Solving algae problems: French expertise and world-wide applications

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ABSTRACT: This paper reviews the various methods available for removing planktonic microalgae (microstraining, direct filtration, sedimentation, flotation, polishing using ozonation and granular activated carbon [O3+GAC], membrane filtration), and discusses their comparative effectiveness, optimisation and limitations. Also described are the treatments considered most effective in the removal of odorous and/or toxic metabolites. In each case French technology and its world-wide applications are compared to those documented in the literature. The article concludes with recommendations on the most appropriate processes for treating eutrophic waters.

INTRODUCTION

The global increase in the number of reservoirs and the worsening of eutrophication in natural and artificial lakes, and even rivers, are a source of growing concern among water treatment engineers and drinking water utilities in terms of the water-quality problems that stem from algal proliferation. These problems fall into two categories:

(i) The presence of biomass itself. This is particularly problematic in plant operations as a result of an abundance of planktonic algae (reagent consumption, sludge rise in sedimentation units, accelerated clogging of filters, etc.) and has been known to exist for a long time [1,2]. Moreover, the residual algae in treated water can be the source of a wide range of problems, including the consumption of residual chlorine and dissolved oxygen and the production of: trihalomethanes (THM) [3,4]; objectionable odours as the algae decay; assimilable organic carbon (AOC) for the growth of the biofilm; and the supply of particulate food matter to microinvertebrates liable to colonise the distribution system [5]. For these reasons, the highest possible degree of removal of microalgae must be achieved. In the absence of a precise international standard, and given that other than membranes, no conventional treatment process will afford 100% algae removal, engineers and operators must be thoroughly knowledgeable about the levels of efficiency of available treatment processes in order to choose the technology best suited to the raw water characteristics and the quality objectives defined. The first part of this paper should assist by describing the removal efficiency that can be anticipated in each case (Table 4).

(ii) The release into the water (either into the natural medium, treatment flow or distribution system) of undesirable metabolites. Although this classification is not exhaustive, two main categories of substances must be distinguished, both of which are derived from the activity of blue-green algae or Cyanophyceae, now known as Cyanobacteria [6] (a name change that, moreover, has not been recognised by all algae specialists [A. Couté, Personal communication (1995), quoting among others: Prochloron, a Microbial Enigma, eds R.A. Lewin & L. Cheng. Chapman & Hall, 1989], since certain prokaryotic species were also discovered among the green algae):

- substances that cause objectionable tastes and odours [7–9], e.g. geosmin and 2-methyl-isoborneol (2-MIB);
- toxins [6] liable to affect the liver (hepatotoxins) or the nervous system (neurotoxins).

Removal of these metabolites will be examined below, concluding with the most appropriate treatment lines for making eutrophic water potable.

METHODS AND MATERIALS

Microalgae counts

In various cases, microalgae have been counted directly under the microscope between the slide and the cover, with or without preliminary centrifugation [10]; or following filtration on a 0.45 μm membrane; or using the inverted microscope method. Algae are reported either as a number of organisms or as areal standard units (asu) per mL.

Tastes and odours

The level of tastes and odours is estimated using the Threshold Odour Number (TON) test, following successive dilutions. Gas chromatography–mass spectroscopy (GC-MS) in combination with two concentration techniques—Closed Loop Stripping Analysis (CLSA) and Steam Distillation Extraction (SDE)—are used as identification techniques [9].

Algal toxins

Hepatotoxins

Reverse phase high-performance liquid chromatography (HPLC) followed by UV detection [11]. For low concentra-
tions, the protein–phosphatase method is used [GA Codd, Personal Communication 1993; mentioned in Fawell [11].

Neurotoxins

REMOVAL OF PLANKTONIC ALGAE
Microstraining
Earlier studies [13–16] have all published similar results: using fabrics of woven stainless steel or polyester wires with apertures ranging from 15 to 45 μm (usually 30–35 μm), the overall rate of algae removal achieved by microstraining is usually between 40 and 70% (with a simultaneous reduction in turbidity of only 5–20%). Even by considerably reducing the aperture size (to 6 μm or even 1 μm) [17], the algal biomass of an aerated lagoon effluent could be reduced by no more than 55%.

Better results have been published by other authors, but these concerned either phytoplanktonic populations consisting essentially of larger species [18–20], or the net plankton, defined as ‘those algae of sufficient size to be retained on passing the water through no. 20 bolting silk’ [21], in contrast to total phytoplankton.

One of the authors has conducted tests in various parts of the world using fabrics ranging in aperture from 25 to 35 μm, and arrived at the following average rates of algae removal:
- 40% for Nile River water in Cairo;
- 55% for Seine River water in Paris;
- 50–65% for water from various lakes and impoundments.

It is noteworthy, too, that the algae removal rates are very scattered (between 10 and 100%) depending on the species (the primary factor being the size of the organisms). According to the authors’ studies, removal rates for various species can be estimated as follows:

For Diatoms, removal rates average from 40 to 80%, consisting of:
- Cyclotella (unicellular) 10–70%;
- Stephanodiscus (unicellular) 10–60%;
- Melosira (filamentous) 80–90%;
- Synedra (unicellular) 40–90%;
- Asterionella (colonial) 75–100%;
- Fragilaria (filamentous) 85–100%.

For Chlorophyceae, removal rates average from 50 to 60%, again with considerable differences from one species to another, for example:
- Chlorella (unicellular) 10–50%;
- Scenedesmus (cenobia with 4 or 8 cells) 15–60%;
- Pediastrum (cenobia with 4–64 cells) 80–95%.

For blue-green algae, removal rates average from 45 to 75%, for example:
- Oscillatoria (filamentous) 40–50%;
- Anabaena (filamentous) 50–70%.

It comes as no surprise that the smallest species are the most difficult to remove (sometimes only 10%) whereas they account for most of the coagulant demand (indeed, coagulation is a phenomenon concerning surface area, and the smallest organisms represent the highest ratio of developed surface area to volume). This, compounded by the low reduction in colloidal turbidity, virtually prevents microstraining from achieving a notable reduction in subsequent coagulant consumption, as illustrated by the results of two jar-tests performed on Nile River water in Egypt (Fig. 1).

Thus, microstraining may be viewed as a useful process for the removal of larger organisms (filamentous or colonial algae; zooplankton), but it does not stop smaller species or reproductive forms. Therefore, although microstraining can be useful in certain cases as a pretreatment prior to direct filtration (slow or rapid) [14,18–22], it cannot constitute a complete treatment or an economical pretreatment ahead of a complete flocculation–settling–filtration line. This had already been observed by other authors [15,23].

Fig. 1 Results of jar test on the Nile River water at Cairo, with and without microstraining.

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Direct filtration

Efficiency and limitations
Without chemicals, only slow filtration is likely to achieve algae removal in the order of 99% (2-log removal). Using rapid filtration without oxidants or coagulants, the authors obtained results in line with those published elsewhere [24] namely, on sand or dual-media filters. Algae reduction ranged from 10 to 75% depending on the species, with an average of ~ 50% (varying according to the particle-size of the filter beds and the filtration rate). Typical orders of magnitude were:

- Green algae *Ankistrodesmus*: 10%;
- Chlorophyta *Chlorella*, *Scenedesmus*: 20–40%;
- Chlamydomonas: 50%;
- Euglenophyta *Euglena*: 50%;
- Diatoms *Melosira*, *Asterionella*: 65–80%;
- Fragilaria: 75%.

When this method of treatment is applied under the best possible conditions—namely, prechlorination, use of a coagulant and a flocculant, optimised filtration—the removal rate achieved averages 95% [14,15], sometimes much lower [25]. The authors obtained similar results on water from various European sources, using the following treatment parameters:

- Reagents: preoxidant, coagulant, flocculant where needed;
- Filter media: either sand, ES = 0.95 mm, or dual-media (anthracite 1.6–2.5 mm + sand, ES = 0.75 mm);
- Filtration rate: between 5 and 8 m/h.

Using dual coagulants (inorganic salt and cationic polymer) further improves algae removal [26], however, to achieve 98–99% removal of algae, preoxidation is almost always necessary [26,27]. When the use of prechlorination was abandoned (a wise decision when dealing with algae-rich waters, as these algae are also THM precursors [3,4]), preozonation was already known to yield excellent results in algae removal by filtration [28], and this efficiency has been recently confirmed [27]. Tests conducted at a plant in southern France demonstrated that the treatment combining hydrogen peroxide with ozone (in this case in a ratio of 0.4:1 by weight) led to a spectacular improvement in performance (Fig. 2) resulting in 99–99.35% (2–2.2 log) removal, vs. 93% (1.155 log) without ozone and 95.3% (1.33 log) using ozone alone (possible water quality problems linked to preoxidation will be discussed further).

In the course of these studies, it was noted that when direct filtration was applied to water whose colour and turbidity were not subject to sharp variations through the year, the duration of the filter run could be expressed as a function of the algal biomass by an equation in the form:

\[ T = T_0 \times (1 + aA^b)^{-1} \]

where: \( T \) = actual filter run (h), \( T_0 \) = time duration of filter run when no or negligible algae are present, \( a \) and \( b \) = coefficients, \( A \) = algae as asu/mL.

Figure 3 shows an example of how this equation can provide a fairly good approximation of results previously published by Hutchinson & Foley [29], taking as the numerical values: \( T_0 = 35 \text{ h}, a = 1.77 \times 10^{-3}, b = 0.98 \).

The numerical coefficients must obviously be determined for each concrete case based on the type of algal population, filtration parameters, etc. A similar relation can be established according to the number of algae or cells contained in each mL of raw water.

The results obtained in the course of these studies led to the conclusion that the filter runs become unacceptably short when the algae content in the raw water exceeds 1000 asu/mL, using 0.9 mm sand or anthracite; or 2500 asu/mL, using 1.5 mm anthracite in dual-media filters. This conclusion is in accordance with the observations of other authors [29–31].

Thus, according to the experience of the authors, direct filtration on single- or dual-media filters is generally not suitable for the treatment of water with a high algae content, at least when a high quality product is requested. More sophisticated multimedia filters with possible application of microstrainers and/or flocculators as initial treatment steps [22], may be used to treat such waters; for instance, the phosphorus elimination plant of Wahnbachtalsperrenverband, consisting of Fe(III) coagulation and flocculation steps followed by 10 three-layer filters (3.3 mm ES GAC + 1.73 mm ES anthracite + 0.87 mm ES quartz sand) backwashed at a velocity of ≥ 70 m/h, can remove about 95% of the chlorophyll (from 25.15 µg/L in raw water to 1.28 µg/L in filtered water on average) and up to 99.9% of the plankton cells (max. 10⁶ cells/mL) [32]; however, it must be pointed out that this plant was designed to treat the maximum flow of a river in order to protect an impounding reservoir situated downstream: consequently, it often works below its maximum capacity and does not directly supply a drinking water network.
Therefore, any project of treatment of an algae-rich water by means of direct filtration involves previous pilot scale investigations in order to estimate the technical and economical advantages of this choice in each case. Generally, a conventional treatment line, including coagulation, flocculation, settling or flotation, and filtration, is preferred to treat algae-rich waters. However, algae removal is somewhat more delicate than turbidity removal and, consequently, greater attention is required when selecting technology and adjusting the chemical treatment.

Selection of clarifier type

Earlier articles have already brought to light the fact that algae removal is much less effective in static settlers [33,34] than in sludge blanket-type clarifiers [35].

The same conclusion emerged from the use of Upflow Pulsed Sludge Blanket Clarifiers [36] (UPSBC) (Pulsator) whose principle can even be applied in combination with that of plate- or tube-type lamellar settling (Superspulsator, Pulsatube, Ultra-pulsator) in treating algae-rich water anywhere in the world. For this application, such combined units have also proven superior to lamellar (plate or tube) clarifiers not equipped with sludge contact [37,38].

As early as the beginning of the 1960s, pilot studies made by one of the authors demonstrated the operating efficiency of UPSBCs for the removal of algae from Seine River water. It was soon observed that a consistent 95–99% reduction in the total phytoplankton population could be achieved on clarified water, and virtually complete removal could be achieved by adding a subsequent sand filtration step. As was the case for turbidity and colour removal, dual-media filtration was not necessary, provided clarification was performed under optimised treatment conditions (see below).

The superiority of the UPSBC over the static settling process has also been verified in warm climates, not only as regards rise rate, but also in terms of clarifier effluent quality and reagent consumption.

For example, on water from Laguna De Bay (Philippines), which contained 50 000–100 000 algae per mL (with 90–95% Cyanobacteria: Microcystis, Anabaenea, Anabaenopsis, Oscillatoria, Lyngbya, etc.), a flocculation test pointed to no better than a 90% reduction in algae content using static settling, whereas a UPSBC pilot unit achieved a reduction of 95–98%, and up to 99.5% with a well-adjusted treatment (7 g/m³ chlorine; 50 g/m³ aluminium sulphate; 0.1 g/m³ of powdered anionic polyelectrolyte).

Other comparative tests were conducted on water from a lake in tropical Africa, where counts showed an average of 2500 filaments of Anabaenae and 2500 colonies of Microcystis per mL of water, plus about 1000 of various Diatoms (Melosira and Cyclotella in particular). Treating this water required either 60–70 g/m³ aluminium sulphate alone, or 40–50 g/m³ of aluminium sulphate, together with the amount of sulphuric acid necessary to bring the pH to ≈6.2. At the industrial-scale plant, the UPSBC was compared with an upflow hopper bottom clarifier (UHBC), which may be considered as a heterogeneous, sludge blanket static settler. Results are shown in Table 1.

In Egypt, numerous UPSBCs are now in service in Cairo and Alexandria for the treatment of Nile River water, which has contained algae ever since the construction of the Aswan High Dam. In Alexandria, a majority of these units are in fact upgrades for older settling tanks, thus giving the Water

![Fig. 3](image-url) Filter runs as a function of algae concentration in raw water (based on Hutchinson & Foley [29]). Media: 30 cm 0.5-mm sand, 30 cm 0.98-mm coal; filtrations rate: 8.8 m/h; alum: <8 p.p.m.; raw water turbidity: <4 Ftu; effluent turbidity: <0.5 Ftu.

\[
T = 35 \left[ 1 + 1.77 \times 10^{-3} (A)^{0.98} \right]^{-1}
\]

Table 1 Comparison of static and pulsed sludge blanket clarifiers at Harare (Zimbabwe)

<table>
<thead>
<tr>
<th>Test no.</th>
<th>Unit</th>
<th>Actual rise rate</th>
<th>Flocculant (activated silica)</th>
<th>pH adjustment (H₂SO₄)</th>
<th>Plankton % reduction after settling</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>UPSBC</td>
<td>3.3 m/ h</td>
<td>no</td>
<td>no</td>
<td>99.5</td>
</tr>
<tr>
<td></td>
<td>UHBC</td>
<td>1.35 m/ h</td>
<td>yes (2 g/m³)</td>
<td>yes</td>
<td>98.7</td>
</tr>
<tr>
<td>2</td>
<td>UPSBC</td>
<td>4.1 m/ h</td>
<td>no</td>
<td>no</td>
<td>96.7</td>
</tr>
<tr>
<td></td>
<td>UHBC</td>
<td>1.1 m/ h</td>
<td>yes (2 g/m³)</td>
<td>no</td>
<td>95.9</td>
</tr>
</tbody>
</table>

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Authority the opportunity to appreciate the advantages of this new technology:
● a 2–2.5-fold increase in production per unit of surface area (with improved clarifier effluent quality);
● a 15–45% reduction in coagulant consumption and a 15–35% reduction in chlorine consumption.
The UPSBC’s efficiency in removing algae may be explained both by the fluidised filter action of the sludge blanket and by the extended contact time between the water upflow and the sludge blanket (about 3/5 of the total retention time inside the unit). The beneficial effect of a longer flocculation time has already been observed [39] and the authors have confirmed it on various other occasions, in particular on Nile River water as indicated in the laboratory test shown in Table 2.

### Adjusting the treatment

No matter how sophisticated the clarification technology, optimal algae removal cannot be achieved, as already outlined by other authors [26,40,41], unless the chemical treatment of the water has been optimised as well, namely:

<table>
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<tr>
<th>Satisfaction of coagulant demand</th>
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| Minimal turbidity as measured in a jar-test is no longer a sufficient criterion. The algal cells are negatively charged (like all suspended colloidal particles) and must be completely destabilised by charge neutralisation to allow the treatment to reach its maximal efficiency; the electrophoretic mobility of planktonic algae, like for inert particles, is usually measured by means of a zetameter, the result being expressed as zeta potential ($pZ$); the treatment is optimised when the algal cells no longer present any electrophoretic mobility, in other words when the $pZ$ reaches a value near zero: this corresponds to the maximum removal of microalgae (see example in Fig. 4, concerning a test conducted on Nile River water in Cairo). Other authors [42,43] have also noted the importance of neutralising the charge of microalgae (possibly using a streaming current detector (SCD) for an on-line control [44,45], although some inconsistencies have sometimes been reported between a complete $pZ$ neutralisation and the SCD results [46]).

If the coagulant dosage is too low and the raw water contains Cyanobacteria, the latter are the last to be removed (clearly shown in Fig. 4) and therefore account for the major portion of residual algae in the treated water. There have been several occasions involving waters from France and Africa, when cyanobacteria were observed to account for the following proportions of total phytoplankton:
● raw water: 10–50%;
● settled water: 60–90%;
● filtered water: 85–100%.

<table>
<thead>
<tr>
<th>Adjustment of the flocculation pH</th>
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</table>
| For a given coagulant dosage, there is generally an optimal pH at which algae removal is at its maximum rate. It stands to reason that a change in the pH (generally in the direction of acidification) can sometimes permit savings on coagulant. For this reason, in the example of the Harare plant (Zimbabwe) mentioned above, the treatment includes an injection of $H_2SO_4$. Without this additional chemical, the coagulant uptake

<table>
<thead>
<tr>
<th>Table 2 Influence of flocculation time on algae removal (jar test at Cairo, Egypt)</th>
</tr>
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<tbody>
<tr>
<td>Treatment:</td>
</tr>
<tr>
<td>Flocculation time (min)</td>
</tr>
<tr>
<td>Residual algae in settled water (per mL)</td>
</tr>
<tr>
<td>Percent removal (raw water: 11 500 per mL)</td>
</tr>
</tbody>
</table>

[Fig. 4 The Nile water in Cairo. Residual phytoplankton as a function of alum dosage; comparison with the evolution of Zeta potential and turbidity (jar-test study).]
depended not only on the number of algae, but also on the alkalinity of the water, due to its influence on the pH. A simple correlation has been established between the alkalinity (Alk, as p.p.m. CaCO₃) and number of algae \( N \) (per mL) in this raw water, and the dosage \( D \) (in g/m³) of coagulant to be used. The results are presented in Fig. 5 and can be expressed by the following formula:

\[
D = a + b \log(N)
\]

and more precisely, in this case:

\[
D = 30 + 2(\text{Alk} - 15) \log(N \times 10^{-3})
\]

for \( N > 10^3 \) algae/mL, in the alkalinity range of 30–70 p.p.m. CaCO₃.

When the amount of coagulant was well adjusted, the UPSBC removed 97–99.5% of the algae, before filtration.

**Use of a polymer, coagulant (instead of or in addition to the inorganic coagulant) and/or flocculant**

In particular, when the coagulant demand is too high, a cationic polymer (when such products are authorised for potable water treatment) can be efficiently associated with the inorganic coagulant [41], as already mentioned in the case of a direct filtration [26].

These various principles illustrate that enhanced coagulation, now recommended for the removal of dissolved natural organic matter (NOM) [47], has long been applied in France for algae removal. However, the process still requires an effective preoxidation step, especially if the water has a high green algae content.

**Using chlorine as a preoxidant**

Breakpoint chlorination has long been the process conventionally used for the purpose of preoxidation [15,23,35]. Figure 6 shows the efficiency of this pretreatment step (industrial tests performed on UPSBCs treating water from the Vistula River in Warsaw, Poland). The raw water contained between 10 000 and 100 000 algae per mL, consisting of Diatoms (1/3) and Chlorophyceae (2/3). Figure 6 shows that the rate of algae removal by clarification (population counts in clarified water prior to filtration) averages 85% in the absence of chlorine and more than 99% when prechlorination is carried out in combination with a well-adjusted coagulant dosage. These results also show that when the parameters of physical–chemical treatment are not optimum, the removal rate is low mainly for the Chlorophyta (green algae); Diatom removal from clarified water is rarely less than 90%.

The consequences of a lack of preoxidation combined with inadequate coagulant treatment are illustrated by another
example which involves taking Seine River water upstream of Paris. Figure 7 summarises the observations made during periods of considerable phytoplankton proliferation in the Seine in late summer and early autumn. The raw water contained between 10,000 and 30,000 algae per mL and the dominant species were the Chlorophyceae *Pediastrum* (mainly *P. clathratum*) and the Diatom *Melosira*. The efficiency of two UPSBCs operating in parallel for the removal of these algae has been studied. When the study began on 28 August, breakpoint prechlorination was applied in UPSBC no. 1 only; the coagulant dosage was defined such that it would allow acceptable flocculation and residual turbidity (40–50 g/m³), rather than being geared to neutralisation of the Zeta potential (80–90 g/m³). These dosages were determined through a combination of electrophoretic measurements using a Zeta-meter, and flocculation tests using a laboratory flocculator (Fig. 8 shows a model of the results).

Under these conditions, algae removal was only 70–80% without chlorine, and 85% with chlorine, as can be seen on the left side of Fig. 7. During the ensuing days, the alum dosage was increased gradually to the value which neutralised the Zeta potential. Beginning on 4 September, the treatment was constantly set for \( pZ = 0 \). Beginning on the same date, the following average reductions were observed:

- 98–99% removal (i.e. 1–2% residual algae in the settled water) in UPSBC no. 1, with breakpoint prechlorination;
- 85% removal (i.e. 15% residual algae in the settled water) in UPSBC no. 2, where no prechlorination was applied.

The final phase of the experiment involved the application of breakpoint prechlorination to UPSBC no. 2 beginning on 2 October. After this step, the two clarifiers showed identical performance (as shown on the right side of Fig. 7).

Under optimised treatment conditions like those described above, microalgae removal rates can reach:

- 90–99% in clarified water (1–2 log);
- 99–99.9% in sand-filtered water (2–3 log).

In certain cases, dual- or multimedia filtration can optimise performance [14,15,48], although certain authors achieved better results following settling, using sand filters [23]. This is generally the solution adopted in France.

If the treatment plant is also equipped with a polishing step (ozonation + GAC filtration), the overall removal efficiency for the plant as a whole will be between 99.9% and 99.995% (3–4.3 log).

![Fig. 7 Seine River at Ivry—study of phytoplankton removal in two UPSBCs.](image)

![Fig. 8 Seine River at Ivry: August–September. Results of jar tests.](image)
substituting preozonation (1 g/m³ on average) for chlorination at the head of the plant, can sometimes yield the same result as chlorination when applied ahead of a conventional treatment line, e.g. on the Seine River water between Rouen and Le Havre, in optimised conditions: i.e. algal removal of 95–98% after settling and 99–99.8% after filtration. However, this technique may sometimes impart an unpleasant odour to the water, hence the need of previous testing before its application.

Whenever applicable, preozonation would be a better choice, although slightly less efficient than prechlorination in the removal of microalgae when no polishing stage is present; however, a main ozonation step, placed further in the treatment line (preferably between settling and filtration), would then be advisable, particularly if the water is organic-rich, in order to complete the oxidation reactions initialised in preozonation and minimise the ozonation by-products.

It must also be pointed out that some cyanobacterial metabolites, probably phenolic compounds, react with chlorine and form by-products developing a very unpleasant chlorophenol-like taste. As a result, prechlorination must be avoided in such cases: tests must be made to choose the best alternative chemical treatment (pH adjustment; preoxidation using ClO₂ or ozone, etc.).

When no preoxidant is available, the use of copper sulphate, injected at a low concentration (around 1 g/m³ as CuSO₄) at the inlet of the plant, can sometimes yield the same result as chlorine but at a lower efficiency (see below).

To conclude this discussion, it must be stressed that prechlorination is still very common world-wide, particularly in developing countries where it is the best answer not only to help microalgae removal, but also to improve all the parameters of water clarification, to keep clarifiers and filters clean, to get rid of ammoniacal nitrogen (in reasonable concentrations), to prepare a safe final disinfection by means of postchlorination, etc. However, the above observations show that its use must be carefully examined in the presence of high concentrations of algae, particularly when the latter are Cyanobacteria; indeed special precautions should be taken, e.g.:

- using GAC, in first or second stage of filtration;
- when this is not possible, using PAC in clarification after a sufficient contact time with chlorine; and
- if no adsorption process is available, chlorination below the breakpoint (in the chloramines range); the authors have found that in some occasions, prechloramination could give results almost similar to those of a breakpoint chlorination when applied ahead of a conventional treatment line, e.g. on the Seine River water between Rouen and Le Havre, in optimised conditions: i.e. algal removal of 95–98% after settling and 99–99.8% after filtration. However, this technique may sometimes impart an unpleasant odour to the water, hence the need of previous testing before its application.

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Sedimentation or flotation? A case study

Dissolved air flotation (DAF) can be a valid alternative to sedimentation when water has a high algae content during the entire or part of the year but is rarely loaded with suspended solids [40,41,58–61]. The Moulle treatment plant in northern France (near Dunkirk) is an interesting example. The plant receives raw water from a very eutrophic river cutoff and is noteworthy for two purposes (Fig. 9):

- to compare two different lines, one of which includes a sedimentation process (1st stage) and the other a flotation process (2nd stage);
- to demonstrate the two possible uses of flotation: in water treatment (algae removal) in the 2nd stage, and in thickening the sludge produced by the 1st stage sedimentation for subsequent dewatering in filter-presses (in contrast, the sludge from the main flotation process selected for the extension is sufficiently concentrated to be pressed directly without intermediate thickening).

At this plant, flotation was found to have two main advantages over sedimentation:

- Coagulant dosage (here, chlorinated copperas) is reduced by 20–40% yielding the same result in both lines (Fig. 10), i.e. 1000–1200 algae per mL on average before filtration and about 10 algae per mL after filtration for both ‘floated’ and clarified water (Raw water algae content: 30 000–50 000 algae per mL);
- Higher solids content of sludge: In the process ‘floated’ sludge has a 25–30 g/L dry solids (DS) concentration, almost 10 times the DS content of ‘clarified’ sludge. This explains why the sludge formed in DAF requires no intermediate thickening before the filter-press. During the thickening of sludge (from the clarifier): the DS concentration increases from 3 to 25 g/L on average. With a static thickener, the required surface area would have been 10 times larger.

Moreover, a comparison between the operating costs of both treatment lines demonstrates that although the two processes are almost equivalent from the standpoint of water treatment only, the use of flotation may reduce operating costs by 10–15%, when sludge treatment is also taken into account.

Thus, DAF has now become a solid competitor to sedimentation—even when the most efficient clarification units are involved—for algae removal applications. In many cases, it is worthwhile to compare these two alternatives, especially since flotation is less sensitive than sedimentation to the type of preoxidation treatment. In other words, whether prechlorination or preozonation is used, algae removal efficiency can be expected to range from 90 to 99% following flotation, and between 99 and 99.9% after sand filtration. Without preoxidation, these figures are 80–95% and 96–99%, respectively, provided in all cases that coagulant application is optimised, as also recommended by other authors [40,41,62].

The advantages of DAF are therefore undeniable, but the treatment is subject nevertheless to certain limitations:

- raw water must not show any periods of high turbidity; and
- the DAF unit must be provided with a stock of spare parts and maintained by qualified personnel, since any interruption in the pressurisation circuit results in an interruption of the treatment itself.

Fig. 9 Flow-sheet of the Moulle Plant.

Fig. 10 Algae removal at Moulle Plant.
Toward new technologies?

The conclusion to be drawn from the above is that modern treatment processes can achieve a very high degree of algae removal, yet do not constitute an absolute barrier. Complete removal can only be achieved by means of membrane processes like microfiltration or ultrafiltration, now key growth technologies. Yet even using these processes, operators must realise that an extremely slight risk still remains, which is related to the seals or the possible rupture of a fibre. Nevertheless, membrane processes may be deemed to offer algae removal efficiency of better than 99.9999% (6-log) under any circumstances, ranking them two orders of magnitude above any other treatment (see Table 4, where removal rates are also reported as Log-Removal rates.). Although the capital and operating costs are still greater in many cases, this is nonetheless a viable investment in view of the water quality and added safety for consumers, since membranes also offer a virtually absolute barrier against bacteria, Giardia cysts, Cryptosporidium oocysts, etc., whereas they do not increase the risk of metabolites release since the cells observed in the concentrate are not damaged, even in the cross-flow mode [63,64].

When seeking complete algae removal, the most promising solution for the treatment of algae-rich waters may lie in a combination of conventional and membrane processes. This was the successful option taken by Chevalier et al. [65] in treating raw water with an algae content of 200 000 algae per mL. The treatment line consisted of DAF/filtration/ultrafiltration (UF).

In the future, the technological choice will depend both on quality objectives and the regulatory framework. Table 4 gives guidelines for making such a choice. If the goal is a few residual algae per mL of water, then conventional treatment lines which include one or more granular media will consistently provide excellent results provided the treatment is well designed and adjusted. On the other hand, a tolerance restricted to between 1 and 10 algae per litre would inevitably call for the use of membranes, as is the case at the Apier/Saint-Cassien plant (27 000 m3/day) near Cannes (France), commissioned in 1995. To treat the impounded water containing algae levels of up to 5000 algae/mL, the first phase of the installation is equipped with eight banks of 20 ultrafiltration modules (Aquasource, Rueil-Malmaison, France) for a total of 160 modules having a unit filter surface area of 50 m2. At the start of the next century, the capacity of the plant will have quadrupled to total 108 000 m3/day.

TASTE AND ODOR REMOVAL

As the effectiveness of O3 and/or GAC as a polishing step is already well documented [9], only more recently developed techniques are mentioned herein, namely:

- The [O3 + H2O2] (Per ozone Process, Degrémont) combination which, by producing free radicals (OH) is highly effective in breaking down geosmin and 2-MIB, where the action of ozone alone has proven incomplete [66,67]. As this process is also effective for the removal of many other trace organic contaminants, it is now widely used in France preferably upstream of sand or GAC filtration, for removing AOC and producing a biologically stable treated water; furthermore, the microorganisms that attach to the GAC can break down the residual geosmin and 2-MIB [68] thus complementing the effects of oxidation. Table 5 gives an example of the efficiency of this process in treating a malodorous water from an impoundment in Iran. As this type of radical oxidation treatment is incompatible with efficient disinfection of the water, it can be performed in a contact tank (patented) consisting of three compartments, allowing for sequential disinfection and trace contaminant removal (Fig. 11).

- Optimisation of the use of PAC which, despite the good efficiency sometimes demonstrated at low dosages [69] and a
higher removal rate when used in a UPSBC [70], generally requires a high treatment dosage in application to raw water in the presence of a coagulant. The need to obtain treated water free of both microorganisms and odour-producing metabolites led to the [PAC + UF] (Cristal Process, Lyonnaise des Eaux [Nanterre], Degrémont and Aquasource [Rueil-Malmaison], France) combination which involves an injection of PAC ahead of an ultrafiltration unit operating in the cross-flow mode [71,72]. This process can be applied directly to raw water as is the case at the Saint-Cassien plant discussed above. It can also be preceded by preozonation or even used as a polishing treatment following a conventional process of sedimentation and ozonation. The latter treatment is implemented at the Vigneux plant (capacity: 55 000 m³/day) which treats Seine River water upstream from Paris: the PAC discharged with the concentrate is returned to the raw water at the head of the plant so it can complete its adsorption function within the sludge blanket of the UPSBC (see Fig. 12), thereby minimising PAC costs. This solution was adopted following testing on pilot units, which demonstrated it to be more reliable than a conventional solution.

### REMOVAL OF ALGAL TOXINS

The toxins produced by blue-green algae may be subdivided into two groups according to the way in which they act [6,73]:

- **Hepatotoxins** are primarily peptides (chains of amino acids). The most commonly identified compounds in this group are microcystins, released by *Microcystis aeruginosa*, and nodularin, produced by *Nodularia spumigena*. Depending on the ingested dosage, the irreversible damage these toxins cause to the liver can lead either to rapid death or to long-term pathologies such as liver cancer. They can also cause temporary symptoms such as gastro-enteritis, vomiting, liver trouble or even dermatosis.

- **Neurotoxins** are alkaloids such as the anatoxins released by *Anabaena flos aquae*. These substances can cause very rapid death by paralysing the respiratory and cardiac muscles.

Although an active secretion of these metabolites cannot be excluded, their presence in the natural environment is generally due to a release caused by the rupture of algal cells. This phenomenon may arise several times in the water management cycle, e.g.: an algicide treatment of the natural medium [74], pumping and conveyance of raw water [75], the e/C128ect of certain substances used in the treatment [54,55].

The following conclusions have emerged from studies on the removal of algal toxins published by other authors:

1. Inadequacy of conventional treatment processes [12,76–79];
2. Oxidation:
   - Ineffectiveness of chloramines and hydrogen peroxide [80]
   - Effectiveness of chlorine, but only at a dosage that yields a residual of between 0.5 and 1 mg/L after 1 h [80,81];
   - Great effectiveness of ozone, even under the restricted conditions of preozonation at a dosage of 1–2 g/m³ with no observable residual [11,12,78,79].
3. Activated carbon adsorption:
   - Powdered Activated Carbon (PAC): good treatment efficiency [12] although PAC demand can reach between 50

### Table 5


<table>
<thead>
<tr>
<th>Ozone dosage (mg/L)</th>
<th>TON for an H2O2/O3 ratio of:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>6</td>
</tr>
<tr>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>3.5</td>
<td>3.5</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
</tr>
</tbody>
</table>

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and 100 g/m³ before satisfactory removal is achieved [11,77,81];
- Granular Activated Carbon (GAC): a 90–100% removal rate can be achieved at an empty bed contact time (EBCT) on the order of 10 min [12,76,78,79,81] but saturation is reached after a few months [77].

The studies described below were aimed at confirming the bibliographical data and clarifying certain points, particularly the influence of NOM—as estimated by the measurement of total organic carbon (TOC)—which is generally found along with algal toxins in eutrophic waters. These studies were conducted within the framework of a joint French-Australian research programme (FAIR) with the participation of the Centre de Recherche Lyonnaise des Eaux-Degrémont (CIRSEE) and the Australian Center for Water Quality Research (ACWQR); they were already described in a comprehensive report [82] and summarised in previous papers [83,84]. Key results may be presented as follows.

Oxidation using ozone

Table 6 shows the experimental data. Figures 13 and 14 illustrate more precisely the effect of NOM on the removal of microcystin-LR by ozone.

The results thus confirm the effectiveness of ozone. However, as the removal efficiency tends to decline when toxins and NOM are simultaneously present in high concentrations, it is advisable to check the efficiency of ozonation (pre and/or postozonation) for each particular case.

Adsorption on PAC

The adsorption isotherms determined in ultrapure water using carbon with a high adsorption capacity indicate good adsorbability for these toxins (Fig. 15). The results are rather comparable to those obtained for trace contaminants such as atrazine. Accordingly, a removal rate of 99% is achieved for a PAC (F400) dosage of 1.5 g/m³ and an initial microcystin-LR concentration of 50 µg/L.

In contrast, in natural waters, adsorbability is diminished due to the NOM that competes with the algal toxins (see Fig. 16). PAC is introduced at the coagulation stage. Table 7 summarises the results of all tests performed.

In addition to the influence of NOM, these results highlight the importance of choosing the right type of PAC, supporting the conclusions already published by Donati et al. [85]. They also confirm the high level of PAC demand (50 g/m³ or higher) that results from a high concentration of toxins.

Adsorption on GAC

Pilot tests were carried out in Australia using F400 carbon at an EBCT of 7.5 min. The filters were supplied with filtered surface

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**Table 6** Destruction of algal toxins by means of ozonation

<table>
<thead>
<tr>
<th>Nature of the toxin</th>
<th>TOC (mg/L)</th>
<th>C₀ (µg/L)</th>
<th>Contact time</th>
<th>Residual O₃ (mg/L)</th>
<th>Percentage removal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microcystin-LR</td>
<td>0.3</td>
<td>100</td>
<td>30 s</td>
<td>0.1</td>
<td>100</td>
</tr>
<tr>
<td>Nodularin</td>
<td>3.5</td>
<td>500</td>
<td>8 min</td>
<td>0.2</td>
<td>99</td>
</tr>
<tr>
<td>Anatoxin-a</td>
<td>0.3</td>
<td>500</td>
<td>10 min</td>
<td>0.5</td>
<td>50</td>
</tr>
</tbody>
</table>

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waters (DOC = 5.0–6.5 mg/L) spiked with microcystin-LR (30–50 µg/L). Results showed a removal of ≥ 90% for up to 7000 m³ of treated water/m³ of carbon in the first filter (Hope Valley water), and for up to 12 000 m³/m³ in the second filter (Morgan water).

After breakthrough in both filters, removal rates ranging from 49 to 63% were observed for an additional 10 000 m³/m³. Following confirmation, the differences between the results of these two tests were attributed to the biodegradation of the toxins.

Thus, these tests confirm the feasibility of applying GAC filtration to remove algal toxins. Indeed:

1. the development of toxins in water resources is generally an isolated occurrence. The adsorption capacities determined experimentally show that GAC can remove peak toxin concentrations (for example, 20 µg/L over a two-week period) provided the filter is not completely saturated.

2. between two toxin occurrences, removal seems to be completed by the biodegradation of the toxins within the filter. This phenomenon had already been observed in the natural environment [11] and amounts to a type of biological regeneration of the GAC, but it takes several weeks to become complete [74,86], being however, more rapid in the particulate living material compared to dissolved toxins [87].

Fig. 13 Oxidation of microcystin-LR by ozone in TOC-free water.

Fig. 14 Influence of NOM on the oxidation of microcystin-LR by ozone.
CONCLUSION

Algae removal

The effective removal of planktonic algae from eutrophic waters used for drinking water purposes generally entails a well-designed and well-operated treatment process. Although two-step direct (single- or multimedia) filtration plants have been recommended in some particular cases [88], the most reliable combination, according to the experience of the authors, includes preoxidation, enhanced coagulation, separation using high-performance settling or DAF followed by sand or multimedia filtration.

In preoxidation, ozone should be preferred to chlorine whenever possible. The efficiency of preozonation is similar to that of prechlorination in a flotation-filtration treatment line, but slightly lower in a settling-filtration treatment line (although it dramatically improves the result compared to a treatment without preoxidation).

The choice between DAF (possibly associated with preozonation [57]) and settling (preferably sludge blanket clarifiers in this case) depends on the type of raw water.
and the local conditions. On the other hand, the choice of a pulsed sludge blanket clarifier can be particularly recommended when the treatment includes metabolite adsorption on PAC.

The efficiency of the clarification treatment can be supplemented by polishing using [O3+GAC] if 3–4 log removal is the goal. To obtain water that is absolutely free of microalgae, micro or ultrafiltration membrane processes are required, alone or following conventional clarification or DAF, depending on the raw water quality.

Table 4 summarises the compared efficiencies of the different processes, as far as planktonic algae removal is concerned.

### Removal of algal metabolites

The best technologies for the removal of odorous or toxic metabolites revolve around the use of ozone (possibly combined with \( \text{H}_2\text{O}_2 \)) and/or GAC, the combined \([\text{O}_3+\text{GAC}]\) treatment being mandatory in difficult cases (especially in water which is permanently eutrophic in nature, notably in warm climates, and/or which is polluted). GAC alone or PAC can be used successfully in cases where problems are of a transitory nature (although a main ozonation step, inserted upstream of the filtration step(s), should now be the rule when the treatment line already includes preozonation).

Among the new techniques, the \([\text{PAC}+\text{UF}]\) process, applied either alone or following conventional sedimentation and/or ozonation, is certainly the most promising for the simultaneous removal of virtually all algae and their metabolites.

### ACKNOWLEDGEMENTS

The authors are indebted to François Bernazeau, Jean Gannier, Philippe Gislette and Pierre Pieronne, their colleagues, for providing some of the data presented in this paper.

### BIBLIOGRAPHY


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**Table 4** Adsorption of algal toxins on PAC (TOC in water: 4–6 mg/L). Influence of various parameters on PAC consumption

<table>
<thead>
<tr>
<th>Percentage removal</th>
<th>90% (from 10 to 1 µg/L)</th>
<th>98% (from 50 to 1 µg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type of PAC</td>
<td>Chemviron RB, Norit W20</td>
<td>Chemviron RB, Norit W20</td>
</tr>
<tr>
<td></td>
<td>Norit SA Super</td>
<td>Norit SA Super, F400</td>
</tr>
<tr>
<td></td>
<td>Ceca TE</td>
<td>Ceca TE</td>
</tr>
<tr>
<td>PAC demand (g/m³)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nodularin</td>
<td>2</td>
<td>9</td>
</tr>
<tr>
<td>Microcystin–LR</td>
<td>7–9</td>
<td>37–50</td>
</tr>
<tr>
<td>Anatoxin-a</td>
<td>11</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td></td>
<td>&gt; 80</td>
</tr>
</tbody>
</table>

Hazen R. Application of the microstrainer to water treatment in Great Britain. JAWWA 1953; 45(7): 723–734.


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