Impact of *Microcystis aeruginosa* on membrane fouling in a biologically treated effluent

Y. T. Goh, J. L. Harris and F. A. Roddick

**ABSTRACT**

*Microcystis aeruginosa* was cultured in biologically treated municipal effluent to simulate blue-green algal bloom conditions in a treatment lagoon. The effect of algae in the early, mid and late phases of growth on membrane fouling, chemical coagulation (alum or aluminium chlorohydrate (ACH)) and hydraulic cleaning on the microfiltration of this effluent was investigated. The effect of *M. aeruginosa* in the early phase was negligible and gave a similar flux profile and permeate volume to that of effluent alone. The increase in *M. aeruginosa* concentration for the mid and late phases caused a significant reduction in permeate volume compared with the early phase. Full flux recovery was achieved with an alum dose of 1 mg Al³⁺ L⁻¹ (early phase) and 10 mg Al³⁺ L⁻¹ (mid phase), demonstrating that membrane fouling was hydraulically reversible. For the late phase, the highest flux recovery was 89%, which was achieved with an alum dose of 5 mg Al³⁺ L⁻¹. Higher alum dosages resulted in a reduction in flux recovery. The use of 1.5 µm pre-filtration after alum treatment showed little improvement in water quality but led to a drastic reduction in flux recovery, which was attributed to diminishing the protective layer on the membrane surface, thus enabling internal fouling. The performance of ACH was comparable to alum at low dissolved organic carbon (DOC) and cell concentration, but was not as effective as alum at high DOC and cell concentration due to the formation of more compact ACH flocs, which resulted in a higher cake layer specific resistance, leading to the deterioration of performance.

**Key words** | biologically treated effluent, coagulation, Cyanobacteria, *Microcystis aeruginosa*, microfiltration, pre-treatment

**INTRODUCTION**

Periodic blooms of Cyanobacteria (commonly known as blue-green algae) in wastewater treatment lagoons lead to water quality problems such as odour, release of toxins, membrane fouling and the production of disinfection by-products on disinfection of the water (Jones & Orr 1994; Gao et al. 2009). Such periodic large blooms typically involve *Microcystis aeruginosa* (Vasconcelos & Pereira 2001). The presence of *M. aeruginosa* can impact severely on the performance of microfiltration and ultrafiltration modules used to condition the feedwater to reverse osmosis units in the desalination of biologically treated effluent.

Various studies have been conducted by spiking *M. aeruginosa* into surface, lake, reservoir or growth media prior to immediate treatment (e.g. dissolved gas flotation, nanofiltration, electro-coagulation) to remove *M. aeruginosa* (Teixeira & Rosa 2006; Gao et al. 2010; Heng et al. 2009; Zhang et al. 2009). Chemical coagulation is widely used and found to be effective in algal removal. Henderson et al. (2008) reviewed several publications on the removal of *M. aeruginosa* from reservoir water and growth media. For the former, chemical coagulation using alum, ferric chloride and a cationic polymer (C-573) as a coagulant aid achieved cell removals of 75%–99%, whereas 62%–82% of *M. aeruginosa* in growth media was removed by ferric sulphate and polyferric sulphate treatment. However, little information is available on the pre-treatment of *M. aeruginosa* at different growth stages in a biologically treated effluent for mitigating the impact of membrane fouling.

In our study, we investigated the impact of *M. aeruginosa* in an activated sludge-lagoon effluent of a municipal sewage treatment plant on its treatment by microfiltration and the efficacy of pre-treatments for fouling mitigation.
during the early, mid and late phases of the algal life cycle. *M. aeruginosa* was grown in the effluent to simulate the actual conditions of an algal bloom, as interactions between algal cells and particulates/foulants, or release of intracellular/extracellular materials, may occur and impact on the filtration performance. Chemical coagulation using alum or aluminium chlorohydrate (ACH) was employed as pre-treatment, and their effects were compared by examining the flux profiles, permeate volume collected, removal of algalogenic and effluent organic matter, reduction of irreversible fouling and residual aluminium concentration in the permeate.

**MATERIALS AND METHODS**

**Algal growth in wastewater**

The effluent was collected from the head of road storage (HORS) at Western Treatment Plant, Victoria, Australia in May 2009. *Microcystis aeruginosa* (CS 566/01-A01) was obtained from CSIRO Microalgae Research Centre (Tasmania, Australia) and was maintained in MLA medium at 22 °C under humidified aeration (0.45 µm filtered) with 16/8 h light/dark cycle. Chlorophyll-a (Chl-a) measurement was determined according to USEPA (1999) as an indication of algal growth. The relationship between cell count and Chl-a measurement was found to be linear ($R^2 = 0.988$) up to a cell concentration of $2.0 \times 10^7$ cells mL$^{-1}$:

$$\text{Cell Count (cells mL}^{-1}) = 6,848 \text{ Chl-a (µg L}^{-1})$$  \hspace{1cm} (1)

HORS (60 L) was inoculated with 120 mL of *M. aeruginosa* (Cyanobacterial biomass on Day 16: $6.4 \times 10^6$ cells mL$^{-1}$ or 928 µg L$^{-1}$ Chl-a). *M. aeruginosa* (AHORS) was grown at 20 °C under humidified aeration (0.45 µm filtered) with 16/8 h light/dark cycle in a water-jacketed enclosed tank. The interior walls of the tank were gently scrubbed occasionally using a brush to minimize the loss of algae. The physical and chemical characteristics of HORS, AHORS$^1$ (Early Phase: Day 0–8), AHORS$^2$ (Mid Phase: Day 19–25) and AHORS$^3$ (Late Phase: Day 46–53) are shown in Table 1.

**Microfiltration (MF)**

An Amicon 8050 stirred cell (membrane area, 13.4 cm$^2$) was connected to a feed reservoir and operated at a transmembrane pressure of 70 kPa regulated by nitrogen gas and stirrer speed of 430 rpm. All experiments were conducted at room temperature (20 ± 2 °C). The permeate flux was determined using a top-loading electronic balance (BP6100, Sartorius, accuracy ±0.1 g) with data logging function connected to a computer. Hydrophilic polyvinylidene fluoride (PVDF) MF membranes (Durapore VVPP, 0.1 µm, Millipore) were used. New membranes were soaked overnight in Milli-Q water and 500 mL of Milli-Q water was passed through each membrane prior to filtration tests. Membranes were selected for use when the measured pure water flux $J_p$ was in the range of 1,400–1,800 L m$^{-2}$ h$^{-1}$. The filtration experiments were conducted till the permeate flux reached a final value of 40 L m$^{-2}$ h$^{-1}$.

For membrane cleaning, the fouled membrane was hydraulically cleaned by surface washing and backflushing by placing the membrane upside down in the filtration cell and filtering 1 L of Milli-Q water. The membrane was returned to its original processing position and the pure water flux $J_p$ measured. Flux recovery ($J_p/J_o$) was used as an indication of the extent of membrane cleaning.

**Table 1 | Characteristics of the HORS and AHORS**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>HORS</th>
<th>AHORS$^1$</th>
<th>AHORS$^2$</th>
<th>AHORS$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>pH</strong></td>
<td>8.2</td>
<td>8.1</td>
<td>8.6</td>
<td>8.8</td>
</tr>
<tr>
<td><strong>Conductivity (µS cm$^{-1}$)</strong></td>
<td>2.080</td>
<td>2.110</td>
<td>1.916</td>
<td>1.952</td>
</tr>
<tr>
<td><strong>Turbidity (NTU)</strong></td>
<td>2.4</td>
<td>2.9</td>
<td>40.2</td>
<td>59.8</td>
</tr>
<tr>
<td><strong>DOC (mg L$^{-1}$)</strong></td>
<td>5.68</td>
<td>5.86</td>
<td>6.05</td>
<td>6.43</td>
</tr>
<tr>
<td><strong>UVA$_{254}$ (cm$^{-1}$)</strong></td>
<td>0.25</td>
<td>0.25</td>
<td>0.26</td>
<td>0.26</td>
</tr>
<tr>
<td><strong>SUVA (L m$^{-1}$ mg$^{-1}$)</strong></td>
<td>4.32</td>
<td>4.24</td>
<td>4.05</td>
<td>4.45</td>
</tr>
<tr>
<td><strong>Al$^{3+}$ (mg L$^{-1}$)</strong></td>
<td>&lt;0.2</td>
<td>&lt;0.2</td>
<td>&lt;0.2</td>
<td>&lt;0.2</td>
</tr>
<tr>
<td><strong>Chl-a (µg L$^{-1}$)</strong></td>
<td>12</td>
<td>17</td>
<td>268</td>
<td>421</td>
</tr>
<tr>
<td><strong>Algae (cells mL$^{-1}$)</strong></td>
<td>$8.2 \times 10^4$</td>
<td>$1.2 \times 10^5$</td>
<td>$1.8 \times 10^6$</td>
<td>$2.9 \times 10^6$</td>
</tr>
</tbody>
</table>

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Pre-treatment methods

Coagulation using alum and aluminium chlorohydrate (ACH), and 1.5 µm pre-filtration (GF/A, Whatman), were investigated as pre-treatments. Alum was dosed at 1, 3, 5, 10 and 20 mg Al\(^{3+}\)/L\(^{-1}\). ACH (MEGAPAC 23, OMEGA), with a basicity of 82\%, was dosed at 5 and 10 mg Al\(^{3+}\)/L\(^{-1}\). Coagulation was conducted at room temperature (20 ± 2 °C) in a laboratory jar tester unit (Phips and Bird, PB-700). Samples were subjected to rapid mixing for 1 min at 200 rpm, followed by slow mixing for 20 min at 30 rpm, and immediately micro-filtered. For selected samples, 1.5 µm pre-filtration (surrogate for media-filtration) was conducted prior to microfiltration.

Characterization and analytical methods

Samples were pre-filtered (0.45 µm, cellulose acetate, Advantec) prior to ultraviolet light absorbance (UVA\(_{254}\)) and dissolved organic carbon (DOC) analysis. UVA\(_{254}\) was measured using a UV/vis spectrophotometer (UV2, Unicam), and DOC was determined using a Sievers 820 TOC analyser. UVA\(_{254}\) was recorded using a HACH 2100P turbidimeter. Residual aluminium (Al\(^{3+}\)) concentration was determined using inductively coupled plasma mass spectroscopy (ICP-MS) (HP4500, Series 300) running on ICP-MS ChemStation software version A.01.02. Samples were adjusted to the same DOC (5 mg L\(^{-1}\)) with Milli-Q water prior to fluorescence excitation emission matrix (EEM) analysis. Fluorescence spectra were obtained using a fluorescence spectrophotometer (LS 55, PerkinElmer).

For selected samples, the fouldant layer on the membrane surface was examined by scanning electron microscopy (SEM) (Quanta 200, FEI) running in environmental scanning electron microscopy (ESEM) mode under low-vacuum conditions.

RESULTS AND DISCUSSION

Flux profiles

The flux pattern of HORS showed a sharp decline and gave a permeate volume of 719 mL (Figure 1). After inoculation of the HORS with *Microcystis aeruginosa* to give AHORS\(^1\), a similar flux pattern was obtained, as anticipated, since the overall algal cell number was not greatly increased. Increasing the alum concentration from 1 to 5 mg Al\(^{3+}\)/L\(^{-1}\) gave a significant improvement in flux profile and permeate volume. Comparable results were obtained with ACH and alum for 5 mg Al\(^{3+}\)/L\(^{-1}\).

For AHORS\(^2\), for which the cell concentration was 15 times that of AHORS\(^1\), microfiltration produced a lower permeate volume of 229 mL (Figure 2). Alum (1 mg Al\(^{3+}\)/L\(^{-1}\)) treatment did not show much improvement in flux profile and led to an even lower permeate volume. This was attributed to too low an alum concentration, which produced smaller particles, leading to higher cake layer specific resistance (Wang et al. 2008), resulting in a sharper flux profile and permeate volume of 187 mL. Increasing the alum concentration from 1 to 10 mg Al\(^{3+}\)/L\(^{-1}\) progressively improved the flux profile of AHORS\(^2\) and increased the permeate volume. ACH (5 mg Al\(^{3+}\)/L\(^{-1}\)) treatment resulted in a sharper flux decline and a lower permeate volume compared with alum at the same concentration. Due to the increased algal concentration, the overall flux performance and permeate volumes were significantly lower compared with the early phase.

For AHORS\(^3\), DOC concentration was increased three-fold and the cell concentration was 35 times that of AHORS\(^1\). Microfiltration of AHORS\(^3\) produced the lowest permeate volume of 99 mL compared with AHORS\(^1\) and AHORS\(^2\) samples (Figure 2). Alum (5 mg Al\(^{3+}\)/L\(^{-1}\)) treatment gave a sharper flux decline and achieved a lower permeate volume of 56 mL compared with AHORS\(^3\) alone due to the low alum concentration. Increasing the alum concentration from 5 to 20 mg Al\(^{3+}\)/L\(^{-1}\) gradually improved the flux profile and increased the permeate volume. The use of alum (5 mg Al\(^{3+}\)/L\(^{-1}\)) followed by 1.5 µm pre-filtration showed significant improvements in flux profile and permeate volume of 179 mL, which is almost twice that of AHORS\(^3\) alone. Comparing at 10 mg Al\(^{3+}\)/L\(^{-1}\), a lower permeate volume of 70 mL was obtained with ACH compared with 146 mL for alum. The poor ACH performance for the mid and late phases was attributed to the differing nature of the flocs.

As reported by Wang et al. (2008), the fractal dimension of flocs formed by the polymeric ACH is higher than for...
those formed by monomeric alum, indicating that the Keggin structure produced by ACH is much more compact than the hexameric ring structure of alum hydrolysis species. Consequently the ACH cake layer specific resistance is far higher, and the flux deteriorates much more severely when coagulated by ACH than by alum (Wang et al. 2008), leading to lower permeate volumes.

**Membrane cleaning**

In addition to the overall permeate volumes, flux recoveries after hydraulic cleaning and backflushing of the fouled MF membranes are shown in Figure 2. As the algal concentration increased, there was a decrease in the permeate volume. Improvement in permeate volume was achieved with the use of alum or ACH treatment on AHORS over the growth cycle. The best alum dosage for full flux recovery increased with algal concentration for AHORS 1 and AHORS 2. However, for AHORS 3, although increased alum concentration (5 to 20 mg Al\(^{3+}\) L\(^{-1}\)) achieved an increase in permeate volume, it led to a severe drop in flux recovery. This suggests that the most effective period for alum treatment to obtain high permeate volume and flux recovery was in the early and mid phases of a *M. aeruginosa* bloom, corresponding to AHORS 1 and AHORS 2. The use of 1.5 µm pre-filtration after alum treatment for AHORS 3 showed a significant increase in permeate volume but led to a low flux recovery of 21%. This drop in flux recovery was mainly due to the removal of the alum-coagulated flocs which diminished the protective layer on the membrane surface, and enabled foulants smaller than 1.5 µm to deposit on the membrane surface and within the pores, resulting in pore blockage and in turn leading to a reduction in flux recovery (Goh et al. 2010).

ACH treatment was comparable to alum treatment for AHORS 1 but was much less effective for AHORS 2, as evidenced by the lower permeate volume and flux recovery, and AHORS 3 in relation to permeate volume. This poor performance could be due to increase in ACH cake layer specific resistance and DOC concentration in the mid and late growth phases of the *M. aeruginosa*.

**Observations of membrane surface**

ESEM images of the membrane surface with and without alum coagulation, and after hydraulic surface cleaning and
backflushing during the late growth phase, are shown in Figure 3. The microfiltration of AHORS\textsuperscript{3} developed a foulant layer over the membrane surface (Figure 3(b)). Increasing the alum concentration from 5 to 20 mg Al\textsuperscript{3+} L\textsuperscript{-1} developed a thicker foulant layer over the membrane surface (Figures 3(c), 3(d) and 3(e)). Hydraulic cleaning of the AHORS\textsuperscript{3}-fouled membrane without alum treatment showed that some of the foulant layer was not removed from the membrane surface (Figure 3(f)), which led to a flux recovery of 53\%. With alum (5 mg Al\textsuperscript{3+} L\textsuperscript{-1}) treatment (Figure 3(g)), a cleaner membrane surface and higher flux recovery of 89\% was achieved. For 10 and 20 mg Al\textsuperscript{3+} L\textsuperscript{-1}, the foulant layer (Figure 3(h) and 3(i)) could not be removed by hydraulic cleaning and caused a drastic reduction in flux recovery. This flux reduction could be due to the overdosing of alum, which led to:

1. excessive counterion adsorption giving rise to charge reversal leading to restabilization of the flocs (Bratby 1980) and so severely fouling the membrane;
2. structural change from monomer to polymer by bidimensional growth of the hexameric ring units during alum hydrolysis (Bertsch 1985), making it harder to remove by hydraulic cleaning.

**Water quality improvement**

DOC removals by pre-treatments for the different growth phases of *M. aeruginosa* are shown in Table 2.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Coagulant dosage (mg Al\textsuperscript{3+} L\textsuperscript{-1})</th>
<th>AHORS\textsuperscript{1}</th>
<th>AHORS\textsuperscript{2} DOC removal (%)</th>
<th>AHORS\textsuperscript{3}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microfiltration (MF)</td>
<td>0</td>
<td>5</td>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>6</td>
<td>13</td>
<td>N.T.</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>10</td>
<td>10</td>
<td>N.T.</td>
</tr>
<tr>
<td>Alum + MF</td>
<td>5</td>
<td>12</td>
<td>11</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>N.T.</td>
<td>18</td>
<td>26</td>
</tr>
<tr>
<td>Alum + MF (1.5 µm filtered)</td>
<td>5</td>
<td>N.T.</td>
<td>N.T.</td>
<td>24</td>
</tr>
<tr>
<td>ACH + MF</td>
<td>5</td>
<td>14</td>
<td>14</td>
<td>N.T.</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>N.T.</td>
<td>N.T.</td>
<td>21</td>
</tr>
</tbody>
</table>

*N.T.* – not tested.
Microfiltration showed similar DOC removal for AHORS\textsuperscript{1} and AHORS\textsuperscript{2}, whereas for AHORS\textsuperscript{3} there was a slight reduction in DOC removal.

For AHORS\textsuperscript{1}, DOC removal was improved with increasing alum concentration. Comparable DOC removals were achieved with ACH.

For AHORS\textsuperscript{2}, due to the increase in cell concentration, the best DOC removal was achieved with alum (10 mg Al\textsuperscript{3+} L\textsuperscript{-1}) concentration. ACH gave a slightly higher DOC removal compared with alum at 5 mg Al\textsuperscript{3+} L\textsuperscript{-1}.

For AHORS\textsuperscript{3}, no further improvement in DOC removals was observed for alum at concentrations greater than 10 mg Al\textsuperscript{3+} L\textsuperscript{-1}. Coupling 1.5 µm pre-filtration to alum coagulation showed insignificant improvement in DOC removal compared with alum coagulation alone. Lower DOC removal was obtained in the late phase with ACH.

In all cases, the removal of turbidity was greater than 98\%, residual aluminium concentrations were below 0.2 mg L\textsuperscript{-1}, and residual chlorophyll-a concentrations were negligible.

**Fluorescence excitation emission matrix (EEM) spectra**

EEM spectra were employed to observe the changes in fluorescent DOC of AHORS during the growth cycle of \textit{M. aeruginosa}. The regions have been designated according to Chen \textit{et al}. (2005) as follows: regions I and II = aromatic protein I and II respectively; region III = fulvic acid-like; region IV = soluble microbial by-products; region V = humic acid-like. Peaks for humic acid and fulvic acid-like matter dominated in all EEMs throughout the growth cycle (Figure 4). The transition from early to mid phase revealed an increase in aromatic proteins and fulvic acid-like regions and may be attributed to the slight increase in DOC from 5.86 to 6.23 mg L\textsuperscript{-1}. The transition from mid to late phase showed an increase in all regions and was accompanied by a drastic rise in DOC from 6.23 to 18.4 mg L\textsuperscript{-1}. These observations are consistent with a study by Stedmon & Markager (2005) on marine algae in experimental enclosures.

**CONCLUSIONS**

In the early growth phase, the presence of \textit{M. aeruginosa} in the biologically treated effluent had negligible impact on flux profile and permeate volume; full flux recovery was achieved with the low alum dose of 1 mg Al\textsuperscript{3+} L\textsuperscript{-1} and comparable results were achieved for ACH treatment.

As the algal population entered the mid and late growth phase, algal cell and DOC concentration (mainly humic acid- and fulvic acid-like materials) increased, leading to poorer flux profiles and lower permeate volumes. Alum coagulation led to greater improvement in flux profile, permeate volume and membrane recovery compared with ACH coagulation. The poorer performance of ACH was attributed to the differing nature of the flocs which developed a higher specific resistance cake layer than alum.

Although chemical coagulation (with alum or ACH) improved the flux rate, water quality and permeate volume during the late growth phase, a higher coagulant concentration was necessary to achieve a satisfactory amount of permeate, but this led to a severe drop in flux recovery. With an increased permeate volume came an increased quantity of deposit on the membrane. At high alum concentrations the deposit formed a cement-like layer which resisted hydraulic cleaning. Hence, it is recommended that treatment of \textit{M. aeruginosa} blooms in wastewater treatment lagoons be conducted during the early and mid growth phase.

** ACKNOWLEDGEMENT**

The authors would like to thank Water Quality Research Australia (WQRA) for their financial support for this project.

**REFERENCES**


First received 4 August 2010; accepted in revised form 3 November 2010