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Association of dissolved air flotation (DAF) with microfiltration for cyanobacterial removal in water supply

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ABSTRACT

The removal of filamentous cyanobacteria in natural water was investigated by a sequential treatment involving dissolved air flotation (DAF) (preceded by coagulation/flocculation) and microfiltration (MF) technologies. The combined treatment was considered to evaluate the contribution of the DAF process as a pre-treatment for MF to mitigate the impact on the microfiltration performance caused by the presence of the cells. The bench-scale experiments were conducted with surface water samples. For C/F steps, a dose of 40 mg L^{-1} of aluminium sulphate was used, and several operational conditions were adopted for the DAF tests. For the MF experiments, polyvinylidene fluoride (PVDF) membranes (0.3 μm) and a working pressure of 1 bar were used. The parameters colour, turbidity, $UV_{254\,nm}$ absorbance, DOC and cyanobacterial density, were evaluated. The DAF-MF sequence showed a considerable reduction in the residual values for all parameters compared with the DAF process alone. MF was able to remove cyanobacterial cells completely. The use of DAF as an MF pre-treatment indicated an increase in the percentages of removal. However, the particulates remaining in the water from the DAF process resulted in clogging of the membrane. The results indicate that the association of DAF and MF technologies can be a viable option to be used for water treatment with cyanobacterial cells.

Keywords: Water treatment; Cyanobacteria; Dissolved air flotation; Microfiltration

1. Introduction

Cyanobacterial blooms have become common events in recent years, and their presence in lakes and water reservoirs is a significant concern for drinking water production. These events have been observed in several places in the world [1–4], normally with exposure to toxins, and in Brazilian reservoirs, cyanobacterial blooms have intensified [5–7]. In eutrophic waters, the growth of cyanobacteria is intensive. Although eutrophication is a natural biological process, in certain water bodies, this process is accelerated by the input of nutrients of anthropogenic origin, which contributes to the massive growth of cyanobacteria [8–10]. These organisms cause drastic changes in the water body and can influence the water treatment process.

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Conventional treatment processes (coagulation, flocculation, sedimentation, filtration and disinfection) are normally used for drinking water production from eutrophic lakes. With the increase in the cyanobacteria population, a higher demand for coagulants and shorter filtration cycles can be observed. These operating conditions are due not only to the higher number of cells but also to the associated algogenic organic matter (AOM) (intracellular or extracellular metabolites) [11]. Both AOM and cells can contribute to the formation of disinfection by-products, such as trihalomethanes (THM) and haloacetic acid (HAA) [11–13]. Conventional drinking water treatment can be considered a safe barrier against cyanobacterial cells but is not highly effective in removing toxins originating from the release of cell-bound toxins [14]. For treatment steps, several authors reported that the conventional treatment may cause cell lysis with the release of intracellular toxins into the water, while others observed no release of these metabolites [8,15,16].

Dissolved air flotation (DAF) has been considered to be an interesting alternative process to treat water from eutrophic lakes. Because of their low density and ability to float, cyanobacterial cells can be removed more effectively by DAF than by the conventional settling treatment [15]. Some studies indicate a higher efficiency of cyanobacterial cell removal by DAF with no cyanobacterial damage and no toxin release [14,15,17]. According to Henderson et al. [18], DAF tends to have the most efficient removal rates for algae and cyanobacterial cells (greater than 90%) compared with sedimentation (between 70 and 80%). Direct filtration is considered to be more susceptible to the changing algae character. Membrane filtration, which is widely used in chemical and biotechnology processes, has become a significant technology for the treatment of surface water. Microfiltration (MF) is typically used as a clarification process, and in water treatment systems, this technology may be used as a polishing step following conventional treatment or as a pre-treatment for other membrane processes, such as nanofiltration and reverse osmosis [19]. With porous diameters measuring between 0.1 and 10 μm [20], MF membranes are less effective in the removal of dissolved compounds, such as humic and inorganic substances [21]. To remove these substances, a coagulation/flocculation step is applicable. For surface water treatment with cyanobacteria, an MF membrane can be regarded as a good treatment for removing the remaining cells after conventional treatment, including DAF. Due to its higher pore size, an MF membrane can reduce the impact on the cyanobacterial cell structure and prevent the release of intracellular substances (e.g. toxins), which was not observed by Gijsbertsen-Abrahamse et al. [22] in the removal of *M. aeruginosa* cells by UF membranes. Thus, a DAF–MF association can favor a higher removal of intact cells of cyanobacteria, with no toxins being released into the treated water.

To evaluate the applicability of DAF and membrane technology for removing cyanobacterial cells in a water treatment system, this study aimed to (1) evaluate the DAF–MF association for cyanobacterial cell removal in natural waters and (2) verify the DAF process as a pre-treatment to MF to mitigate the impact of cells on the performance of microfiltration.

2. Methodology

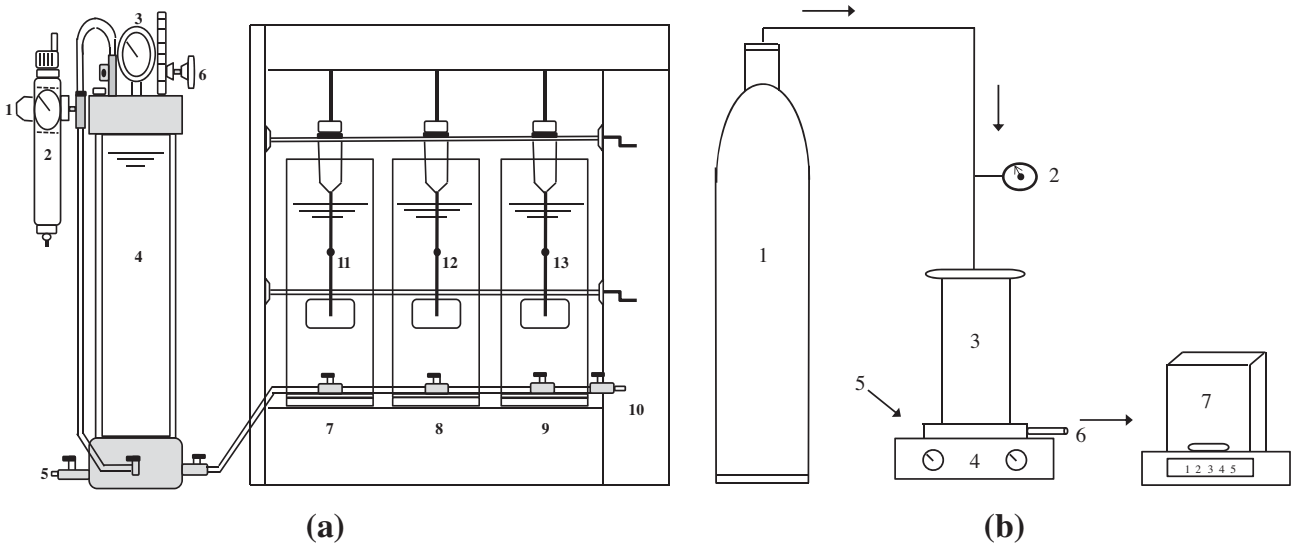
2.1. Natural water samples

The water used in this study was collected from Lagoa do Peri (Peri Lagoon), a freshwater coastal lagoon in Florianopolis, Santa Catarina, Brazil, which is used to provide drinking water to approximately 113,000 people. High densities of microalgae and potentially toxic cyanobacteria were verified, and the predominant species was *Cylindrospermopsis raciborskii*. These organisms in the water have resulted in operational difficulties in the currently employed treatment system (rapid filtration with a descending flux).

During this study, the color, turbidity, $UV_{254\,nm}$ absorbance, dissolved organic carbon (DOC), cyanobacterial density, and chlorophyll-a (chl_a) were constantly analyzed, with mean values of 55 HU, 5.3 NTU, $0.0675\,cm^{-1}$, $5.1\,mg\,L^{-1}$, $39{,}000\,ind\,mL^{-1}$, and $19.4\,\mu g\,L^{-1}$, respectively.

2.2. Coagulation/flocculation/DAF experiments

The DAF experiments were performed at room temperature (20 ± 2°C) using laboratory flotation equipment (Fig. 1(a)). This apparatus has a 2 L pressure chamber and three calibrated cylinders with 2 L each. Preliminary tests were performed to determine an optimum coagulant dosage (aluminium sulphate – $Al_2(SO_4)_3{\cdot}16H_2O$) considering the best removal rates for color, turbidity, and cyanobacterial density. From these tests, a coagulant dose of $40\,mg\,L^{-1}$ (coagulation pH of 4.91) was established. The DAF operating conditions studied were: (a) coagulation at G_C $1{,}000\,s^{-1}$ for 10 s; (b) flocculation at G_F $25\,s^{-1}$ for 10 min; and (c) DAF for 8 min, a relative pressure of 4 bar, an applied recycle ratio (R) of 10%, and a bubble rising velocity (V_F) of $5\,cm\,min^{-1}$. The sampling time was determined from Eq. (1).



(1) air inlet; (2) pressure valve; (3) manometer; (4) pressure chamber; (5) water inlet; (6) air outlet; (7/8/9) distribution of saturated water; (10) depressurization; (11/12/13) sampling.

(1) synthetic air cylinder; (2) pressure meter; (3) stirred cell; (4) magnetic stirrer; (5) membrane compartment; (6) permeate outlet (7) electronic balance

Fig. 1. Laboratory bench-scale flotation test (a); dead-end stirred cell set-up (b).

$$S_T\ (\text{min}) = \frac{12\ (\text{cm})}{V_F\ (\text{cm} \cdot \text{min}^{-1})} \quad (1)$$

where 12 cm is the height of sampling point.

Samples of treated water were analyzed for color (HACH 2010 spectrophotometer), turbidity (HACH 2100P turbidimeter), $UV_{254\,nm}$ (Varian UV/VIS spectrophotometer), DOC (Shimadzu TOC 5000A analyzer), and cyanobacterial density (Sedgewich Rafter chamber), all of which were determined using standard methods for analysis [22], and chl_a (HACH 4,000 spectrophotometer) was determined according to Nusch [23]. The water treated by DAF was used for sequential MF tests. A total of 20 tests were performed during one year of sampling.

2.3. Microfiltration experiments

The MF experiments were performed in a dead-end stirred cell system (Fig. 1(b)) made of stainless steel and with 200 mL of useful volume. The membrane used is an asymmetrical microporous polyvinylidene fluoride (PVDF) flat-sheet membrane (0.3 μm, GE Hosmonics®) with an effective filtration area of $3.8 \times 10^{-4}\,m^2$. Scanning electron microscopy (SEM) analyses were performed to observe the morphological characteristics of the PVDF membrane (Fig. 2). From the initial tests, a pressure of 1 bar was established as the ideal working pressure for the unit of filtration used.

In each previous test, to eliminate the impurities inherent in the manufacturing process, the membranes remained soaked in ultra-pure water for at least 24 h [24,25]. The membranes were first compacted with ultra-pure water for 60 min, which is the time required for achieving a steady permeate flux.

The MF experiments were performed with water treated after DAF (DAF–MF) and surface water without treatment. For the first goal, the water previously treated by DAF (Section 2.2) was transferred to the dead-end stirred cell unit and was filtered for 180 min at constant pressure (1 bar). After the filtration time, samples from the permeate solution were taken and analyzed for color, turbidity, $UV_{254\,nm}$, DOC, and cyanobacterial density. The flux behavior during the filtration experiment was also evaluated. After passing the sample (water treated by DAF), a new filtration with ultra-pure water was performed for 60 min to verify the membrane *fouling* occurrence.

In tests using only microfiltration, the surface water was placed directly on the dead-end stirred cell unit. A filtration time of 180 min was performed, and samples were taken at 0, 30, 60, 90, 120, 150, and 180 min for analyses for the same analytic parameters. The behavior of permeate flux was also evaluated. The analytic

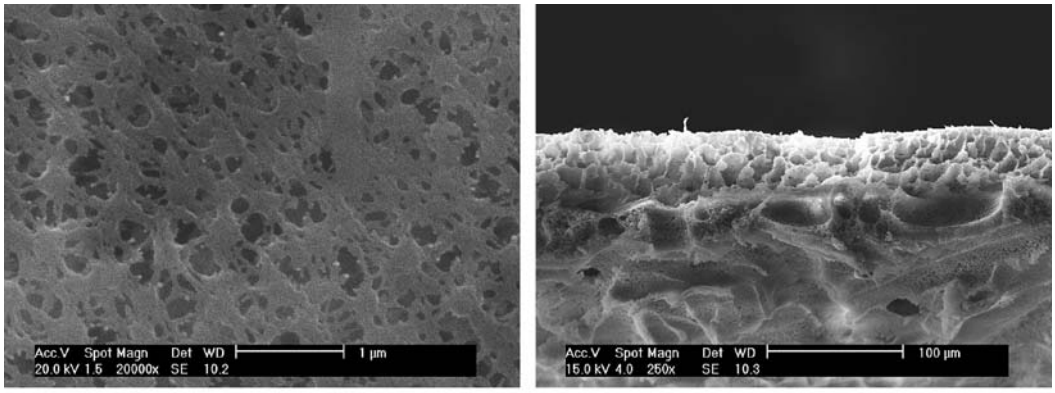


Fig. 2. SEM micrographs of the MF membrane surface (a) and membrane cross-section (b).

results of this step were compared with DAF-MF experiments for the same surface water sample.

3. Results and discussion

3.1. DAF and DAF + MF experiments

Table 1 shows the mean values obtained for the evaluated analytical parameters (color, turbidity, $UV_{254\,nm}$, DOC, and cyanobacterial density) for raw water and for water treated by DAF and by DAF + MF.

The mean color value found after the coagulation/flocculation/DAF (C/F/DAF) process was 16.7 HU, which does not meet the maximum color value in the treated water recommended by Brazilian legislation (15 HU), confirming the need for a post-treatment to obtain better quality treated water. The mean value for this parameter removal was 69%. After microfiltration, the residual values of color were significantly lower than those obtained only with DAF: 1 HU (98% of removal) (Fig. 3(a)). Similar values with over 95% removal were obtained by other authors [26] using microfiltration membranes (mean pore size of 0.2 µm) associated with coagulation for drinking water treatment.

For turbidity, a mean residual of 2.08 NTU and a mean percent removal of 60% were obtained after C/F/DAF treatment. With microfiltration, significant turbidity reductions were obtained with a mean residual value of 0.43 NTU (92% removal) (Fig. 3(b)). A value of 99.6% (0.2 NTU) removal was observed by Bottino et al. [27] for a ceramic microfiltration membrane (mean pore of 0.2 µm) to treat water from Lake Brugneto (Genoa, Italy). Removal up to 71% for cyanobacteria was obtained only with C/F/DAF (Fig. 3(c)). Nevertheless, the remaining amount of organisms (9,872 ind mL^{-1}) is still considered high. After membrane filtration, complete cyanobacterial removal was obtained, which was due to the morphology of the cyanobacterial species in the surface water studied (filamentous) and the small pore size in relation to the filament size.

The permeate fluxes for MF membranes are shown in Fig. 4. The mean permeability for the MF membrane used was 3,357 L m^{-2} h^{-1}. The permeate flux behavior for sample filtration presented a notably irregular pattern, with volumes ranging from 159 to 697 L m^{-2} h^{-1}, which is much lower than the mean permeate flux for clean membranes. The *fouling* effect on the membranes can be observed by assessing the permeate flux with ultra-pure water after sample filtration. The similar values indicate membrane pore obstruction by the cyanobacteria and other particulates not removed by DAF.

Table 1
Mean values and percentages of color, turbidity, $UV_{254\,nm}$, DOC and cyanobacterial removal from raw water, DAF, and DAF + MF

	Color (HU)	Turbidity (NTU)	Absorbance ($UV_{254\,nm}$) (cm^{-1})	DOC (mg L^{-1})	Cyanobacterial density (ind mL^{-1})
Raw water	54.25	5.32	0.07	4.87	36,000
DAF	16.7 (69%)	2.08 (60%)	0.03 (52%)	3.03 (38%)	9,872 (71%)
DAF + MF	1 (98%)	0.43 (92%)	0.02 (71%)	2.33 (52%)	0,00 (100%)

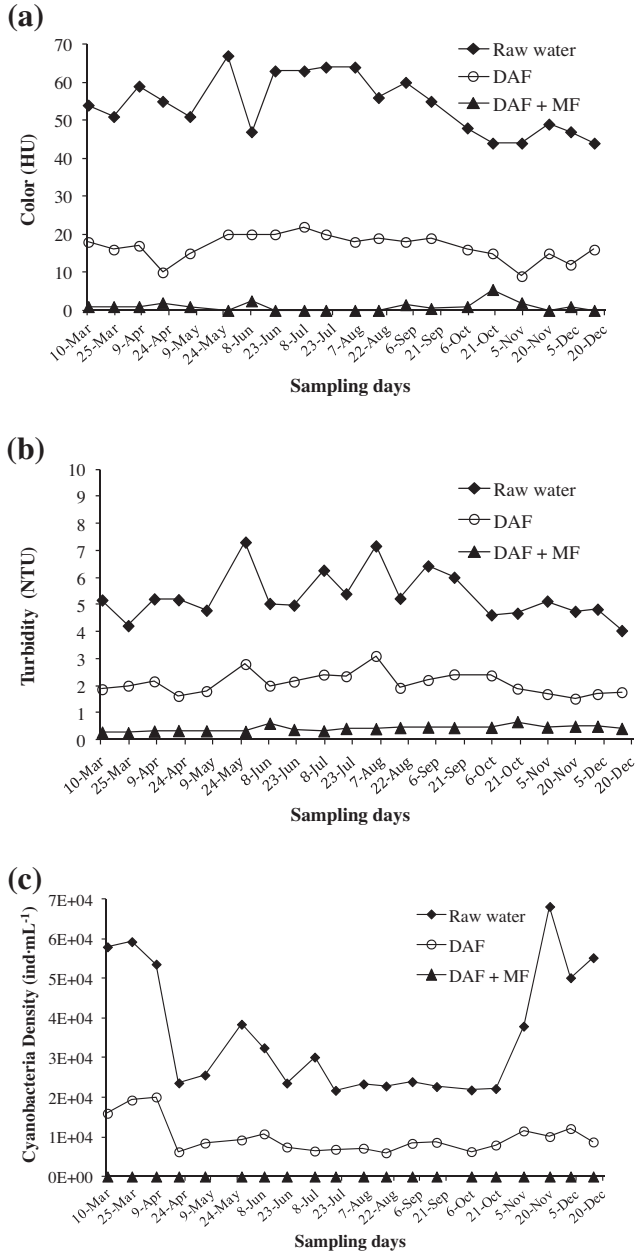


Fig. 3. Results from DAF and DAF + MF experiments: color (a), turbidity, (b) and cyanobacterial density (c).

According to Wakeman and William [28], the fouling formation in MF membranes is due to two distinct phenomena: the blocking of the membrane pores by the deposition of larger particles on the membrane surface and the deposition of smaller particles into the membrane pores. For the first condition, Eagles and Wakeman [29] believe that the larger the particle diameter deposited on the membrane surface, the greater the reduction in permeate. Another phenomenon that may cause *fouling* in microfiltration is the concentration polarization. The effects of this phenomenon on the

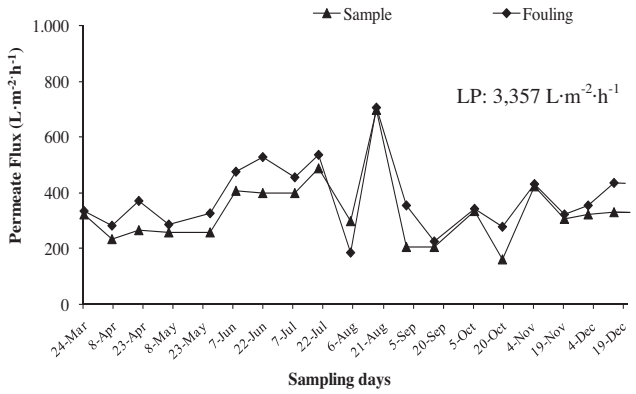


Fig. 4. MF means flux for sample filtration and *fouling* evaluation with ultra-pure water. (Lp = is the average of permeate flux with a clear membrane).

microfiltration can be severe because the flux in this type of filtration is high, and the coefficient of mass transfer is reduced due to the low diffusion coefficients of macromolecular solutes, small particles, colloids, and emulsions [28].

3.2. MF and DAF + MF experiments

In this experiment, it was considered to evaluate whether the pre-treatment with DAF could improve the performance of the MF membrane and possibly increase its useful life.

The permeate flux behavior for MF and DAF + MF experiments can be observed in Fig. 5. For microfiltration, a notably low initial and constant flux was observed, indicating the immediate blocking of membrane pores by the direct filtration of water. Fluxes between 543 $L\,m^{-2}\,h^{-1}$ (initial) and 159 $L\,m^{-2}\,h^{-1}$ (after 180 min) were registered. With prior treatment by DAF, the initial flux was higher (5,435 $L\,m^{-2}\,h^{-1}$) with a sudden flux drop after 10 min of filtration (1,442 $L\,m^{-2}\,h^{-1}$) and constant reduction. Notably

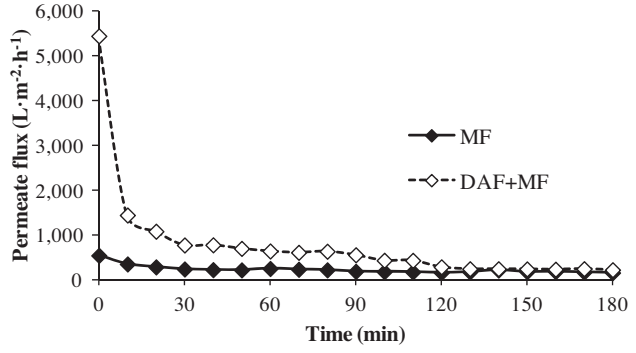


Fig. 5. Flux behavior for MF membrane for the filtration of the sample after DAF and direct filtration.

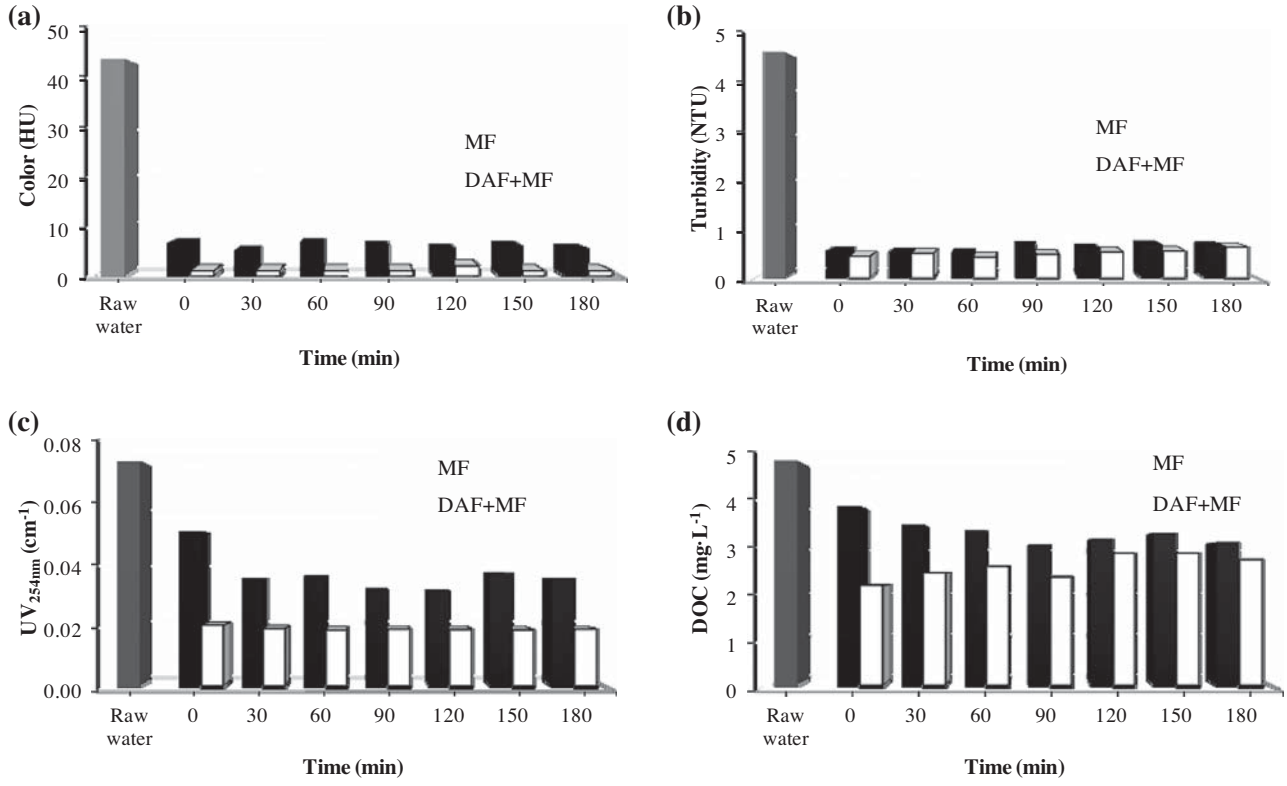


Fig. 6. Results from the MF and DAF + MF experiments: color (a), turbidity (b), $UV_{254\ nm}$ (c), and cyanobacterial density (d).

close permeate flux values for DAF + MF and MF were verified after 120 min. Although the pre-treatment by DAF initially reduced the impact of the water quality on the membrane, the remaining particulate material similarly promoted the clogging of the membrane. Goh et al. [30] also observed an enhancement in the flux rate when they applied DAF as a pre-treatment for microfiltration. The authors identified that the particulates larger than 1.5 μm contributed to membrane fouling by depositing onto the membrane surface.

For the analytical parameters analyzed, the percentages of removal were higher in DAF + MF experiments than in MF-only experiments with lower residual values. The higher removal efficiencies for the water pre-treated by DAF are observed mostly for color (Fig. 6(a)) and absorbance (Fig. 6(c)) parameters. Whereas the color is mainly related to dissolved substances in water, the step of coagulation/flocculation allowed an aggregation of these substances in particles to be retained by the membrane, thereby reducing the final color of the treated water. The percentage removals ranged from 95 to 98% for DAF + MF (1.14 HU) and from 85 to 89% for MF (5.8 HU), which indicates that the removal efficiency was 13% higher in DAF + MF. In terms of absorbance, the results indicate a reduction between 73 and 78% for DAF + MF treatment and 31–57% for MF treatment. For turbidity (Fig. 6(b)) and DOC (Fig. 6(d)), the results indicate no expressive difference in reduction between the two treatments. For turbidity, percentages of removal up to 91% for DAF + MF and 89% for MF were obtained, and for DOC, values of up to 55% for DAF + MF and 37% for MF were obtained. Complete removal of the cyanobacteria was observed for both treatments.

From the obtained results, we can verify that a pre-treatment can improve the microfiltration performance. Kim et al. [31], using a microfiltration membrane (with a pore size of 0.1 μm) associated with fluidized activated carbon for the treatment of water from the Tama river (Japan), obtained 84% DOC removal and a 94% absorbance reduction. The removal values were higher when only the microfiltration treatment was used, with values of 7.8% for DOC and 19% for absorbance reduction being observed.

4. Conclusions

In this study, the association of DAF and MF technologies were evaluated for cyanobacterial cell removal in natural waters. For DAF alone, the residual value of color in the treated water was above the maximum value allowed by Brazilian legislation. The DAF

removed a considerable amount of the cyanobacteria, but due to the high density of these organisms in raw water, the concentration in the treated water was still considered high. With the association of DAF with MF, the residual values in the treated water, mostly for the parameters of color and turbidity, were significantly lower compared with the values obtained when using DAF alone. Total cyanobacterial removal was verified when MF was used in association with DAF. The sample flux obtained using the PVDF membranes for microfiltration showed significant variations between the tests. These fluxes were much smaller than the flux obtained with a clean membrane and smaller than the flux obtained with the ultra-pure water filtration performed after the sample filtration, which indicates the formation of membrane fouling. When using microfiltration alone, a notably low initial and constant flux was observed, indicating the immediate blocking of the membrane pores by the direct filtration of water. With DAF + MF, the initial flux was higher with a sudden flux drop after 10 min of filtration. A constant reduction was observed. Similar permeate flux values for MF and MF+DAF were verified at 120 min. Although the pretreatment by DAF initially reduced the impact of the water quality on the membrane, the particulate material remaining still eventually clogged the membrane. For the analytical parameters analyzed, color, turbidity, $UV_{254\,nm}$, and DOC, the percentages of removal were higher in DAF + MF than in the MF-only experiments with lower residual values. From these conclusions, it was observed that the association of these two technologies promotes an improvement in the quality of treated water and in the permeate flux. This association of technologies proved to be a viable option that can be used for water treatment of water containing cyanobacteria.

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