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Control strategies against algal fouling in membrane processes applied for microalgae biomass harvesting / Malaguti, M.; Novoa, A. F.; Ricceri, F.; Giagnorio, M.; Vrouwenvelder, J. S.; Tiraferri, A.; Fortunato, L.. - In: JOURNAL OF WATER PROCESS ENGINEERING. - ISSN 2214-7144. - 47:(2022), p. 102787. [10.1016/j.jwpe.2022.102787]

Availability:
This version is available at: 11583/2967476 since: 2022-06-15T02:33:23Z

Publisher:
Elsevier Ltd

Published
DOI:10.1016/j.jwpe.2022.102787

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12 October 2024

Control strategies against algal fouling in membrane processes applied for microalgae biomass harvesting

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Abstract

Microalgae biomass is increasingly applied in a variety of high-end applications, such as biofuel production, CO_2 fixation, food, and cosmetics. As the demand for microalgae increases, improvements in biomass harvesting techniques are required since dewatering represents a significant fraction of the total algae production cost. While membrane technology is growing as a means to achieve effective biomass harvesting, fouling from microalgae suspensions is a major drawback, since these streams are rich in organic compounds, nutrients, and biological materials. The aim of this paper is to present the state-of-the-art of the control strategies to manage algal fouling. The control strategies are divided into: (i) mitigation strategies, including pre-treatment options, modified membrane surfaces, and hydrodynamic approaches; and (ii) adaptation strategies, which include physical, mechanical, and chemical cleaning. Fouling mitigation strategies are implemented in membrane separation processes seeking to maintain high productivity without compromising biomass quality, while minimizing the energy cost related to fouling control. Adaptation techniques include optimization of the cleaning time and effective removal of the irreversible foulants. Further, minimization in the use of chemicals and of the backflush permeate must be achieved to ensure an efficient performance in chemical cleaning and backwash approaches, respectively. Finally, the article discusses future research perspectives in membrane-based microalgae harvesting with a focus on zero liquid discharge and effective fouling control strategies within the water-energy nexus.

Keywords: Microalgae Dewatering; Fouling control; Membrane photobioreactor; Membrane filtration; Microalgae cultivation; Wastewater treatment;

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1. Introduction

Increasing stress in the water-energy system mirrors the challenges caused by rapid industrialization and population growth. In fact, water scarcity hits two-thirds of the global population for at least one month each year, putting a spotlight on the need for alternative water supply strategies [1, 2]. The use of large-scale solutions for desalination and water reuse has gained momentum owing their potential to increase the freshwater available worldwide; in turn, these technologies demand large amounts of energy and consequently, entail high greenhouse gas emissions [3]. Thus, a deeper understanding of the so-called water-energy nexus (WEN) is required to address the trade-offs between both resources [4, 5]. Coupling renewable energies to water production is a promising strategy to facilitate both sustainable development and water access. In this context, the production of algal biomass is a promising field owing a growing interest for commercial applications in biotechnology, wastewater treatment, food, and cosmetics [6, 7]. Further, applications in the energy sector, such as biofuel production and CO_2 fixation, pose great potential [8-11]. In fact, it is now possible to industrially produce more than three times higher oil per hectare using algae when compared to other feedstocks, e.g., corn , while a CO_2 fixation rate of 5 g/CO_2 L^{-1} day^{-1} can be reached in ambient air, which is roughly 10-50 times more efficient as compared to terrestrial plants [12]. In addition, microalgae can also be cultivated utilizing CO_2 from non-conventional sources, such as facilities which produce carbon dioxide as a by-product, thus reducing the overall CO_2 emissions [13].

The most used harvesting solutions for concentrating algae biomass are: (i) coagulation, (ii) dissolved air flotation, (iii) centrifugation, and (iv) membrane filtration. Briefly, (i) Coagulation consists of adding coagulants in the feed solution in order to reduce the electrostatic repulsion between the microalgae cells, thus causing their settling [14]. (ii) Dissolved air flotation (DAF) relies on air micro-bubble generation to promote microalgae flocs rising to the interface where the biomass is accumulated [15]. (iii) Centrifugation is based on a physical method which acts radially to separate colloids such as algae from their liquid medium according to their density difference [16]. Finally, (iv) membrane filtration

83 uses a selective barrier to divide one stream (feed) into two different phases (concentrate and permeate)
84 according to size differences under a pressure gradient applied across the membrane [17]. Among these
85 solutions, membrane-driven separation is advantageous in terms of compactness, high water recovery
86 rates and in maintaining high-quality biomass upon concentration [18, 19]. Microfiltration and
87 ultrafiltration are the typical and the most effective membrane processes used for concentrating
88 microalgae biomass [18, 20, 21].
89 To secure an efficient process performance, membrane fouling phenomena must be reduced: fouling
90 causes a dramatic decrease of permeate water production and it requires stoppage periods for
91 membrane cleaning and eventually replacement [22]. Biofouling and organic fouling are especially
92 significant when the feed stream contains highly concentrated microalgae suspensions. Fouling-related
93 loss in performance is a major challenge to membrane technologies for harvesting algal biomass and for
94 the downstream processes seeking to recover value-added products [23, 24]. Numerous recent
95 publications have addressed these phenomena, describing in detail the fouling mechanisms and effects
96 when membranes are applied to concentrate microalgae [18, 25]. This review focuses on effective
97 mechanisms for fouling control and mitigation during microalgae harvesting in membrane-driven
98 separation technologies.

2. Mechanisms and characteristics of algal fouling

Membrane-driven algae separation is highly prone to extensive fouling [18]. Different factors, intrinsic to process design and algal biomass, determine the fouling severity. In this section, a summary of the main algal foulants and fouling regulating factors is reported. This information is discussed in the light of energy aspects related to loss of productivity and overall performance.

2.1. Main foulants

Microalgae are microorganisms with cell sizes ranging from 3 to 30 μm that exhibit cultivation concentrations lower than 1 kg/m^3 [26]. Microalgae suspensions also include other components, such as vitamins, proteins, pigments, nutrients, and organic substances [27]. Organic and biological foulants are regarded as dominant in microalgae separation processes, while inorganic fouling and scaling are considered negligible, as low amounts of total dissolved solids (TDS) and inorganic colloids are typically present in algae-rich streams, especially if compared to organic and biological components [28]. It is important to note that, in algae membrane-based processes, the distinction between biofouling and organic fouling often is not clear since they occur simultaneously. Therefore, algal fouling results mainly from the deposition of algal cells, algal organic matter (AOM), and transparent exopolymer particles (TEP) present in the feed solution [17, 28-31], together with the deposition and adsorption of organic compounds, such as proteins, carbohydrates, and fatty acids.

The algal cell characteristics are strongly dependent on the species of algae themselves, varying also within different strains [18, 29, 30]. Cells are larger than the pores of MF membranes (typically 0.1-10 μm) and UF membranes (typically <0.1 μm), so they are rejected by the membrane due to size exclusion. In this perspective, has been observed that large particles with spherical shape are responsible of the formation of a cake layer with high porosity values and good filterability properties. [32]. Furthermore, more stable and uniform cake layers in terms of porosity, which also generally guarantee higher filterability, were associated to algae strains characterized by rigid cell walls. The initial

123 biomass concentration is another key aspect when dealing with fouling severity, since highly
124 concentrated feed streams are associated to high viscosity values, in turn responsible for lowering the
125 effects of air scouring. Lastly, several literature reports have shown direct correlations between biomass
126 concentration of the starting stream and flux decline over time [31, 33].
127 Another major foulant is represented by the soluble organic matter, which includes compounds of
128 diverse chemical composition and molecular weight, whose characteristics depend on factors such as
129 algal strain characteristics and nutrients available. AOM that results from the algal metabolic activity is
130 termed external organic matter (EOM), while internal organic matter (IOM) is the AOM fraction
131 released due to cell ageing and lysis [34]. The latter occurs in tandem with the generation of smaller cell
132 fragments and cell debris and is prone to cause pore blocking and narrowing [2]. The algal particle
133 characteristics substantially affect the fouling propensity during membrane-based biomass harvesting,
134 and their size distribution stems from the dynamic equilibrium between formation, transformation, and
135 breakage of algal cells and AOM [35].
136 AOM is mainly characterized by negatively charged organics with high hydrophilicity and low specific
137 ultraviolet absorbance [36]. AOM also includes other compounds, such as proteins, peptides, and
138 amino acids, which are responsible for the heterogeneity of the algal stream [34, 37, 38]. However,
139 AOM shows large variations among different algal species even if, in general, large molecules are
140 usually biopolymers (e.g., proteins), while the small molecules are usually represented by acids and
141 humic substances [28, 39]. High molecular weight (HMW) and low molecular weight (LMW)
142 compounds in AOM act very differently in relation to membrane fouling. The formers are rejected
143 mostly due to size exclusion and, similarly to what happens with algal cells, they tend to accumulate at
144 the membrane surface and contribute to a formation of a cake layer. Contrarily, LMW compounds can
145 more easily enter membrane pores, thus causing pore blocking, which represents the main cause of
146 irreversible fouling [40-42]. It should also be noted that, cells and HMW compounds are instead prone
147 to build a cake layer, which may act as a secondary sieving layer for LMW substances, thus protecting
148 the membrane by reducing the likelihood of pore blocking and irreversible fouling.

149 The last main algal foulant is represented by TEPs, which are the high viscosity part of AOM. TEPs
150 can be of different sizes ranging from 0.05 up to 200 μm [43-45]. Their characteristics in terms of
151 viscosity are responsible for a gel-like structure that increases attachment probability and considerably
152 hamper membrane permeability [18, 29, 46, 47]. Ideally, in an algal stream in the absence of AOM, the
153 cake resistance would increase linearly with the number of deposited algal cells; in reality, TEPs
154 generate a dynamic structure which links algal cells and AOM and significantly reduces membrane
155 performance and productivity [40, 42].

156 2.2. Fouling regulating factors

157 Algal fouling is described as a multi-stages process, whose development is influenced by the
158 interactions between feed stream and the membrane surface under a convective force that triggers the
159 gradual deposition of fouling materials over time [46]. In most cases, algal fouling develops as: (i) a first
160 rapid flux decline in the very first minutes of filtration, which is due to pore narrowing (irreversible and
161 removed only by chemical methods), caused by the attachment of compounds and colloids smaller than
162 the membrane pores, usually extracellular polymeric substances (EPS) released by algae [18, 48, 49].
163 This first effect is enhanced by the simultaneous adsorption of biopolymers on the membrane surface
164 and inside the membrane pores. From here, (ii/a) the cake layer begins to develop during the first
165 transitory filtration phase; its formation is due to particles and solutes larger than membrane pores that
166 are rejected, creating additional resistance to water passage as they accumulate on the membrane
167 surface, thereby referred as a dynamic membrane [17, 50, 51]. This characteristic effect is mostly due to
168 cell-cell and cell-AOM interactions rather than feed-membrane interactions. Further, (ii/b) a gel layer
169 structure, typical in algal harvesting processes, can be present and it is mostly associated to the presence
170 of bacteria and to biopolymers dominance over algae cells when accumulating on the membrane
171 surface. Finally, (iii) the last fouling mechanism stage exhibits flux stabilisation, resulting from the
172 equilibrium reached between cake layer growth and the tangential transport imposed by the cross-flow
173 hydrodynamics in the boundary layer. An illustrated representation of the foulants formation position
174 and on fouling stages is displayed in Fig. 1.

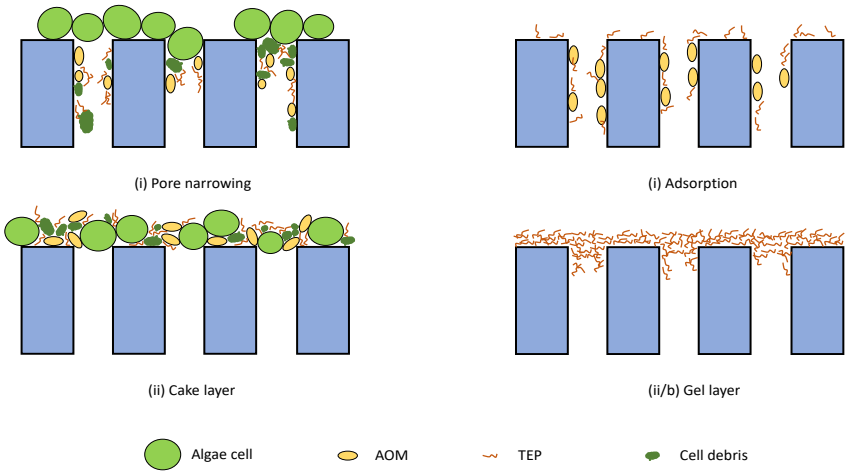


Fig. 1: Mechanisms of membrane fouling in membrane processes for biomass harvesting: (i) pore narrowing usually occurs first, due the action of compounds (algal organic matter (AOM), transparent exopolymer particles (TEP) and cell debris) smaller than membrane pores. Pore narrowing is enhanced by the simultaneous adsorption effect (i) related mainly to biopolymers, i.e., AOM and TEPs. Cake layers are instead associated to larger particles, such as algal cells, which are less likely to penetrate into the membrane pores and more prone to give rise to the so-called dynamic membrane. A gel-layer structure (ii/b) is formed when biopolymers and bacteria are more present than algal cells in the feed stream.

The hydrodynamic conditions, i.e., cross-flow velocity (CFV) and trans-membrane pressure (TMP), by controlling drag forces and shear stress have a fundamental influence on filtration performance and fouling mechanisms [52, 53]. On the other hand, the characteristics of the feed microalgae streams and of the membrane also play a crucial role. The impacts of microalgae biomass on fouling steam from species, cell dimensions, particle size distribution (PSD), hydrophobicity and zeta potential [54]. Moreover, feed streams with microalgae concentrations >0.6 g/L have been shown to result in a much faster cake layer formation while greater membrane porosity usually entails larger permeate fluxes [55]. The ratio between foulant and membrane pore size reportedly influences the extent of pore blockage, and hence fouling irreversibility. Some studies observed larger irreversible fouling rates when using less selective membranes, with larger pore sizes, when compared to less productive ones with smaller pores; this reportedly stems from a wider range of foulants passing through the membrane, henceforth inducing more pore blockage [47, 56]. On the contrary, opposite trends wherein large pore sizes are less prone to irreversible fouling are described as an archetype of a lower hydraulic resistance and decreased drag forces [57, 58]. Clearly, fouling phenomena is a complex mechanism, which depends upon numerous factors. In spite of several investigations on colloidal and biological fouling, there is a lack of

models in the realm of algal membrane fouling [59, 60]. From this point of view, a considerable range of process optimization still potentially exists in algae harvesting by membrane filtration [61].

2.3. Fouling and productivity

In typical membrane operation, the productivity values (water fluxes) vary substantially depending on CFV and TMP. In general, medium and high pore-size membranes are more suitable for filtering algae since their selectivity is the most adequate. Their average mean productivities, once reached steady-state conditions, vary from 10 and 100 LMH [62-64], even if some mitigation strategies can ensure steady state fluxes up to > 200 LMH [65, 66]. Quantitatively, in terms of productivity loss, literature reports estimate that over sub-critical conditions, the fouling phenomena reduce the water fluxes approximatively between 50 to 90% [18, 29, 40, 67]. Specifically, Liu et al. observed that, without pre-treatments, an ultrafiltration module for algae-laden treatment recorded a decrease of water flux of around 85% [68], while Zhao et al. reported that an ultrafiltration process working under sub-critical conditions is subjected to productivity reductions of around 50% [69]. Thereupon, a direct correlation between flux decline due to fouling and increase in energy demand exists; these patterns are aligned with the pilot scale application analysis of Wang et al. [70] which found that fouling influence is the main bottleneck since capital and operating costs accounted for 9% and 91% respectively, and specifically power requirement contribution was ~50%. The estimated energy requirements were similar to those recorded in other membrane processes (0.17-2 kwh/m^3) reviewed by Mo et al. [71].

Fig. 2 qualitatively summarizes and compares the influence of the main parameters on the filtration performance. The microalgae species influences the process productivity because different strains may give rise to AOM, EPS, and TEP of different size and chemistry, which are then responsible of the interactions with the membranes described above. However, the strain shape and subsequent physical interaction with membrane pore size may even be more important than these effects. CFV typically shows a stronger impact on productivity than TMP, especially above the threshold flux, which is defined here as the maximum permeate flux at which no significant fouling occurs. Above this value, an increase in TMP does not translate into a consequent increase in water flux [72], while increasing CFV

may still have a positive effect on productivity. Lastly, the membrane porosity is a critical parameter in relation to the extent of irreversible fouling, which may be argued to be the ultimate effect that should be avoided [56].

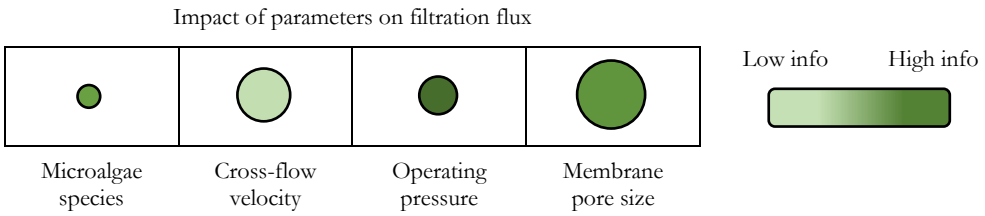


Fig. 2: Assessment of the influence of important parameters on processes productivity. The circle size is qualitatively proportional to the impact on process performance and does not refer to the process performance or fouling behaviour in absolute terms. The intensity of the colour filling each circle is a qualitatively representation of the amount of information available for each parameter.

3. Algal fouling control strategies

The fouling phenomena must be reduced and properly controlled to guarantee high process performance. In this section, a review of the fouling control strategies is reported; Fig. 3 summarizes the approaches, classified into mitigation and adaptation strategies. Briefly, mitigation strategies are aimed to reduce the impact of fouling phenomena by acting directly during the run of the process while adaptation (or cleaning) ones are applied at the end of each cycle in order to re-establish as much as possible the starting conditions.

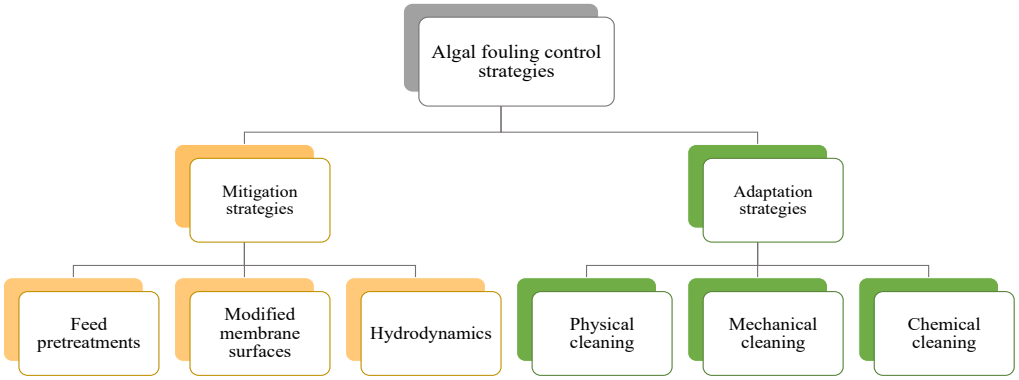


Fig. 3: Schematic of algal fouling control strategies. The objective of mitigation strategies is the reduction of fouling severity during membrane filtration cycles, while adaptation refers to actions carried out at the end of each filtration cycle to restore initial membrane performance.

3.1. Mitigation strategies

There are mainly three categories of mitigation strategies. Feed pretreatment is reported as an effective approach to control the accumulation of compounds known for aggravating reversible and irreversible fouling as well as precursors in the formation of undesired disinfection by-products [73].

They are increasingly studied for algal streams and include coagulation, adsorption, oxidation, air flotation, and their combination [29, 74-77]. Utilization of modified membranes and hydrodynamic approaches are also effective strategies to mitigate and reduce fouling. A detailed summary of the fouling mitigation strategies available in the field of membrane-based separation processes for algae biomass harvesting is presented in the Table 1 and further specific details are described in the next paragraphs.

249 **Table 1:** Fouling mitigation strategies resume

Fouling control strategy	Control mechanism	Main inputs	Control principle	Drawbacks	Findings	Ref.
Feed pretreatment (Section 3.1.1.)	**Coagulation**	Iron coagulants	Lower the surface tension between foulants Promote formation of big aggregates to form a porous cake layer	Large demand of chemicals and alteration of biomass properties.	Fe(IV) coagulant/oxidant have a dual effect that outperforms traditional $FeCl_3$ Fe(VI) ↑ removal LMW & biopolymers than $FeCl_3$	[36, 73, 78]
		Polymeric coagulants			Chitosan ↑removal LMW & biopolymers than $FeCl_3$ ↓concentration required to achieve high efficiency	[36, 79]
	Electrocoagulation	Electricity and iron/aluminum/steel electrodes	Apply dc to destabilize foulants by double layer compression or charge neutralization	High energy and equipment costs. Changing polarity and electrode passivation.	Low DC in electrode ↑ concentration of microalgae (CF>100) ↑ removal of DOC, proteins and carbohydrate	[80, 81]
	Oxidation	Ozone, ultraviolet light (UV), chlorine, Fe(II)/persulfate or cold plasma	Generate hydroxyl and sulfate radicals decompose organic foulants and mineralize the smaller fractions	High equipment and energetical cost. Cell breakage risks with consequent IOM release. In chlorine, generation of hazardous by-products.	0_3 & UV ↑ transform organics to LMW ↑ pore blockage ↑ release of IOM and cell debris ↑ pore blockage	[74, 82, 83]
					Fe(II)/persulfate ↓ cell breakage ↓ dose required	[83]
					UV/ persulfate ↑ specific flux	[84]
	Electro-oxidation	Boron-doped diamond anodes	Generation of reactive free radicals OH• that decompose organic foulants	High equipment and energy costs. Changing polarity and electrode passivation.	Mineralization of AOM responsible of irreversible fouling. ↓ resistance to aggregation → ↑ large flocs → ↑ filterability	[76, 85]
	Adsorption	Powdered activated carbon (PAC), kaolin, metal oxide	Adsorb LMW organics and particles	Low effect in biopolymers, large organics and algal cells	↓ LMW fractions ↓ irreversible fouling ↑ DOC removal ↑ floc stability ↑ cake filterability	[75, 86, 87]
	Pre-coagulation + pre-oxidation	a) $KMnO_4$ - Fe(II) b) Fe(III) - permonosulfate c) Sodium percarbonate - Fe^{2+}	Create a dual coagulation/oxidation effect	High demand of chemicals	$KMnO_4$ + Fe(II) → ↑ floc size → ↑ cake porosity ↓ pore blockage and irreversible fouling SPC + Fe^{2+} → ↑ formation of $Fe(OH)_3$ → ↑ floc size	[76, 84, 88]

Modified membrane surfaces (Section 3.1.2.)	**Nanomaterial blends**	TiO_2, Fe_2O_3, zeolites, silica, silver nanoparticles	Coat of blend membranes with particles capable of modifying hydrophilicity or surface charge	Demand of expensive membrane materials	TiO_2 / Fe_2O_3 → ↑ hydrophilicity → ↑ filterability Hydrophilic polymers → ↑ hydrophilicity → ↑ filterability Ag-polymers → ↑ surface roughness and permeability	[89-91]
	Nanomaterial blends + oxidation	Zero valent iron (ZVI) + peroxymonosulfate (PMS)	Activate PMS via ZVI, generating SO_4 and Fe^{3+} and a dual coagulation/oxidation	Cost of materials and chemicals	*In-situ* oxidation created on membrane-fouling interface significantly decreases the oxidative cell damage effect created in the feed by other oxidation methods, protecting algal cells from breaking Dual coagulative/oxidative effect → ↑ filterability	[65, 92]
	Modified membrane polymers	Equipment and membranes	Modify the membrane structure and pore geometry	Demand of equipment and materials	Solvent vapor treatment ↑ mechanical strength of membranes slight permeability ↓	[66]
	Customized tilted membrane panels	Energy, air and equipment	Maximize the membrane-bubbles interaction, maximizing shear	Equipment and energy costs Cell damage and release of IOM	Increasing the tilting angle to 20° improves water flux by 20-30%	[66, 93]
Hydrodynamics (Section 3.1.3.)	**Modifications in HRT, SRT in MPBRs**	-	Control growth stage and the generation/release of AOM	Alteration of biomass properties Decrease in productivity	↑ HRT → ↓ organic loading rate and fouling severity ↑ SRT → ↑ LMWO and irreversible resistance	[35, 41, 54, 94]
	Flux control	Pumping energy	Decrease convective forces that drag foulants towards the membrane	Need of large membrane areas to keep low fluxes	Subcritical flux → ↓ irreversible fouling ↑ filtration stability	[18, 29, 40]
	Cross-flow velocity	Pumping energy	Increase CFV to enhance turbulence and shear	High energy demand Cell damage and release of IOM at high CFV	↑CF → ↑ turbulence values which reduce the deposition of foulants	[52, 95]
	Membrane vibration	Energy and equipment to vibrate/rotate	Mechanically create shear in the surface-feed interface to disrupt cake and gel polarization layers	High equipment and energy costs. Cell damage and release of IOM	*In-situ* shear created on membrane-fouling interface decreases the shear stress created to the feed, protecting algal cells from breaking ↑ removal of cake and gel polarization layers ↑ collision → ↑ large algal aggregates with better filterability	[63, 96, 97]

3.1.1. Pretreatment of the algal stream

3.1.1.1. Coagulation and electrocoagulation

The use of iron-based coagulants with amphiphilic characteristics is commonly applied to lower the surface tension between liquids and solids and this technique may be applied to promote the formation of cell and AOM aggregates that form a porous cake layer with better filterability which can easily be removed by membrane cleaning. Iron in the ferric chloride ($FeCl_3$) form is widely used in water treatment for coagulation and flocculation processes [78]; however, the high doses required limits its application [36, 73]. Thereby, recent developments proved a better suitability for the use of alternative iron-based coagulants, such as Fe(IV)/Fe(II). Iron at high oxidation state, i.e., Fe(IV) (ferrate), is considered promising, as this acts simultaneously as coagulant, oxidant, and disinfectant. Alshahri et al. [78] reported an improvement in the AOM removal from 58-87% to 88-93% when comparing the performance of $FeCl_3$ with *in-situ* generated liquid ferrate (FeO_4^{-2}) in the coagulation of *Chaetoceros affinis*. A higher adsorptive and oxidative capacity was achieved through the formation of $Fe(OH)_3$ ions that resulted from the decomposition of ferrate and thus, dosing Fe(VI) led to a higher removal of LMW neutral organics and other biopolymers (38–65% and 97–100%, respectively) in comparison with Fe (III) (14–29% and 74%, respectively) [78]. In a following study, Alshahri et al. [73] found a similar trend when coupling liquid Fe(IV) and Fe(III) with dissolved air flotation (DAF); at a concentration of 3 mg Fe/L, Fe(IV) and Fe(III) enhanced the removal of AOM by 99% and 52% respectively, at a flotation time of 10 minutes.

Alternative inorganic coagulants have been found to form stable structures with negatively charged proteins. Such effect was reported by Huang et al. [87] when dosing a metallic chelator in the form of Cu^{+2}: increasing the Cu^{+2} concentration lessened the contribution of IOM to the overall fouling resistance. Similarly, the use of polymeric organic coagulants, e.g., chitosan, is gaining popularity due to a low production cost (>2 USD/kg) and high efficiency at algal flocculation at lower doses [36, 79]. Discart et al. [36] reported a 50% flux improvement by dosing 10 mg/L of $FeCl_3$, whilst the same improvement was achieved at a concentration of 2-5 mg/L of chitosan. Likewise, Du et al. [79] proved the efficiency of chitosan to remove LMW organics and alleviate pore blockage; in a comparison of the efficiency of aluminum sulfate and a

276 composite aluminum sulfate-chitosan (AS-CS) coagulant, the presence of CS polymeric chains in the AS-CS
277 composite led to a larger removal of LMW organics, thereby alleviating membrane fouling by 58.8%,
278 compared to AS alone (23.7%).

279 Coagulation is widely regarded as a successful fouling control treatment. However, despite enhancing the cake
280 layer compressibility, severe membrane fouling due to pore-blocking mechanism has been reported under
281 certain conditions when coagulants combine with LMW organics. Thereby, recent developments explored
282 ways to eliminate undesired chlorides or sulfates from conventional coagulation salts by using
283 electrocoagulation [84]. This technique consists in the application of a direct current that destabilizes
284 negatively charged foulants by double layer compression or charge neutralization. As a result, soluble AOM
285 and algal cells coagulate, and the hydrogen bubbles resulting as a by-product trigger their removal by flotation
286 [80, 81, 98]. Parmentier et al. [81] achieved a >100 algal concentration factor when applying low direct
287 currents to iron and aluminum electrodes (0.8 and 0.3 A, respectively), concentrating suspensions of *Chlorella*
288 *vulgaris* from 0.2 to 18.5 and 35.2 g/L when using iron and aluminum electrodes, respectively. Similarly, Rafiee
289 et al. [80] reported the removal of proteins, carbohydrates and DOC by 21, 60 and 47%, respectively, when
290 applying electrocoagulation to a mixed algal culture.

291 Overall, some energetic and economic evaluations were performed to evaluate the feasibility of the
292 coagulation pretreatment, albeit not directly for algal streams separation. Yoo et al. [65] observed that in a
293 water treatment plant the costs of chemicals rose from 0.0066 to 0.0146 $/m^3 due to the high doses of
294 needed coagulants, but the energy saving due to better treatment performance (permeate flux increased from
295 50 to 70 LMH) and the lower sludge production counteracted this effect, thus decreasing the total operating
296 cost of 11.2%. Further studies are necessary to obtain an adequate assessment of the feasibility of coagulation
297 in algae separation. In this context, new plant-based coagulants are gaining popularity as environmentally
298 friendly solutions owing to their safety and biodegradability, being more sustainable compared to
299 conventional chemical coagulants (i.e., Fe and Al salts, or polymers).

300 **3.1.1.2. Oxidation and electro-oxidation**

301 Oxidation is gaining spotlight in the mitigation of membrane fouling and in the degradation of AOM [83].
302 The pre-oxidation of feed water by means of, e.g., ozone, ultraviolet light (UV), chlorine, Fe(II)/persulfate,
303 and cold plasma, has been reported as an effective fouling mitigation technique. The generated hydroxyl and
304 sulfate radicals decompose organic foulants and mineralize the smaller fractions [74, 83, 99]. Nevertheless,
305 large doses of an oxidant pose a risk for cell breakage and IOM release into the medium. Liu et al. [83]
306 compared the oxidation effect of ozonation and oxidation/coagulation by Fe(II)/persulfate. Both oxidant
307 strategies were found to trigger cell breakage and thus, to increase the concentration of IOM in solution.
308 Specifically, cell breakage of 58 and 81% was observed when dosing ozone at 0.015 and 0.06 mM,
309 respectively. In contrast, Fe(II)/persulfate reduced cell breakage (5%) at the lowest concentration (0.05 mM)
310 of reagent investigated. Thereupon, ozonation was correlated with an increase in the irreversible fouling
311 fraction, as it was found to degrade biopolymers into smaller foulants, prone to cause pore blockage. In
312 contrast, dosing Fe(II)/persulfate at low concentrations (0.2 and 0.4 mM) improved the final specific flux.
313 Moreover, ferric ions were *in-situ* generated during the oxidation with Fe(II)/persulfate, thereby promoting an
314 additional coagulant effect that reduced fouling and favored the formation of a porous cake layer, by
315 enhancing the agglomeration of cells and AOM into large flocs. Comparatively, Lee at al. [84] showed a
316 significant decrease in the fouling membrane index and TOC concentration in solution (85% and 17%,
317 respectively) when using UV/permonosulfate (PMS) oxidation in suspensions of *Pseudo Nitzchia*.

318

319 UV-driven advanced oxidation is advantageous owing to its great oxidative power. Coupled oxidation
320 treatments have shown large performance in fouling control. Wan et al. [74] compared the fouling control
321 efficiency of several composite UV-based oxidation processes (UV/persulfate (UV/PS), UV/chlorine, and
322 UV/H_2O_2). Beyond a critical threshold of oxidative stress, UV-radiation was found to trigger cell lyses and
323 therefore increase irreversible fouling caused by the IOM and cell debris released by dead cells. Upon
324 experiments, the composite UV/PS was found to be more effective to control the membrane fouling,
325 enhancing the final specific flux from 0.26 to 0.29 and 0.81 at PS doses of 0.25 and 1 mM, respectively. Better

326 efficiency at controlling reversible fouling was associated with the coagulant effect of PF that decreased the
327 concentration of irreversible foulants such as soluble hydrophobic LMW organics and debris. Analogously, a
328 positive effect of UV/H_2O_2 in fouling control was exhibited at large doses (1 mM), reaching a final specific
329 flux of 0.38. In contrast, UV/chlorine pretreatment exacerbated membrane fouling, and the resistance
330 associated with irreversible fouling increased fivefold compared to raw water. The effect was due to the
331 breakage of large biopolymers into LMW that progressively accumulated in membrane pores, indicating a low
332 efficiency in biopolymer mineralization.
333 The use of UV oxidation allows reduction in the use and release of chemical oxidants into the water;
334 nevertheless, the resulting cell damage limits its application. Alternatively, the use of cold plasma (CP) was
335 proposed by Lee et al. [99] as a strategy to mitigate membrane fouling by active decomposition of AOM, as it
336 displayed suitable removal of TSS, TOC, and COD. However, research on CP is needed to clarify its energy
337 expenditure and fouling control.
338 Likewise, Liu et al. [85] proved as positive the effect of electrochemical pre-oxidation with boron-doped
339 diamond anodes at reducing reversible and irreversible fouling; with an increase in the oxidation time, the zeta
340 potential of algal biomass decreased from −18,2 to −8,9 mV, weakening the resistance to aggregation and
341 thereby, leading to the formation of particles with sizes larger than 100 μm. These particles promoted the
342 formation of a loose and fuzzy cake layer with a lower resistance to water permeation. Nevertheless, the high
343 energy and equipment cost, along with the frequent changing polarity and electrode passivation, pose a
344 bottleneck for the process, and thereby its efficiency on fouling control necessitates further research [76]. In
345 general, some environmental issues must be considered when dealing with oxidation processes: some toxic
346 by-products can be formed upon degradation of organic compounds as well as disinfection by-products
347 associated to the use of chlorine [100].

348 **3.1.1.3. Adsorption**

349 The use of adsorption has been proved to remove the LMW fractions and micropollutants responsible of
350 irreversible membrane fouling, as well as disinfection by-products [75, 86, 87]. The interactions between
351 adsorbents and algal foulants have been found to correlate with the cake layer stratification and deformation

352 properties [86]. Commonly used adsorbents include powdered activated carbon (PAC), kaolin, and metal
353 oxide particles [75, 86, 87]. Zhang et al. [86] reported a stable efficiency in EOM adsorption and fouling
354 mitigation when using pre-mixed PAC and kaolin particles. The fouling control efficiency increased for
355 higher PAC sizes, as this parameter increased the cake layer porosity and thereby, enhanced the transport of
356 the algal foulants back into the feed stream, lowering the overall fouling rates. On the other hand, smaller
357 PAC/kaolin sizes led to micropores with a larger adsorptive capacity that was mirrored in higher DOC
358 removal efficiencies and cake layer resistance. Thereby, when using PAC_{12}, PAC_{25}, and PAC_{30}, the DOC
359 removal efficiencies reached 22.3%, 23%, and 15.9% respectively. Nevertheless, while large biopolymers in
360 the EOM where difficult to remove by PAC adsorption, kaolin proved to be more efficient owing the
361 hydrogen bonding between O atoms in the kaolin and OH groups of the EOM. The polyhedral face-edge
362 structure of kaolin favored foulant-kaolin aggregates with low adhesion energies, thus building loose
363 aggregates and a porous cake layer.
364 Furthermore, the sole use of PAC has been proven insufficient at removing high MW organics, and UV alone
365 hardly degrades the AOM due to the limited and short lifetime of $OH^{\bullet}$ radicals in solution. Accordingly, Xing
366 et al. [75] employed UV/Cl oxidation and PAC adsorption (UV/Cl/PAC) to offset the negative impacts of
367 DBPs prior to filtration. UV/Cl pretreatment led to the formation of $OH^{\bullet}$ and $Cl^{-\bullet}$ radicals that triggered the
368 decomposition of HMW organics into activated LMW that could be easily adsorbed by PAC. The
369 combination of UV/PAC and UV/Cl/PAC enhanced the DOC removal to 7.2 and 9.3%, and this was
370 mirrored in the TMP development along filtration operation. Raw feed water was associated to a final TMP
371 of 0.81 bar, whereas UV/PAC and UV/Cl/PAC pre-treatment resulted in TMPs of 0.67 and 0.56 bar,
372 respectively. The alleviating effect of UV/Cl/PAC was ascribed to the combined effects of oxidation and
373 adsorption. The composite use of UV/PAC and UV/Cl/PAC proved to be efficient at degrading organic
374 foulants and alleviating reversible and irreversible fouling, reducing them by 26-36% and 13-22%, and by 44-
375 48 and 33-53%, respectively. In general, adsorbent materials do not present any relevant environmental
376 issues. However, the steps involved with their management, regeneration, or disposal depend strongly on the
377 types of adsorbent and algal process.

3.1.1.4. Blend pre-treatments

Pre-treatments are important to increase the efficiency and service time of the membrane operation, and improve water quality. On the other hand, the implementation of pre-treatment steps involves an increment of investment and maintenance costs, which might not always justify the overall benefit. Moreover, when the target is biomass harvesting and recovery, pre-treatments should not compromise the quality of the final product. To this extent, the application of strong oxidative products, such as hypochlorite, may be unfeasible. Compact and simple system designs combining the use of adsorbents, oxidants, and coagulants were recently reported as highly efficient approach toward fouling control. For example, Ma et al. [76] proposed the *in-situ* generation of Fe(III) by a composite pre-coagulation/pre-oxidation method, using $KMnO_4$- Fe(II) in the filtration of *Microcystis aeruginosa*. Larger floc sizes, generated when using $KMnO_4$-Fe(II), led to a porous cake layer that, translating into a lower TMP increase in comparison with the Fe(III) coagulation alone (0.25 bar vs 0.43 bar, respectively). Furthermore, pore blockage was alleviated during the $KMnO_4$-Fe(II) pretreatment, as the cake layer contribution increased from 31.6% to 54.6%. Similar results were reported by Lee at al. [84] when combining Fe(III) and permonosulfate to alleviate membrane fouling. Further, Ren et al. [88] coupled pre-oxidation and coagulation by using sodium percarbonate activated with ferrous ion (Fe^{2+}/SPC). The SPC was catalyzed by Fe^{2+} to generate OH• oxidants, whilst the Fe^{2+} led the formation of $Fe(OH)_3$ as it was oxidized. The combined effect of Fe^{2+} and SPC enhanced the specific final flux (J_f/J_0) from 0.284 to 0.710, and reduced reversible fouling resistance by 80% at doses of 0.25 mmol/L.

3.1.2. Modified membrane surfaces

Increased understanding of the combined effect of membrane-foulant interaction forces, including electrostatic repulsion/attraction, hydrophobic forces, and Van der Waals forces, has brought momentum to research in modified membrane surfaces as a strategy to manipulate these colloidal forces on fouling development [18]. Thereby, growing attention is being devoted to modifying membrane antifouling properties by, e.g., blends and surface coatings with polymers and inorganic nanoparticles [91]. Compared to traditional pretreatments, membrane modification allows an easier management of the stream and is a potential strategy to reduce the number of pre-treatment steps. On the other hand, membrane modifications

404 still involve high investment costs and challenges related to the scaling-up. The development of super
405 hydrophilic cost-efficient membranes is required to improve the filtration performance while leading to
406 considerable cost reduction within the harvesting process.

407 Membrane materials, such as PVDF, PES, PVC, and polysulfone (PSF), are highly prone to algal fouling
408 because of their hydrophobic nature [101, 102]. For this reason, surface coatings with, e.g., hydrophilic
409 monomers, polymer bushes, and nanomaterials have been applied to increase the hydrophilicity and to
410 improve water filtration performance [29, 89, 91]. For example, TiO_2, Fe_2O_3, zeolites, silica, and silver
411 particles have been shown to increase the water flux owing to their hydrophilic nature [89]. Hu et al.
412 evaluated the antifouling properties of nano-TiO_2 particles incorporated into PVDF membranes, as TiO_2–
413 modified surfaces reportedly became superhydrophilic. The resistance of PVDF/TiO_2 membranes was 49%
414 lower than that observed with pristine PVDF membranes, and the effect was associated with an increase in
415 membrane wettability, displayed by a decrease of the contact angle of water on the membrane (from 54.4 to
416 46°) [89]. Similar results were reported by Yogarathinam et al., as the addition of TiO_2 resulted in a better
417 water recovery percentage and a more stable flux [90]. Additionally, Yogarathinam et al. incorporated a
418 hydrophilic additive (polyethylene glycol), polyelectrolyte additive (polyethylenimine), inorganic additive (zinc
419 chloride) and charged polymer (sulfonated polyether ether ketone) as pore formers in PES membranes. As a
420 result, the water contact angle decreased from 76 to 55° and the water flux was enhanced [90]. Analogously,
421 Liu et al. evaluated the performance of Fe_2O_3 incorporation into PVC membranes during their formation by
422 phase inversion. Incorporating 1% of Fe_2O_3 into the membrane increased its hydrophilicity, lowering the
423 water contact angle by 22%, and thereafter achieving a 66% flux increase by decreasing the deposition of
424 irreversible fouling precursors, such as aromatic proteins, fulvic acids, and humic substances [91]. Similarly,
425 Soleimani et al. evaluated graphene/polydimethylsiloxane (PDMS) with silver nanoparticle coatings (GOH-
426 Ag) in PDMS membranes. The GOH-Ag membranes exhibited better antifouling efficiency by increasing the
427 surface roughness and lowering the water contact angle [103].

428 Iron nanoparticles were studied by Huang et al., by incorporating zerovalent iron (ZVI) on polycarbonate
429 (PC) membranes in combination with peroxymonosulfate (PMS) oxidation. ZVI activated the PMS oxidative

430 effect, generating $SO_4^{\bullet}$ and Fe^{3+} that entailed a dual coagulative/oxidant antifouling effect during the
431 filtration of *C. vulgaris* suspensions. The oxidation zone located solely on the membrane surface and therefore,
432 minimized algal cell breakage as well as the formation of disinfection by-products. As a result, the ZVI-PMS
433 approach led to a larger flux (345 LMH) in comparison with that of the pristine PC membranes (129 LMH)
434 [65]. In contrast, the use of powdered activated carbon coatings was proved to be inefficient at fouling
435 control. A recent approach discussed by Zhang et al. evaluated the pre-deposition of a powdered activated
436 carbon (PAC) coatings onto the membrane surface: the resulting cake layer exhibited stratified layers with a
437 higher resistance to water permeation that aggravated the cell deformation rate and decreased the back-
438 transport of foulants to the bulk solution [92].
439 Differing from surface coatings and material blends, solvent vapor treatment (SVT) modifies the membrane
440 structure by altering the links between fibers. Mat et al. enhanced the mechanical strength of 6,6 electrospun
441 nanofiber membranes (NFM) by using SVT. As a result, the mechanical strength of NFM was increased.
442 Pristine NFM proved to be efficient for algal filtration, although its low mechanical strength limited a
443 prolonged use. In contrast, SVT enhanced the fusion and melting of overlapping fibers, increasing the
444 mechanical resistance by 221% at the expense of a slight decrease in permeability; the thickness, porosity and
445 mean pore size of the treated NFM were lowered by 18, 4, and 40%, respectively, in comparison with the
446 pristine NFM [66]. Moreover, Mat et al. improved the fouling cleaning efficiency by tilting the nanofiber
447 membranes panel to 20°. Increasing the aeration rates further enhanced the productivity, and intermittent
448 aeration was found to produce a higher flux of around 7% when compared to continuous aeration [66].
449 Analogously, Eliseus et al. developed a customized tilted membrane panel to enhance the air scouring
450 efficiency, improving the water flux up to 30% during the filtration of *Euglena sp* [93].
451 Electrically-enhanced membrane systems have gained momentum owing to their capacity to exploit the
452 electrostatic repulsion between the foulants and the membrane surface: electricity applied across the
453 membrane surfaces produces strong electrostatic repulsion forces that reduce the probability of foulants
454 attachment onto the surfaces. Increasing the magnitude of the electric field on conductive membranes

455 augments the electrostatic repulsion and lowers the fouling propensity [104]. So far, electrically-enhanced
456 membranes lack research specifically on algal filtration, thereby demanding further study.

457 **3.1.3. Hydrodynamics**

458 Lower permeate drag force, smooth fluxes, and high surface shear are effective strategies to mitigate fouling
459 [95]. A decrease of permeate flux leads to a reduction in the fouling rate but also requires larger active areas in
460 order to increase the overall productivity [31]; this solution is seldom adopted in real plants due to capital
461 costs and footprint.

462 The relationship between TMP and fluxes is not so straightforward. For example, Sun et al. [23] found that a
463 TMP increase in the range from 1.3 to 1.8 bar brought an increase in permeate flux and productivity, while
464 TMPs above 1.8 bar led to a decrease of productivity. This result is rationalized with the formation of a
465 thicker foulant layer with reduced porosity generated by high compression of microalgae cells and low
466 turbulence values associated to higher TMPs. Adopting optimal CFV values allows direct control over
467 turbulence and shear rates, which are closely connected to foulant accumulation on the membrane surface
468 [52, 95]. Typically, higher CFV values are associated with larger permeate fluxes.

469 In membrane photobioreactor (MPBRs) configurations, i.e., membrane immersed directly into the algal
470 growth reactor, there are also other parameters that regulate the performance, including the hydraulic
471 retention time (HRT) and the solid retention time (SRT). A take-home message from the main literature
472 reported is that an increase in HRT can reduce the organic loading rate and the amount of the nutrients
473 during microalgal cultivation, thus causing a decrease of fouling and cake layer development [35, 41, 54].

474 Shear rates are commonly applied in membrane systems to promote a turbulent flow regime that facilitates
475 the back-transport of foulants to the feed solution [50, 66]. They might be provided from the feed via
476 crossflow velocity, from the scouring systems via air bubbles, or by mechanical means such as vibration or
477 rotation of the membrane modules [105]. These strategies are based on the so-called dynamic filtration
478 systems (DMF): a relative movement between fouling interface and membrane surface in the range of 0 to 0.5
479 mm is created in order to protect the algal cells from damage due to shear. [18, 96]. Furthermore, the shear
480 force applied on the membrane surface disrupts both the cake and the concentration polarization layers,

481 although careful consideration should be made to maintain the shear forces below the threshold of cell
482 breakage, which varies among algal species [18, 105, 106]. This effect was reported by Ladner et al. [106] in a
483 study whereby an exponential flux decline resulting from the cell lysis and the release of organelles and
484 organic matter was observed under shear force conditions.

485 The use of vibration membranes is a recent development applied for fouling mitigation. It relies on high
486 vibration frequencies or amplitudes to create a share rate at the membrane-feed interface [96, 97]. Jiang et al.
487 [96] found an increase in the antifouling performance when the vibration frequency was increased from 1 to 5
488 Hz. The balance in the total interfacial forces was shifted to increase the lifting forces with respect to the drag
489 forces, thereby alleviating membrane fouling. Furthermore, mild vibrations enhanced the inter-foulant
490 collision, thus promoting the formation of large algal aggregates with a lower tendency to deposit on the
491 membrane, although no impact was observed on the relieving of reversible fouling by EOM. Similar results
492 were reported by Zhao et al. [97] within the same frequency range in a uniform shearing vibration membrane
493 (USVM) with constant shear rate by uniform circular motion. By increasing from 1 to 5 Hz, the water flux
494 rose from 296 to 1527 LMH and the final specific flux rose from 0.13 to 0.69, respectively. Furthermore, as
495 the tendency to cell deposition decreased, the biomass was reduced from 0.64 to 0.08 g/m^2 at 1 and 5 Hz.

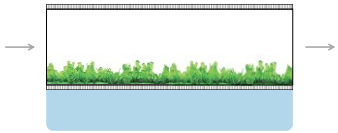
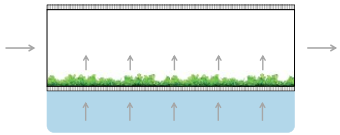
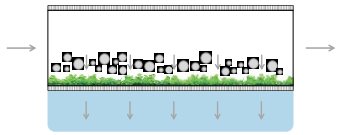
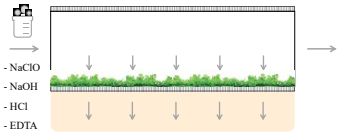
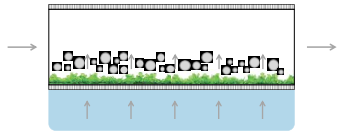
496 Recent developments created a composited fouling control system made of vibration, negative charge, and
497 patterned polysulfone/sulfonated polysulfone (PSf/sPSf) membrane blends [63]. The waves-patterned
498 membranes exhibited larger pore size, porosity and hydrophilicity when increasing the sPSf concentration,
499 thereby displaying higher water permeability. The patterns promoted turbulence, generating a shear effect that
500 was also triggered by the turbulent regime, induced by vibrations of frequencies above 7 Hz. Patterns and
501 vibrations promoted the generation of turbulence eddies between patterns, thereby scouring the foulants with
502 a high efficiency. Furthermore, turbulence and the negatively charged membranes induced a joint effect that
503 minimized irreversible fouling. Altogether, the energy consumption was significantly lower than that of
504 traditional pumping systems that induce shear through a cross-flow velocity, and the synergistic fouling
505 removal relied mostly on the eddy mixing, shear and scouring effects, according to a computer fluid dynamic

506 modeling. The composited fouling control displayed a higher clean water permeance and critical flux, as well
507 as a lower membrane intrinsic resistance.
508 Fortunato et al. [50] reported the impacts of cleaning on the cake layer morphology using *in-situ* non-
509 destructive real-time monitoring for the filtration of *Chlorella vulgaris*. Despite significantly reducing the
510 biomass deposition on the membrane surface (5-10 μm thickness) and the fouling rates (−60%), air scouring
511 was found to be inefficient against irreversible fouling caused by AOM and small colloids. In contrast, the
512 shear stress created by air scouring was found to actually increase irreversible fouling by breaking algal
513 foulants into smaller particles, as also elsewhere reported [105, 107]. Similarly, scouring does not prevent
514 small proteins and carbohydrates from depositing on the membrane surface, and higher shear stress
515 conditions increase the formation of submicron particles that transport through the cake layer into the
516 membrane [40]. Thereafter, as the filtration time increases, scouring leads to the formation of a thin packed
517 layer that cannot be removed by physical cleanings [50, 107]. According to Alipourzadeh et al. [29], there is an
518 optimal air sparging intensity which guarantee the minimum fouling, as stable hydrodynamic conditions can
519 enhance the membrane filtration capacity, whereas the extreme opposite might strongly exacerbate the
520 fouling rate.

521 3.2. Adaptation strategies

522 As fouling occurs, control mechanisms can be applied to maintain or recover membrane performance; they
523 are generally classified as physical, mechanical, and chemical strategies. Their efficiencies rely on both
524 environmental scenario and operating conditions. When productivity or permeate quality reach a threshold
525 low value, cleaning procedures are necessary to recover the performance. Unfortunately, cleaning becomes
526 less effective at each new cycle, reduces membranes lifetime, and requires pausing the harvesting process [97].
527 Therefore, selecting the proper membrane cleaning strategy is of capital importance in terms of energy
528 consumption and process performance [97]. Table 2 reports the main working principles of cleaning
529 strategies, their effect on fouling layers and the main challenges.

530 **Table 2.** Fouling adaptation strategies resume

Fouling adaptation strategy	Cleaning mechanism	Schematic Conceptualization	Control principle	Drawbacks	Challenges	Ref.
Physical cleaning (Section 3.2.1.)	**Relaxation**		Stop the flux and convective drag forces towards membrane	– Decrease in filtration time. – Lower efficiency over time.	↑ back-transport ↓ reversible fouling ↑ decompressing → ↑ cake porosity ↓ gel polarization layer → ↑ filterability	[40, 50, 66]
	Backwash		Reverse the permeate flow to fluidize cake layer and remove the deposited particles from membrane surface	– Loss of permeate. – Extra pumping energy input. – Lower efficiency over time.	↓ compaction of cake layer ↑ cake porosity and filterability ↓ pore blockage, irreversible fouling (decreased efficiency over time)	[33, 40, 50, 82, 108]
	Air scouring		Air bubbles scrap-off the foulants from the top of the cake layer. Induce shear stress to facilitates back-transport of particles	– High equipment and energy costs. – Cell damage and release of IOM.	↑ IOM and cell debris → ↑ irreversible fouling ↓ cake thickness → ↓ reversible fouling ↓ efficiency in removing LMW organics ↑ air flow rates → ↑ bubble size and fouling removal	[93]
Chemical and combined cleaning (Sections 3.2.2. and 3.2.3.)	**Membrane soaking**		Soaking membranes in a cleaning solution with a consequent flush aimed to remove both used chemicals and detached foulants	– Membrane deterioration. – Formation of by-products. – Chemical and energy consume.	Choosing the adequate cleaning solution for each situation. Problems to achieve high recovery efficiencies, especially when numerous cleaning cycle are performed on the same membrane.	[48, 109, 110]
Mechanical cleaning (Section 3.2.4.)	**Backwash + air-water flushing**		Backwash disturbs foulants deposited on the membrane surface and the following forward flushing remove the debris	– High equipment and energy costs. – Wasn't proved to be more efficient than backwash.	Flux recovery of 29%- 66% achieved by forward air–water flushing. Flux recovery of 90% - 100% was achieved by backwash.	[111]

531 **3.2.1. Physical cleaning**

532 The main physical strategies aimed to control the fouling phenomena in membrane filtration are:
533 relaxation, backwash, and air scouring. Briefly, relaxation consists of a temporary interruption of the
534 filtration, facilitating the back transport of the foulants from the cake layer toward the bulk of the feed
535 solution [50, 66]. When the relaxation period ends, the foulants are redistributed more uniformly along
536 the cake layer, which typically becomes more porous [50]. On the other hand, long relaxation periods
537 inevitably reduce the filtration time, thus lowering the overall productivity [50, 66].

538 Backwashing instead consists of reversing the direction of the flux across the membrane using the
539 permeate itself as cleaning solution to exert a physical force aimed at removing the particles attached on
540 the membrane surface, reducing pore blocking and cake layer effects [33, 82, 108]. The bottleneck of
541 this approach steams from a large permeate loss, especially when dealing with LMW compounds that
542 demand large permeate backwash flow rates [50]. Lastly, air scouring consists of gas bubbles injected
543 across the membrane surface that generate localized shear stress zones aimed to detach foulants, and
544 also creating a turbulent regime that enhance the compounds back-transport into the feed.
545 Furthermore, cleaning via air bubbles is largely affected by the bubble sizes; achieving optimal slug-flow
546 conditions demands high energy to pump large volumetric air flow rates [93]. The energy-intensive
547 nature of the process hampers its operational efficiency, and flows above an optimal bubble number
548 might hinder the contact between the liquid feed and the membrane surface, thus reducing the liquid
549 flow across the membrane [50, 66].

550 Cake layer is largely considered to be the main contributor to fouling resistance in algal filtration
551 systems and for this reason much attention has been drawn to its responses to membrane physical
552 cleaning [40, 50, 54]. Fortunato et al. [50] found that relaxation and backwash can both alleviate the
553 final fouling rates by nearly 50%. Nevertheless, differences were found on their impact in the
554 distribution of foulants across the cake layer and thereby, in the mechanisms affecting the flux; during
555 backwash, the cake layer was partially detached from the membrane, but quick redeposition occurred
556 during the following filtration stage with the formation of a similar cake layer. Moreover, the protection

557 exerted by the cake layer against LMW organics deposition was not available upon its detachment. In
558 fact, Kanchanatip et al. [40] reported a decrease in the fouling control efficiency as the number of
559 backwash cycles increased the share of irreversible fouling. Comparatively, relaxation resulted in a
560 thicker and rougher cake layer with similar filterability properties to that of backwash, cutting off the
561 expenses of permeate and energy required for the backflow. It is important to highlight that, prolonging
562 relaxation periods reduce the productivity and has been found to also impact the process negatively by
563 resistance related to irreversible fouling [66, 93]. Mat et al. [66] found that, in a filtration cycle of 5
564 minutes, relaxation periods of 1-2 minutes resulted in the highest steady-state flux. Accordingly,
565 carefully adjusting the filtration/relaxation intervals is required to mitigate fouling without severely
566 reducing the permeability [66]. It is also important to highlight that, cake layer thickness has been found
567 inadequate at predicting fouling severity: the distribution of foulants across the layer, surface roughness,
568 and cake porosity determine together the degree of compression and resistance to water filtration [40,
569 111].

570 **3.2.2. Chemical cleaning**

571 As physical cleaning can counteract only reversible membrane fouling, chemical cleaning has been
572 applied to remove irreversible fouling. This approach consists of soaking membranes in a cleaning
573 solution with a consequent flush aimed to remove both used chemicals and detached foulants [112,
574 113]. It is clear that the optimal cleaning approach depends on a multitude of factors, such as feed
575 composition, fouling nature and severity, cleaning frequency, temperature, pH [114]. Chemical cleaning
576 could be realized in different scenarios: (i) in-situ (without removing the membrane from its vessel), (ii)
577 ex-situ (outside the vessel in an ad hoc tank), (iii) by adding chemicals during the filtration cycle, (iv) by
578 chemical backwash, i.e., adding reagents during the physical cleaning phase [115]. The cleaning agents
579 are classified in: alkaline; acidic; metal chelators; enzymes; oxidizing agents. A combination of these is
580 typically applied.

581 Liang et al. [48] observed a suitable cleaning efficiency during the combined use of NaOH (0.02 N) and
582 NaClO (100 mg/L) when dealing with UF membranes fouled by algae reservoir water. Moreover,

583 mirroring other literature results, they discovered that cleaning by NaOH for 2–6 h ensured lower
584 recovery than that obtained using NaClO. The citric acid instead was not effective in cleaning the
585 membrane, providing only 40–60% flux recovery. The advantages of using NaClO are that it is a strong
586 oxidant, which is able to attack NOM and to inactivate algae cells and bacteria [48, 116]. On the other
587 hand, Ding et al. [109] showed that proteins, amino acids, and polysaccharides adsorbed on the
588 membrane surface strongly contributed to halogenated by-products formation upon NaClO addition.
589 Moreover, the increase of NaClO concentration, as well as temperature, promoted the formation of
590 these hazardous compounds, which are recognized as serious threats to water quality and human
591 health. In accordance with the previous results, Zhang et al. [110] found that cleaning fouled PVC
592 membranes with 500 mg/L NaOH for 1.0 hours exhibited a negligible effect on irreversible fouling
593 resistance (1.5% ± 1.0%) while much higher efficiencies (88.4 ± 1.1)% were registered using 100 mg/L
594 NaClO. Intermediate cleaning performances in terms of efficiency (47.1% ± 1.2% and 21.6% ± 3.5%)
595 were obtained with HCl (500 mg/L) and EDTA (150 mg/L). However, it is worth noting that most of
596 these studies have focused only on the flux recovery without considering the potential negative impact
597 of invasive cleaning on long-term operation.

598 **3.2.3. Combined cleaning**

599 New research approaches are focusing on the combination of the shear effect provided by air scouring
600 systems with traditional cleaning methods. Dual membrane cleaning techniques applying backwash and
601 air-water flushing were studied by Huang et al. [111] for harvesting *Chlorella sp.* Backwash using
602 permeate water was found to be more effective than forward air-water flushing in the fouling control,
603 reaching flux recoveries of 90-100% and 29-66%, respectively. Additionally, a cleaning strategy whereby
604 a first hydraulic cleaning by backwash is followed by a second forward flushing was recommended
605 based on the hypothesis that backwash can disturb foulant deposits on the membrane surface and the
606 following forward flushing may hopefully remove the fluidized foulants detached during backwash.
607 Recent applications on Chemically Enhanced Backwash (CEB) whereby, a dosage of chemical reagents
608 such as NaCl and NaClO is applied in parallel with backwash, have proved to confer supplementary

609 effects between both processes and particularly, to minimize irreversible biofouling. By means of
610 oxidation and dissociation, the chemicals weaken cohesive in the foulant-membrane interface and
611 enhance the effect of backwash, although this is highly reliant on dosages and frequencies applied and
612 further investigation is required on this realm. Tang et al. [117] found in that in freshwater-containing
613 algae, NaOCl applied at a dosage of 10 ml/L twice per day was optimal in fouling control due to the
614 dissociation of cross-linked gel structures and to the inhibitory effect in microorganisms; differently,
615 NaCl is cheaper and greener in terms of biodegradation and toxicity, yet it led to a lower fouling control
616 effect and demanded large dosages of 500 mg/L. Similarly, Kang et al. [118] used dosages of 300 mg/L
617 in CEB to cope with algal inflow in membrane desalination, reaching recovery rates of 98.1% TMP
618 upon CEB treatment.

619 **3.2.4. Mechanical cleaning**

620 Shear rates are commonly applied in membrane systems to promote a turbulent flow regime that
621 facilitates the back-transport of foulants into the feed solution [66]. They might be provided from the
622 feed via crossflow velocity, from the scouring systems via air bubbles, or by mechanical means, such as
623 vibration or rotation of the membrane modules [105]. The latter techniques are termed dynamic
624 membrane filtration systems (DMF) [86, 105]. In DMFs, a relative motion between the membrane
625 surface and the fouling interface is applied within a 0 to 0.5 mm distance; this is intended to protect the
626 algal cells in the feed from shear-induced damage [96]. The shear force applied on the membrane
627 surface disrupts both the cake layer and the concentration polarization layer, although shear forces
628 should be kept below a threshold to prevent cell breakage [106].

629 The vibration.based systems use high vibration frequencies or amplitudes to create a shear rate at the
630 membrane-feed interface. Jiang et al. [96] found an increase in the antifouling performance when the
631 vibration frequency was increased from 1 to 5 Hz. This result was due to a shift in interfacial forces,
632 becoming lifting forces predominant over drag forces at higher frequencies, thus minimizing the
633 deposition on foulants on the membrane. Further, mild vibrations enhanced the inter-foulant collision,

634 promoting the formation of large algal aggregates with a lower tendency to deposit on the membrane.
635 Recently, Zhao et al. [97] created a composited fouling control system made of vibration, negative
636 charge, and patterned polysulfone/sulfonated polysulfone (PSf/sPSf) membrane blends. The waves-
637 patterned membranes exhibited larger pore size, porosity and hydrophilicity when increasing the sPSf
638 concentration, thereby displaying higher water permeability. The patterns promoted turbulence,
639 generating a shear effect that was enhanced by the turbulent regime that the vibrations induced at
640 frequencies above 7 Hz. Patterns and vibrations promoted the generation of turbulence eddies between
641 patterns, thereby scouring the foulants with a higher efficiency.

642 # 4. Sustainable microalgae membrane-based separation
643 perspectives

644 Membrane-driven separation processes have the several advantages respect other possible algae
645 harvesting solutions due to lower equipment footprint, high recovery, and the ability to preserve high
646 quality of the concentrated biomass for downstream processing [18, 19]. However, membrane fouling
647 is responsible for the increase in energy consumption and for membrane deterioration [119]. While a
648 slight flux decline is observed when filtering algae-free growth medium alone, high values of flow
649 reduction occur only during the filtration of the algal suspension, indicating that the algal cake layer
650 dominates the process performance efficiency [37, 120]. A reduction of fouling and its better control
651 will translate directly into energy savings due to the easier filterability linked to higher algae harvesting
652 performances [51].

653 Based on this literature review, further necessary research investigations in microalgae harvesting and
654 fouling control have been identified in the following areas, which are also summarized in **Fig. 4**:

655 (i) Optimization of membrane configurations for an efficient fouling control and sustainable algal
656 harvesting.

657 (ii) In-depth investigation of microalgae and AOM composition and their interactions with
658 membranes.

659 (iii) Integration of effective and standardized fouling characterization procedures, fundamental for a
660 comprehensive understanding of fouling dynamics. To this extent, a variety of parameters such as
661 flux, retention rate, fouling resistance, and concentration factors have been used as reference
662 performance and fouling indicators, but without uniformity among research protocols; different
663 studies can hardly be compared with each other.

664 (iv) Understanding the role of algae biofilm and cake layer developed on the membrane, in both
665 separation efficiency and biochemical transformations during algae separation [29].

(v) Develop models to predict the fouling mechanism in algal processes for biomass harvesting.

(vi) Quantitative environmental and energetic assessments of membrane-based algae concentration processes aimed to precisely estimate and reduce the energy demand during production of biofuels from algae biomass.

(vii) Develop tailored fouling control strategy to allow the simultaneous recovery of valuable algal by-products and restoration of membrane performance [121].

(viii) Processes scale-up, crucial to link the lab-scale literature results with full scale applications, focusing on the energetic aspect [71].

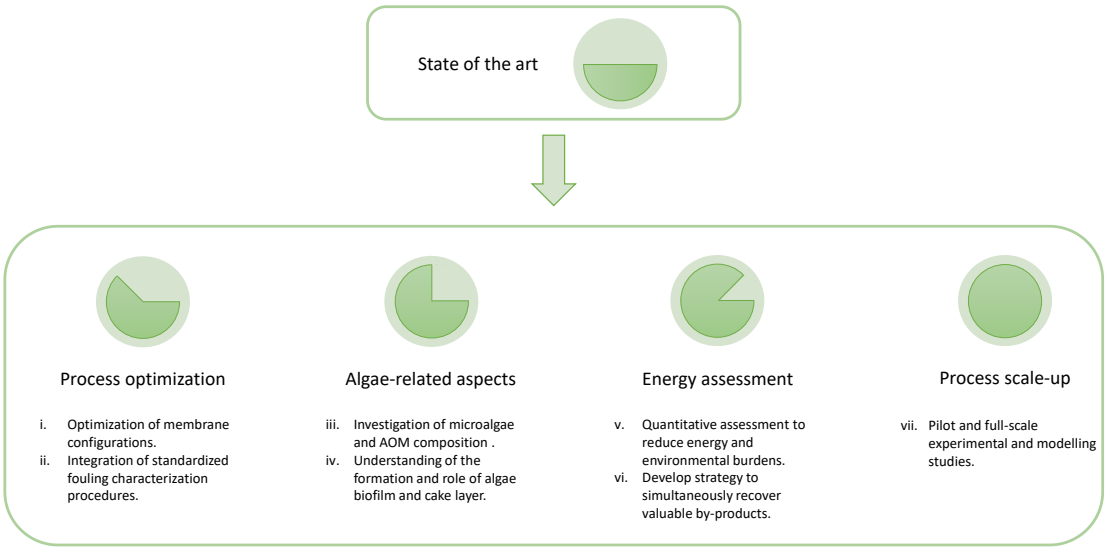


Fig. 4: Summary of the main research needs to step up microalgae harvesting processes.

The harvesting and concentration process of microalgae still requires large water volumes, high energy consumption and investments, which combined are estimated to account for over 30% of the total production cost of algal biomass [122-124]. Research is required to investigate the possibility to reuse the collected permeate directly or indirectly as a new growth medium for subsequent microalgae growth cycles, thus approaching zero liquid discharge (ZLD) perspectives, improving the process sustainability. This goal will require high-quality permeate streams that can only be achieved if the fouling phenomena is carefully controlled. Moreover, to reduce the water footprint of algae cultivation systems, seawater,

saline aquifer water, and wastewater may be also used as lower-value water sources for algae production with careful consideration about the content of N and P, and other micronutrients [125-127].

Acknowledgements

The research reported in this paper was supported by funding from King Abdullah University of Science and Technology (KAUST), Saudi Arabia and by Politecnico di Torino, Italy. Francesco Ricceri acknowledges funding from the CleanWaterCenter@PoliTo for his Ph.D. scholarship (01_TRIN_CI_CWC).

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6. Abbreviation appendix

Nomenclature

AOM	Algal organic matter	MPBR	Membrane photobioreactor
AS-CS	Sulfate-chitosan coagulant	NFM	Nanofiber membrane
CEB	Chemically enhanced backwash	PAC	Powdered activated carbon
CFV	Cross-flow velocity	PES	Polyethersulfone
CP	Cold plasma	PS	Persulfate
DAF	Dissolved air flotation	PSD	Particle size distribution
DBP	Disinfection by-products	PSF	Polysulfone
DMF	Dynamic membrane filtration system	PMS	Permonosulfate
DC	Direct current	PVDF	Polyvinylidene fluoride
DOC	Dissolved organic carbon	SPC	Sodium percarbonate
EOM	External organic matter	SRT	Solids retention time
EPS	Extracellular polymeric substances	TDS	Total dissolved solids
HMW	High molecular weight compounds	TEP	Transparent exopolymer particles
HRT	Hydraulic retention time	TMP	Transmembrane pressure
IOM	Internal organic matter	UF	Ultrafiltration
LMH	Flow in liters per square meter per hour	WEN	Water-energy nexus
LMW	Low molecular weight compounds	ZLD	Zero liquid discharge
MF	Microfiltration		