***MICROCYSTIS AERUGINOSA* REMOVAL BY DISSOLVED AIR FLOTATION (DAF)**

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Microcystis aeruginosa Removal by Dissolved Air Flotation (DAF) *Options for Enhanced Process Operation and Kinetic Modelling*

DISSERTATION

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Abstract

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Seven Dutch surface water treatment companies share the problem of periodical troublesome algal blooms in their raw water sources and reservoirs, and participated in this research : Amsterdam Water Supply (GWA), North Holland Water Supply Companies (PWN), Rhine-Kennemerland Water Transport Company (WRK), Dune Water Supply Company South Holland (DZH), Energy and Water Supply Company Rhineland (EWR), Water Supply Association Overijssel - Regio Oost (WMO, formerly Water Supply Company Oost-Twente - WOT), and Groningen Water Supply Company (GWG). KIWA Research and Consultancy also participated actively in the research.

The eutrophication status of the raw water utilised by the water treatment companies varies substantially. Phosphorus was found to be the limiting nutrient; concentrations of >0.05 mg P/L were recognised as potentially troublesome in terms of algal growth (>10 µg chlorophyll-*a*/L). The predominant algal species depend on a range of additional factors, including pH, temperature, combined ratio of other nutrients, light availability, etc. Different sets of problems are experienced due to the resultant seasonal algal population fluctuations (from 8.4 µg/L chlorophyll-*a* for Loenderveen Lake to 238 µg/L chlorophyll-*a* for the PWN reservoir, on a yearly average maximum basis). In all cases however, the periodical peak concentrations of algae cause substantial treatment problems. Cyanobacteria in general and the *Microcystis aeruginosa* and the *Oscillatoria agardhii* species in particular, proved to be the most persistent and associated with most treatment problems. The Dutch experiences are generally representative of industrialised countries where increased eutrophication results commonly in algal blooms and related treatment problems. These included an up to 100% higher coagulant dose, substantially increased filter backwash quantities, trihalomethane formation (during short periods of intermittent chlorination for mussels growth control), (surface) clogging of filters, passing of algae through treatment in objectionable quantities, increase of MFI (modified fouling index) and AOC (assimilable organic carbon) values, etc. The spherical *M. aeruginosa* single cells form is also representative of particles in the size range that are typically difficult to remove in treatment (3-10 µm). Its size and shape make it also a suitable representative of the pathogenic *Cryptosporidium* oocysts and *Giardia* cysts.

This study targeted the removal of the cyanobacteria *M. aeruginosa* by conventional and advanced treatment technologies, that are available or considered feasible under raw water quality and process circumstances that are typical for the Netherlands. The ultimate barrier preventing them, and their products, from penetrating into the distribution system is rapid or/and slow sand filtration. Pre-treatment modes practised in the involved companies include sedimentation-

filtration and more advanced dissolved air flotation (DAF)-filtration. The coagulation/flocculation process is of critical importance regardless of the down-stream treatment; however, the down-stream treatment itself is defined by the coagulation/flocculation. The option of algae conditioning by oxidants, e.g. ozone or KMnO_4 , although not actively practised in the Netherlands, was considered attractive to (periodically or continuously) increase the algae removal efficiency. Consequently, this study aimed to investigate a range of treatment options for the removal of *M. aeruginosa*, with an emphasis on DAF and different modes for its enhancement. The evaluation of process efficiency was to be complemented with insight into the process mechanisms, and to serve as a basis for the evaluation of process kinetics.

For this purpose two experimental modes were used: a bench-scale modified jar test filtration apparatus with facilities for both DAF and sedimentation, and a commercial DAF pilot plant (Purac, Sweden). The former utilised model water prepared by spiking water originating from the Biesbosch reservoirs (the Netherlands) with laboratory cultured *M. aeruginosa* to a standard initial concentration of $\approx 10,000 \pm \text{cells/mL}$. The standard experimental temperature was 20°C and coagulation G was $1,000 \text{ s}^{-1}$ for 30 s at pH 8. Other coagulation pH conditions were also tested (pH 4, 6, 7, 8, and 9). The flocculation G and time were varied ($G_f=10, 30, 50, 70, 100$ and 120 s^{-1} and $t_f=5, 10, 15, 25, 30$ and 35 min). The coagulant ($\text{FeCl}_3 \cdot 6 \text{ H}_2\text{O}$) doses were 0 - 15 mg Fe(III)/l , while the cationic polyelectrolytes Superfloc C-573 and Wisprofloc-P, the non ionic Wisprofloc-N, and the anionic Superfloc A-100 were tested as sole coagulants and as coagulant aids. Ozone was used as an algae conditioner in the dose range of 0.48-2.16 mg O_3/L , or 0.2-0.9 mg $\text{O}_3/\text{mg TOC}$ at pH 7.5. KMnO_4 conditioning was tested in the dose range of 0-2 mg/L at pH 8, the optimal dose being based on the visual determination technique. The KMnO_4 contact time was varied from 0-30 min. The DAF pilot plant was (operated at a flow of $Q=4\text{-}6 \text{ m}^3/\text{h}$) followed by a multi media rapid sand up-flow filtration ($Q=0.4 \text{ m}^3/\text{h}$, $v=10 \text{ m/h}$) and situated at the WRK III treatment plant in Andijk, the Netherlands. It was compared with the full-scale WRK III treatment line comprising of flocculation, lamella sedimentation and filtration, during the cyanobacteria blooms (primarily *M. aeruginosa*) occurring in the raw water reservoir. Analytical techniques for the assessment of process efficiency included turbidity, residual coagulant (Fe), TOC/DOC, UV absorbance at 254 nm, and residual Mn. They were supported by more advanced methodologies including bromate and MFI measurements, particle count (in the size range of $d_p > 2.75 \mu\text{m}$ and in a limited number of cases $d_p > 0.3 \mu\text{m}$) and computer image analysis ($d_p > 1.9 \mu\text{m}$). Coupled with the high resolution SEM (scanning electron microscopy), the latter provided valuable information for assessment of process mechanisms and kinetics.

DAF proved to be a viable and efficient alternative to sedimentation for the treatment of heavily algae (*M. aeruginosa*) laden water. It consistently resulted in equal or better algae removal efficiency than the sedimentation process (71% versus 87% for jar test conditions, and 96.3% versus 95.9% for pilot plant conditions), requiring at the same time an up to 50% lower coagulant dose than the sedimentation (3 mg Fe(III)/L versus 10 mg Fe(III)/L in the jar test conditions, and 7-12 g Fe(III)/m^3 versus 20-24 g Fe(III)/m^3 + 0.2-0.5 g Wisprofloc-P/ m^3 in the pilot plant conditions and the full scale sedimentation), which is significant from an environmental point of view. Also, the produced sludge was of high solids content. Other positive aspects included the relatively short flocculation time (however, not shorter than 15 min, compared to $>30 \text{ min}$ for sedimentation) and the low flocculation energy input requirement ($G=10 \text{ s}^{-1}$ compared to $G=30 \text{ s}^{-1}$ for sedimentation), as well as the high process loading rate (leading to 5-6 times overall lower space requirements). Furthermore, the DAF-filtration treatment scheme resulted consistently in

high (>2 log) algae removal efficiency. The relatively low coagulant demand for DAF, however, produced less efficient sweep coagulation conditions than in the case of the sedimentation, resulting in organo-Fe complexes formation and consequently higher DAF effluent Fe and turbidity residuals. This was overcome by coagulation at pH<IEP (iso-electrical point), suggesting the potential of using cationic polyelectrolyte coagulant aids. These improved the particle (algae) removal efficiency significantly in case of the model water experiments (20-40%), compared to a minor rise of only 1-2% in pilot plant experiments. This is owed to the promotion of particle (algae) adsorption coagulation based on charge neutralisation. Furthermore, the observed charge neutralisation phenomena resulted in improved particle-bubble attachment and hence more efficient DAF. Non-ionic and anionic polyelectrolytes (and the bridging mechanism) were found to be less efficient, partly because of the lower particle concentration (lower coagulant dose) and the negative charge of the air bubbles.

The particle (algae) removal efficiency the pilot plant achieved on reservoir water was by 20-30% higher than that in the jar tests with model water. The NOM (natural organic matter) concentration and composition was found to be the likely cause of the differences, especially as it is present in the form of organic matter meshes and fibrillar structures. Although the complexing of metal coagulants by NOM imparts additional coagulant demand, these structures may eventually promote flocculation, serving as sites for easier floc growth and embedment, as well as providing a gel-like structure that is easier to be removed by the rising air bubbles in DAF.

Relatively low doses of ozone (0.2-0.5 mg O₃/mg TOC, or 0.6-1.5 mg O₃ /L) as an algae conditioner under bench-scale conditions significantly (by 30-40%) increased DAF efficiency towards 2 log removal. Combined with cationic polyelectrolytes, the DAF process efficiency rose further typically by another 5%. Although not yet confirmed by pilot plant results with reservoir water, the application of the lower dose range of 0.2 mg O₃ /mg C at pH 7 resulted in bromate levels within the range of the 10 µg/L MAC prescribed by the EU. The further improvement of organic matter removal by down-stream GAC (granular activated carbon) filtration would enhance interest in this treatment, especially if ozone is already used for other purposes within the plant (e.g. disinfection), and providing the MAC value remains within the currently prescribed range.

The use of KMnO₄ as an algae conditioner also tended to increase DAF process efficiency towards 2 log particle removal, resulting in equal or better performance than the full-scale conventional sedimentation, on occasions even better than the full-scale sedimentation + filtration in terms of particle (d_p>2.75 µm) removal. However, the resultant higher effluent Fe (organo-Fe complexes) and Mn (mostly MnO₂) concentration raised the particle count in the colloidal size range (<0.5 µm) and the turbidity. This situation improved when cationic polymers were introduced as coagulant aids, significantly improving the DAF process efficiency (typically by 5%) on the account of reduced Fe and Mn residuals below their respective MAC values. This option, however, did not lower the accompanying filtrate MFI value to the desired 5 s/L² value. The lowest MFI value achieved by the non-optimised filtration (within the DAF + filtration scheme) was in the range of 20 s/L². This suggests the need for optimisation of the filtration step to fully utilise the conditioning benefits of KMnO₄.

The improved algae removal in the case of conditioning by an oxidant is probably caused by a number of process mechanisms. Motile algal cells are immobilised by the oxidation of the outer

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cell layer, and their metabolic processes are disrupted. This is accompanied with the creation of stress conditions in the algal environment, resulting in EOM (extra-cellular organic matter) excretion, partial algae lysis and IOM (intra-cellular organic matter) leakage. The EOM and IOM act as natural coagulant aids, resulting in spontaneous microfloculation even before coagulant addition. Oxidation and removal of the organic coating on the particles, as well as oxidation of NOM, result in more favourable coagulation conditions through the production of in-situ coagulant, which otherwise would have been complexed and unavailable for coagulation. The resultant MnO_2 in the case of KMnO_4 conditioning increases the particle concentration and promotes sweep coagulation.

The particle size frequency and volume distributions obtained under different process conditions and after different process stages served as the basis for an empirical calculation of the relative and absolute floc density, an input variable in the single collector collision efficiency DAF reaction zone model. Other input variables such as the mean particle (floc) and bubble size, as well as their number and volume concentrations, were also directly measured or calculated. The calculated values for the particle-bubble attachment efficiency α_{pb} support the previous assertion regarding the positive impact of cationic polyelectrolytes, and conditioners. Results suggest that under optimal DAF process conditions almost every second particle (floc)-bubble collision resulted in their attachment. The role of the recirculation ratio (5-10%) and the saturator pressure (500-700 kPa), although significantly affecting the bubble size distribution, proved less important for DAF process efficiency. Thus, critical attention should be paid to the coagulation/floculation process as the major determinant of DAF efficiency. Although difficult to model, the impact of NOM concentration and characteristics on the particle-bubble attachment efficiency is of great significance. Thus, the simplified modelling approach that particle-bubble agglomeration is solely the result of their collision is not fully justified.

DAF proved to be an efficient, robust and flexible water treatment process, and a highly appropriate alternative to sedimentation for the treatment of algae laden water. The lower civil engineering costs and chemicals consumption often will compensate for the higher energy and maintenance costs. Its high algae removal efficiency makes it particularly attractive as pre-treatment before direct filtration, or membrane filtration. Where it is already applied, it's efficiency can be significantly enhanced temporarily (e.g. during short or long term algae blooms) by the application of e.g. cationic polyelectrolytes, oxidants, or a combination of the two. The application of KMnO_4 seems particularly attractive, since no hazardous by-products are produced. Similarly existing sedimentation units can be retrofitted with (enhanced) DAF in case of increased algae problems. Nevertheless, the final choice of the treatment technology or mode should be made after pilot plant investigations and cost-benefit analysis.

Chapter 1

THE ALGAE PROBLEM IN THE NETHERLANDS FROM A WATER TREATMENT PERSPECTIVE

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ABSTRACT : Surface water treatment in the Netherlands receives increased attention requiring better characterisation and evaluation of the 'algae problem'. Investigations and analysis carried out at five drinking water production locations in the Netherlands utilising surface water (amounting to 60% of the overall Dutch surface water production) show that the most significant problem are the cyanobacteria. Seasonal blooms of *Microcystis aeruginosa* and *Oscillatoria agardhii* force short- and long-term process modifications, affecting water production cost. Assessment of possible solutions suggests that, apart from the application of appropriate water quality management measures, new insights on more efficient (pre)treatment processes are critical. In treatment, the agglomeration (coagulation/flocculation) phase is considered the essential step in the removal of the algae. It should be optimised in relation to the pre-treatment preceding it (e.g. application of chemical oxidant/s, and/or microstraining), as well as to the applied solid-liquid separation treatment process. Dissolved air flotation (DAF) emerges as a feasible option given the natural tendency of algae to float. The existing discrepancies between literature and practice call for further research on process mechanisms and suggest that considerable gains can still be made in process optimisation. This research should notably increase insight into the influence of algae morphology and physiology on treatment efficiency.

1.1 INTRODUCTION

1.1.1 Algae in water supply - a growing concern

VEWIN (the Association of Dutch Water Supply Companies) forecasts the drinking water production in the Netherlands to amount to 1360 Mm³/year by the year 2000, of which 16 % (220 Mm³/year) will originate from treated surface water, another 16 % (220 Mm³/year) from dune-infiltrated water, and the remainder (930 Mm³/year) from groundwater [1]. Although this projection for all the sources shows a substantial amount of the infiltrated water is of surface water origin, surface water is not recognized by VEWIN as the preferred alternative. Very often it contradicts one or more of the four basic principles that have to be met for any water source : (i) good water quality, (ii) constant water quality, (iii) low treatment cost and (iv) security from calamities. Nevertheless, surface water is an inevitable choice due to increased water consumption and increasing number of restrictions on groundwater abstraction. The main surface water sources in the Netherlands are the rivers Rhine and Meuse. These two rivers receive a substantial industrial, municipal and agricultural pollution load, resulting in water treatment problems such as high turbidity, colour, organic micropollutants content and high nutrient loads. Eutrophication is recognized as the cause of numerous problems, both in surface water quality and in drinking water treatment. It causes plankton blooms in stagnant and slowly flowing water bodies; their removal emerges as a priority problem in surface water treatment.

Algae cause a wide range of technical and health problems in the treatment and in the distribution step [2]:

i) problems related to algae laden water treatment:

- interference with coagulation and flocculation;
- increased disinfectant demand of the water;
- production of haloform precursors;
- filter clogging and increased use of backwash water; and

- filter penetration;

ii) problems related to algae presence in the distribution system:

- taste and odour;
- precipitation of algal mucopolysaccharides at low pH;
- corrosion of iron mains and discolouration of water;
- shielding of pathogenic bacteria from disinfection;
- aftergrowth of bacteria and higher organisms in the mains;
- infestation of zooplankton; and
- release of toxins.

Nevertheless, water supply experts disagree about the required degree and approach to algae removal in water treatment. Lacking a proper water quality standard, one has to rely on the philosophy that the more algae are removed during common treatment, the better, accepting however that removal efficiency for different types of algae varies widely. The diversity of algal species with respect to their size, form, surface characteristics, production of extra-cellular organic matter (EOM), motility, etc., makes it almost impossible to propose a simple solution that fits water quality management and treatment concerns. Therefore, the 'best available technology' for treatment tends to set the standards.

This chapter intends to address the issue of raw water quality for five Dutch surface water resources, contributing to approximately 60% of the total surface water origin treated water in the Netherlands. Statistical analysis of the water quality parameters contributing to the eutrophication phenomenon will be performed. This will be related to the resultant impact on the algal population, quantity- and species-wise, defining the most significant species from the point of view of imparted treatment problems. The treatment problems, as well as the adopted short and long term reservoir management and treatment strategies will also be discussed. Finally, an overview of existing conventional treatment technologies, as well as more advanced options for their enhanced operation, will be discussed in the context of most efficient algae removal in drinking water treatment.

1.1.2 Eutrophication and algae

Long term changes in the nutrient supply to water bodies cause shifts in the balance and spectra of resources to which phytoplankton is known to respond quantity- and quality-wise. Such changes occur naturally due to a variety of causes. The large increases in the amount of nutrients discharged into water bodies during the past decades, especially in the industrialized countries, and the subsequent changes in the ecology of these water bodies, have called attention to the problem of nutrient enrichment, or eutrophication. Generally, the adjective 'eutrophic' ('well feeding') is used to describe biological aquatic systems with a high input of otherwise growth limiting nutrients, and which therefore support a high level of organic productivity [3].

Eutrophic conditions occur naturally (by leaching of soils and vegetation decomposition), but are nowadays primarily due to human activities (domestic sewage, agricultural run-off, etc.) [4]. The anthropogenic eutrophication, should be tackled at the source of the nutrients discharge, rather

than by the symptoms. In the Netherlands, as well as in some other West European countries, measures like tertiary sewage treatment and a ban on detergent phosphate have drastically reduced nutrient inputs. However, the nutrient rich under-water sediments, the contained fertilizer use in agriculture, and the inevitable leaching of N and P from wastewater, will keep the trophic levels high.

Eutrophication manifests itself as increased water fertility which results in higher primary productivity, i.e. algal growth. Under extreme conditions this can lead to temporary excessive algal biomass and ecosystem instability. As a result of increased eutrophication, the diversity of algae, especially with respect to the planktonic forms, is reduced in favour of large concentrations of a limited taxonomic range of cyanobacteria, chlorophytes and diatoms. The trophic level of the water body thus defines the range and concentration of species present within it [3].

A common annual cycle for eutrophic impounded water in northern temperate regions like the Netherlands consists of a spring population of diatoms which may include *Asterionella formosa*, *Fragilaria crotonensis*, *Tabellaria flocculosa*, *Melosira* spp., *Cyclotella* spp., *Stephanodiscus astraea*, and/or *S. hantzschii*. During early summer, diatoms may be replaced by small species of cryptophyceae or chlorophyceae, and at a later stage by dinoflagellates such as *Ceratium hirundinella*, chrysophytes such as *Dynobryon* spp., *Synura* spp., or chlorophyte genera such as species of *Chlorella*, *Scenedesmus*, *Dictyosphaerium*, *Ankistrodesmus*, *Eudorina* and *Pandorina*. However, in late summer and throughout autumn, dominant populations of cyanobacteria (blue-green algae) may occur, including *Anabaena* spp., *Aphanizomenon flos-aquae*, *Gloeotrichia*, and *Coelosphaerium*; especially *Microcystis aeruginosa* and *Oscillatoria* spp. can be abundant and are able to form surface blooms of aggregated cells under windless conditions [4].

In addition, cyanobacteria tend to dominate a large part of the growing season. Objectionable odours may emanate from water bodies due to decaying biomass, and the water may suffer from offensive tastes and odours, while serious disruption of water treatment processes is experienced. Predation of algae is reduced by colony forming and filamentous cyanobacteria, resulting in the preclusion of effective filter feeding zooplankton (algae size related). This further influences the plankton abundance and composition. In addition, toxins produced by cyanobacteria, such as microcystin, have been responsible for occasional outbreaks of wildlife and livestock illness and death, and has been related to human illnesses [4].

Principles of freshwater ecology phytoplankton form the basis for appropriate design and management strategies of lakes and reservoirs [3]. On the other hand, knowledge of the freshwater ecology does not guarantee the accurate prediction of the plankton responses to particular changes in their environment. Furthermore, the primary problem of water treatment engineers in practice is the problem of biomass 'peaks', composed of different dominant species, regardless of the average level of biomass. Therefore, more knowledge is needed on the behaviour and characteristics of individual organisms. This knowledge should preferably overcome the indiscriminative algae related lumped material parameters approach (chlorophyll, organic carbon, turbidity, etc.). Species differ in their ecological, morphological and physiological characteristics that allow them to survive and grow, and that can strongly influence water treatment.

1.1.3 Growth strategies and characteristics of cyanobacteria

The spatial and temporal distributions of an organism within an ecological system are the result of the evolutionary strategies the organism adopted for growth and survival [5]. Strategies can be defined as sets of similar morphological, physiological, reproductive or behavioural traits that have evolved among species or populations and that are better suited to particular environmental conditions than that of others. Accordingly, different organisms that adopted similar strategies, are likely to resemble in their ecological behaviour.

Based on the logistic growth equation phytoplankton communities have commonly been classified into two main categories : r (characterised by rapid growth and colonisation of a nutrient rich environment) and K (characterised by the ability to out compete other organisms in a nutrient limited environment) [3]. However, other approaches exist that are seemingly more suitable for algae and which classify organisms into three categories marked C, S or R [5]:

- C/competitors : exploitation of environments saturated with light and nutrients, through the investment in rapid growth and reproduction, and to do so before other species;
- S/stress tolerant : operation under conditions of severe depletion of externally supplied essential nutrients; and
- R/ruderals : tolerate frequent or continuous turbulent transport through the light gradient.

There is a considerable overlap of morphometric and physiological characteristics (e.g. unit volume, surface area/volume, maximum linear dimension, photosynthetic efficiency, projected area, maximum growth rate, cellular phosphorus uptake, temperature sensitivity, threshold dose of exposure to saturating light intensity, motility, minimum sinking rate and susceptibility to grazing) of these classes, but the growth and recruitment of a particular strategist will be preferentially promoted in function of specific environmental circumstances. These species are then more likely to build up the largest fraction of sustainable biomass (i.e. become dominant) for as long as the same circumstances persist.

Environmental variables that are supposed to influence the selection among the primary strategists (C, S or R) may be grouped as physical factors (e.g. temperature, stability of the water column, absolute mixed depth, turbidity of mixed layer), chemical factors (notably nutrient concentration) and biotic factors (grazers filtering rate). These may be referred to as externally imposed (allogenic) perturbations. On the other hand, self-imposed (autogenic) perturbations play a substantial role in the definition and selection among the strategists, like euphotic depth (depth of light penetration), reduction due to high biomass concentration, nutrients concentration decline, or increased rate of zooplankton grazing.

Extreme eutrophication expressed in a range of unusual physio-chemical conditions has been proven advantageous to the physiologically and ecologically adaptive cyanobacteria. Ideally, physical and to a lesser extent chemical stability must accompany such extremes. The more a species is versatile in adapting itself to a wider set of survival associated factors, the more abundant, and the longer lasting is its population during the yearly cycle [5]. The cyanobacteria have proven to be highly versatile and adaptable, able to survive and to dominate in conditions highly unfavourable for other species. This versatility makes cyanobacteria able to survive and dominate in water bodies during protracted periods of the annual plankton cycle. In some

relatively large and shallow lakes, e.g. the Hartbeespoort Dam and the Vaalkop Dam in South Africa, cyanobacteria compose the most significant part of the population throughout the year; this situation probably reflects a final eutrophication stage of a lake, considering the comparatively constant climatic and nutrient conditions [6].

Carbon, nitrogen and phosphorus are the essential nutrients for cyanobacteria. The nutrient uptake pattern influences the population structure in the water body. It has been noticed that high phosphorus levels generally result in cyanobacterial dominance, though blooms also occurred in low or undetectable phosphate concentrations [7]. The latter is probably caused by fast and almost complete uptake of phosphate by blooming species, leaving the water with low phosphate concentrations for a period of time. A large diversity in response towards phosphorus deficiency has been found, with little or no growth rate dependence on external (i.e. soluble reactive phosphorus, or total dissolved) phosphorus. The rate of phosphate uptake is optimal between pH 7.5 and 8.5 and declines sharply below neutral pH; it is larger in light than in dark conditions.

Most cyanobacteria are N_2 fixing organisms [7]. Fixation of dissolved atmospheric N_2 fixation is carried out by unicellular organisms, by filamentous, heterocystic forms (such as *Anabaena* and *Nostoc*) and by filamentous strains lacking heterocysts (heterocysts and akinetes emerge in nutrient exhausted surroundings, akinetes having a preservatory role and being able to germinate in better nutrient conditions).

The role of carbon in the nutrition and life cycle of cyanobacteria is still not sufficiently clarified, but together with nitrogen it is readily available from the atmosphere, unlike phosphorus. The deficiency in nitrogen and phosphorus is known to lower the respiration rate, and causes accumulation of carbohydrates during the light period of the day, preparing cells for survival in the dark [7].

Cyanobacteria depend on efficient photosynthesis. Like other taxa, they show phototactic/phobic and chemotactic/phobic responses, indicating their affinity to light and limnological conditions. Cyanobacteria, however, are versatile and adaptive, which allows them to out compete other algae in unfavourable environments with regard to nutrients and light. They can adapt to the light-dark regime in a way that gives them the possibility to maintain a higher photosynthetic activity during short photoperiods, so that they may survive for prolonged periods in the dark. They can also adapt to irradiance intensity, whereas a planktonic species from a temperate lake may be killed by prolonged exposure to irradiance exceeding a certain high intensity. A characteristic adaptive mechanism adjusts their chlorophyll content to the light's wave length, to maximize light-efficiency [7].

Cyanobacteria also show motility within the water column. Two types of movement are exerted by cyanobacteria when searching favourable living conditions [7]: phototactic gliding of cyanobacteria attached to surfaces (of suspended sand, clay particles, etc.), and light regulated buoyancy. The rate of displacement, and the frequency of path reversals are related to e.g. the vertical movement in a water body, and are influenced by temperature, viscosity and chemical factors. On the other hand, factors that change rapidly or a concentration gradient, like light intensity, wind and chemical factors, influence the direction of the movement.

Finally, circumstantial evidence exists that some cyanobacteria secrete organic substances of which some are inhibitory or toxic to other organisms, including fish, waterfowl domestic livestock, and humans [3]. An assessment from almost 300 sites all over the world, in the period 1981-1989, shows that the likelihood of an individual bloom being toxic is 45-75 %. Usually, *Microcystis*, *Anabaena*, *Oscillatoria* and *Aphanizomenon*, and less often *Gomphosphaeria*, *Coelosphaerium*, *Nodularia*, *Nostoc* and *Cylindrospermum*, were involved [8]. Research conducted on toxic algae blooms in the UK in 1989 indicated associated death of dogs and sheep, and illness in people. The toxicity was attributed to microcystin poisoning, originating from *M. aeruginosa*. Such blooms and associated toxicity were also recorded in Norway, Denmark, Sweden, USSR, Finland [8], Australia [9] and Brazil [10]. In Finland, cattle poisoning attributed to cyanobacterial blooms took place at three sites in 1985 and 1986, and similar blooms occurred in drinking water sources as well. The isolation of strains of cyanobacteria confirmed the presence of hepatotoxic and neurotoxic strains of *Anabaena*, as well as hepatotoxic strains of *Microcystis* and *Oscillatoria* [11]. The National Rivers Authority in UK reported in 1990 about the 1989 cattle kill and human infestations. 68% of the 78 water bodies tested for toxicity were found positive, which was related mostly to the presence of *M. aeruginosa*, though other cyanobacteria were present as well, notably *Aphanizomenon flos-aquae*, *Anabaena*, and *Oscillatoria* [6]. In 1990 the incidence of *Anabaena* spp. bloom in a recreational lake in the Netherlands was found to have caused summer flu in humans. Toxicity analysis of water containing *Microcystis* spp. blooms and originating from the Andels Meuse and Braassemer Lake in the Netherlands showed the presence of hepatotoxic microcystin toxin. This led to the conclusion that consumption of these toxins via drinking water is possible [12]. Further research was advocated on the influence of environmental factors on the characteristics and levels of cyanobacterial toxicity, as well as on the stability and persistence of the toxins involved.

No consensus exist in literature on the question whether cyanobacterial toxins are relevant from the point of view of water treatment and distribution. However, it is suggested that the public health aspect should receive more attention. Distinction is to be made between physical contact with surface water containing cyanobacterial toxins, consumption of such water, and consumption of water that contained cyanobacteria before treatment. Similarly, it still is uncertain whether the toxic substances, rarely identified, are secreted by viable algae, or whether they are produced during putrescence of algal matter [3]. Significant levels of toxins found during water treatment may be provoked by cell lysis due to for example oxidants application.

Controversy also exists regarding the nature of the human infestations caused by ingestion of cyanobacterial toxins. Some species of cyanobacteria have been identified as causing gastroenteritis, however, only when consumed in large numbers [13]. High cyanobacteria concentrations in the raw water supply have been found to cause an epidemic of pyrogenic reaction among kidney patients consuming such water [14]. Lysis of cyanobacteria, when an algicide or oxidant disinfectant is applied, may result in harmful concentrations of toxins in domestic water supplies causing liver injury, gastroenteritis, or hepatoenteritis [15]. Others on the other hand, state that conclusive evidence exists that cyanobacterial toxins are a health hazard in drinking water [4].

However, not all cyanobacteria species are toxic, and differences exist between strains of the same species. The recognition of different environmental conditions (nutrient composition, light availability, presence of competitor algae species, etc.) and the governing mechanisms which may

result in differing toxicity of the same strain at different locations, has not been addressed so far and deserves further research attention.

In conclusion, it appears that the excretion of toxins is a widely recognized problem, and that high cyanobacteria concentrations warrant much caution. It is therefore recommended that a careful approach is adopted to the application of certain chemicals such as oxidants (e.g. ozone or KMnO_4) as particle (algae) conditioners due to the potential danger of toxins leakage from ruptured algae cells. The fate of toxins along the treatment line, especially the final product toxin levels deserves research attention. The issue of long term toxins consumption and health implications has not been addressed so far and also attracts attention. The recommended approach is to face and assess the potential toxicological problems regardless of the high cost and long duration of such analysis.

1.1.4 *Microcystis* and *Oscillatoria* spp.

Whilst cyanobacteria species are generally recognized as K-strategists, species differ with regard to their R or S character. The most widely spread cyanobacteria are *Microcystis* spp., which are characteristic for eutrophied lakes, and a typical S-species, able to dominate periods of late summer (high temperatures), and severe nutrient depletion. Their physiological and morphological characteristics make them a particular nuisance in water treatment. On the other hand, in the Netherlands concern grows about the incidence of the *Oscillatoria* spp. in some water resources. *Oscillatoria* spp. are known to possess an effective light-capturing mechanism across a wide band of the visible spectrum, as well as a buoyancy regulating mechanism. These make for a pronounced R-species, allowing the species to inhabit constantly mixed lake environments [5]. There is obvious differentiation between these two cyanobacterial genera, with respect to their shape, size, extra cellular organic matter (EOM) characteristics, nutrient uptake, buoyancy regulating mechanism, etc. On the other hand, they are both identified as cause of numerous problems in water treatment [4, 6, 8, 12, 16, 17, 18]. Therefore, they are of particular further interest to this study.

Microcystis spp. are encountered worldwide. They are spherical in shape, of 3-7 μm in size, and can occur in irregularly shaped colonies of often more thousands cells, as well as single cells. There are indications that the single cell form is an adaptation of this species developing at later stages of their life cycle (i.e. later during the season), due to the dissociation of colonies in order to provide better light availability for all cells. These species favour warmer water ($>14^\circ\text{C}$) and stable, relatively low nutrient availability, combined with stable hydraulic conditions [3, 16].

Oscillatoria spp. are characterized by an oscillating movement and lack of definite sheath. Their cells exceed 100 μm in length and are 3-5 μm in diameter; trichomes are solitary or intermingled, forming non-parallel bundle-like structures. Generally, they inhabit temperate lakes of two distinct types : (i) mildly eutrophied, large, deep, and usually alpine lakes, and (ii) shallow, enriched, unstratified basins, where they form stable maximum concentrations in the summer metalimnion, provided that this is located within the euphotic zone. They possess an extremely efficient light capturing mechanism, functional across a wide band of the visible spectrum, favouring low irradiance and achieving maximum growth rates at relatively low nutrient availability [3].

Cyanobacteria in general, and especially *Microcystis* spp., are also able to survive in conditions unfavourable for other species. For example, *Microcystis* spp. can overcome O₂ toxicity. With the high light irradiance of late summer, and high O₂ concentrations and low CO₂ concentrations, O₂⁻ (superoxide) is produced that damages cytochromes, pigments and other segments of the algal photosynthetic system. To prevent the toxicity of O₂⁻ the enzyme superoxide dismutase (SOD) is manufactured in algal cells. However, prolonged high light intensity triggers a drop in SOD level and die-off of algae. This phenomenon has been observed with most algal species, except for *Microcystis* spp. which are able to maintain high levels of SOD and survive such conditions [7].

Characteristic for *Microcystis* spp. and *Oscillatoria* spp. is the existence of gas vacuoles [7]. Up to 10,000 of these organelles are present in a single cell. Generally they have the shape of a hollow cylinder of 70 nm diameter with a conical cap at each end. They are composed of protein, the wall being permeable to dissolved gases in surrounding liquid, but impermeable to liquid (the pressure inside being usually atmospheric). Nutrients as well as Cl⁻ and K⁺ uptake are known to play a major role in rise or fall of turgor (difference in hydrostatic pressure between the inside and outside of the cell) and consequent buoyancy. This enables the cells to attain an as good as possible position in a water body (mostly in the vertical sense) with regard to light and nutrients [7].

Other characteristics of the genera establishing their dominance are : allelopathy and light attenuation [7]. Allelopathy involves chemical inhibition of other organisms by the excretion of inhibitory organic compounds. Attenuation of light occurs as the cyanobacteria can increase their biomass very fast, thus increasing turbidity and limiting the growth of competitors. Cyanobacteria are vertically motile during diurnal or higher order nutrient fluctuations, favouring them as inhabitants of water bodies during the late summer and autumn. In addition *Oscillatoria* spp. are able to maintain approximately neutral buoyancy, and cannot be ingested by grazing zooplankton because of their length of >100 µm, ensuring low loss rates [3].

However, characteristic differences exist between the two genera with respect to nutrient affinity and uptake. *Oscillatoria* spp. have been reported to fix nitrogen [3]. *Microcystis* spp. in contrast, have been reported as non-nitrogen-fixing species [7]. Low N/P ratios can be expected to favour N₂ fixing *Oscillatoria* spp., whilst the opposite would favour the growth of *Microcystis* spp.

The toxins related to cyanobacteria blooms may be grouped into three families: neurotoxins, lipopolysaccharide endotoxins, and hepatotoxic peptides. Microcystin belongs to the hepatotoxic family and is a potent and specific in vitro inhibitor of phosphatases, with an action similar to that of the tumour promoting toxin of shellfish poisoning, okadaic acid. This particular toxin, consists of polypeptide containing D-serine, L-orithine and some protein L-amino acids (circular peptide) [8, 16].

1.2 PROBLEM ASSESSMENT

Analysis of operational data of five major surface water treatment plants in the Netherlands has been performed, of which a representative cross-section is given in Table 1.1 [19]. These all produce water for different purposes (drinking water, industrial water and dune infiltration water), and face growing problems caused by algae. They are interested in efficient short- and long-term

treatment strategies and process modifications to address the algae problem. The enterprises are North Holland Water Supply Companies (PWN), Rhine-Kennemerland Water Transport Company (WRK), Dune Water Supply Company South Holland (DZH), Amsterdam Water Supply (GWA) and Energy and Water Supply Company Rhineland (EWR). At a later stage of the research after the further presented analyses were completed, two more enterprises with similar interests were involved, namely Water Supply Association Overijssel - Regio Oost (WMO, formerly Water Supply Company Oost-Twente - WOT), and Groningen Water Supply Company (GWG) (Fig. 1.1).

Fig. 1.1 Locations of the water treatment plants of the seven water treatment enterprises.

Although the origin of all concerned surface water is in essence river Rhine or Meuse water, the water quality in the five sources largely varies, because abstraction is almost never practised directly from the rivers. GWA treats water from the Loenderveen Lake, which is regularly replenished with Bethune polder drainage water (in the future to be mixed with Amsterdam-Rhine canal water at a ratio of 1:1). PWN and WRK treat water from the IJssel Lake, which is fed with water from river Rhine via one of its branches, the IJssel river. DZH practices dune infiltration of water abstracted from the Andels Meuse (a dammed arm of river Meuse, practically serving as a reservoir), and EWR of Rhineland polder drainage water. The purpose of their treatment also varies (drinking water, industrial water, water for dune infiltration, etc.) and so does the water treatment technology applied. Table 1.5 summarises the selected water quality management and treatment technology.

Table 1.1 Year-average minimum and maximum water quality of surface water resources, based on average monthly measurements by the water enterprises.

Water Quality Parameter	IJssel Lake (Reservoir) PWN Andijk 1984-89	IJssel Lake (Reservoir) WRK Andijk 1986-92	Loenderveen Lake GWA Amsterdam 1986-92	Andels Meuse DZH Den Haag 1986-92	Polder Water EWR Katwijk 1989-92
Temperature °C	1.2-20.7	1.4-20.3	3.6-22.3	1.8-22.4	1.5-21.6
pH	7.9-9.45	7.7-8.9	7.5-8.4	7.7-8.6	7.6-8.5
PO ₄ -o mgP/L	N.M.	0.0077-0.095	0-0.014	0.006-0.063	0.124-1.04
PO ₄ -t mgP/L	0.06-0.76	0.05-0.25	0.007-0.04	0.04-0.2	0.39-1.15
NO ₃ +NO ₂ mgN/L	N.M.	0.57-4.52	0.42-1.44	1.74-5.5	1.1-6.6
NH ₄ mgN/L	0.04-0.79	0.04-0.9	0.12-0.89	0.08-0.55	0.17-2.78
SiO ₂ mgSi/L	N.M.	0.05-2.8	3.7-13.1	0.25-5.6	1.04-11
Chl <i>a</i> µg/L	8.6-198	6.5-101	1.3-8.4	1.6-37	1.5-51
Chl <i>t</i> µg/L	18-238	13-132	2.7-10.3	2.6-58	N.M.
Algal Vol. mm ³ /L	1.1-411	0.4-97	N.M.	N.M.	N.M.

Chl *a* : Chlorophyll-*a*

PO₄-o : ortho phosphate

N.M.: not measured

Chl *t* : Chlorophyll-total

PO₄-t : total phosphate

During the collection and processing of the data, it appeared that the degree of attention paid to algae, as demonstrated by the frequency and diversity of algae related measurements, varies between the enterprises, and is proportional to the number of problems experienced in the past. The attention is determined, for example, by whether the end product is drinking, industrial or infiltration water. This inconsistency in the data set is compounded by the absence of a proper algae related standard for product water, and by the absence of a proper measuring technique that is able to include all types of algae in proportion to their actual concentrations.

Statistical analysis of the data related to comparison of the five water resources was performed. The aim of the two way analysis of variance (ANOVA) was to test the similarity between the (year-average) raw water quality of the five water resource localities, and to check whether the raw water quality of each of the water resources has changed within the analysed time period (see Table 1.1). The results show : (i) significant differences between the raw water quality at the five localities (raw water resources), and (ii) no significant changes of the water quality of each of the localities during the analysed periods. Based on a multiple range test, Table 1.2 reflects the similarities for the analysed water quality parameters at different localities.

Table 1.2 Similarity of raw water quality between the five locations. a, b, c, d denote typical water quality categories, based on the two way ANOVA.

Local	°t	pH	PO ₄ -o	PO ₄ -t	NO ₂₊₃	NH ₄	SiO ₂	Chl-a	Chl-t
1	ab	c	N.M.	c	N.M.	a	N.M.	c	c
2	a	a	a	a	a	a	a	a	a
3	a	b	a	b	b	a	a	b	b
4	bc	a	b	d	b	b	c	ab	N.M.
5	c	ab	a	b	b	a	b	a	a

1 - PWN reservoir 3 - DZH Andels Meuse 5 -GWA Loenderveen Lake

2 - WRK III reservoir 4 - EWR polder water

N.M.: no measurements

available for the locality

Chl a :

Chlorophyll-a

Chl t :

Chlorophyll-total

NO₂₊₃ :

nitrites + nitrates

Multiple correlation analysis of the variables (analysed raw water quality parameters) for all the localities (raw water resources) shows high correlation coefficients between chlorophyll-tot on one hand and pH, ortho-PO₄, total PO₄, NO₂ + NO₃, and SiO₂ on the other, and low correlation coefficients between chlorophyll-tot and °t and NH₄ (Table 1.3). The lack of data for SiO₂ for the polder water may explain to some extent the high negative correlation coefficient obtained.

Similar correlations were obtained for chlorophyll-a. Based on the maximum correlation coefficients, simple and multiple linear regression analyses were carried out, showing highest r^2 values for simple linear regression between log(chlorophyll-tot) and log(PO₄-t) [$r^2=0.87$], and for multiple linear regression between log(chlorophyll-a), log(PO₄-t) and pH [$r^2=0.80$]. Fig. 1.2 represents a plot of the observed versus the predicted log(chlorophyll-a) values in function of the phosphate concentration and pH. The transformation equation predicting the (chlorophyll-a) value on a year-average maximum basis is : $\text{pr.log(chlorophyll-a)} = 0.52 \log(\text{PO}_4\text{-t}) + 0.6(\text{pH}) - 3.2$. The highest r^2 values for PO₄-tot suggest that phosphorus is the key element which controls algae growth in surface water impoundments, consistent with the conclusions of Vollenweider [20] and also elaborated by many authors like Sakamoto [21], Dillon and Rigler [22], and Tillman et al. [23], for water impoundments under a range of conditions.

Table 1.3 Correlation coefficients (r) and significance levels (P) for correlation of log (chlorophyll-tot) with log values for different variables (except for pH which is a log value itself). Number of data $n=15$.

Coef.	°t	pH	PO ₄ - o	PO ₄ - t	NO ₂₊₃	NH ₄	SiO ₂
r	0.11	0.76	0.96	0.93	0.81	-0.09	-0.8
P	0.7	0.0011	0.0000	0.0000	0.0002	0.75	0.0003

Fig. 1.2 Observed versus predicted log(chlorophyll-*a*) values, $r^2=0.8$.

The reservoirs' chlorophyll and phosphorus data were superimposed on a graph by Harris [24] (based on results for a combination of lakes analysed by Janus and Vollenweider, and a number of oligotrophic Canadian lakes); the results fall in the 95% confidence limit of the regression line (Fig. 1.3).

Fig. 1.3 Annual mean chlorophyll vs. P concentrations for the five impoundments, superimposed on a graph from Harris [23].

The close relation between chlorophyll-*a* and phosphorus, emerging from the multiple regression analysis, suggests causality and is again in accordance with Vollenweiders results. However, the causal relation between chlorophyll-*a* and pH is more complex. Although high correlation coefficients do not automatically imply the presence of causal relations, the causal relation between chlorophyll-*a* and pH may be supported by the CO₂/pH relation hypothesis. Indeed, the relatively high pH of the five impoundments (pH 7.5-9.45 on year average basis) favours the growth of cyanobacteria at high eutrophication levels [25], as recorded in our case [19]. In addition it may be hypothesized that a feedback mechanism exists between chlorophyll, PO₄ and pH, in the sense that resuspension of phosphorus from reservoir sediments occurs under the recorded high pH conditions, which supports further algae growth [26, 27].

The intensive photosynthetic activity of high biomass concentrations further shifts pH to high values, possibly to the point where C becomes a limiting factor. As cyanobacteria have lower saturation constants K_m for uptake of total carbon and CO₂, this condition combined with the high pH promotes cyanobacteria dominance [25].

Summaries of the observations on the most significant species encountered in the five impoundments describing their characteristics and treatability, are presented in Table 1.4. The treatability of the algae species, i.e. the extent of their removal in water treatment, is mainly affected by (i) the treatment technology applied, (ii) optimization of the applied treatment technology, and (iii) characteristics of the algal species involved. Although the choice of the treatment technology rarely is made on the basis of efficient algae removal alone, the occurrence of the periodical algae blooms plays an important role. Under normal circumstances (moderate algae concentrations) algae can be removed more or less efficiently by commonly applied treatment technologies.

However, this technology is not necessarily optimal for the removal of all the algae species concurrently or separately (at other time periods) present in the water. The algae characteristics and properties influence their removal, including size, shape, cell surface characteristics (composition of algal cell wall and resultant colloidal surface charge characteristics), excretion of EOM and motility. Many of these characteristics vary with algae age, as well as within subspecies, limiting the removal predictability. In this light, the terms 'Problematic', 'Good', etc. treatability of Table 1.4 are only indicative. They are used to characterize the general degree of problems related to a particular algal species removal, the effect on effluent water quality, and the water production costs.

Each of the studied water impoundments is characterised by seasonal (yearly) variations. Algal blooms are common during certain periods of the year, but they are proportional to the degree of eutrophication and most acute in the cases of WRK (Figs. 1.4 and 1.5) and PWN treating IJssel Lake water. A pronounced seasonal occurrence of cyanobacteria (especially *M. aeruginosa*) is found in all five water resources, again showing proportionality between concentration and the degree of eutrophication. This was found to be most typical in Loenderveen Lake. While diatoms are characterised as superior competitors under phosphorus limitation (high N/P ratio) and water temperature below 14°C, green algae and cyanobacteria are dominant under nitrogen limitation (low N/P and Si/P ratios) and high temperatures [23].

Table 1.4 The most significant algae species encountered in the five impoundments, indicating their basic characteristics and treatability.

Algae species and taxonomy Location/Treatment plant	Figurative representation	Characteristics	Treatability
<i>Melosira</i> spp. (Diatomeae) 1. 4. 5.		Porous, capsule like, cylindrical cells forming filaments; blooms; L=10-40 µm; W=5-20µm.	Problematic
<i>Stephanodiscus</i> spp. (Diatomeae) 1. 2. 4.		Single, porous cells, spines at edges; in hard waters or basic lakes; L=8-20 µm.	Problematic
<i>Asterionella</i> spp. (Diatomeae) 1. 3.		Often very abundant, spoke like arrangement of rectangular frustules; in hard waters; L=40-130 µm; W=2 µm.	Moderately problematic
<i>Nitzschia</i> spp. (Diatomeae) 3.		Commonly solitary frustules, tapered at ends, straight or sigmoid, porous; L=25-110 µm; W=5-10 µm.	Moderately problematic
µ-algae (Diatomeae) 3.		Small capsule like cylindrical cells, very much like <i>Stephanodiscus hantzschii</i> .	Problematic
<i>Microcystis</i> spp. (Cyanophyta) 1. 2. 3. 4. 5.		Irregularly shaped colonies of thousands of sphere cells, also single cells; gas vacuoles (N ₂); extensive blooms-surface mat; D=3-7µm.	Problematic
<i>Oscillatoria</i> spp. (Cyanophyta) 1. 2. 3. 4.		Cells forming filaments - solitary or intermingled, cells lack definite sheath; blooms; L=2-4 µm, (L ₁ =100 µm); W=3-5µm.	Problematic
<i>Aphanizomenon</i> spp. (Cyanophyta) 4.		Cells forming filamentous bundles, heterocysts present; blooms; L=5-15 µm, (L ₁ =150 µm); W=5-6 µm.	Problematic
<i>Anabaena</i> spp. (Cyanophyta) 3.		Cells forming filaments, heterocysts present; upper layers; blooms - surface mats; L=6-8 µm, (L ₁ =30-35 µm); W=6 µm.	Moderately problematic
<i>Scenedesmus</i> spp. (Chlorophyta) 4.		4, 8 or 12 oval or fusiform cells forming colonies, 1 or 2 spines at end cells; L=10-15 µm	Moderately good
<i>Coelastrum</i> spp. (Chlorophyta) 1. 4.		Spherical or polygonal cells forming hollow colonies, by protuberances from mucilaginous sheaths; D=8-12 µm.	Moderately good
<i>Oocystis</i> spp. (Chlorophyta) 1.		Elliptic or lemon shaped, one or more generations of mother cell walls enclose daughter cells; L=14-26 µm; W=10-20 µm.	Good
<i>Cryptophyceae</i> spp. 1. 3. 5.		Solitary cells, rarely colonial, protozoan like, 2 flagella, chloroplast pigments - brown, blue or red.	Moderately problematic
<i>Chrysophyceae</i> spp. 3. 5.		Unicellular or colonial, rarely filamentous, chloroplast pigments - yellow, brown, golden brown, thick wall, 1 or 2 flagella, motile.	Moderately problematic

1.- PWN reservoir

2.- EWR polder water

3.- GWA Loenderveen Lake;

4.- WRK III reservoir

5.- Andels Meuse.

L - cell length

L₁ - total length of filament

W - cell width

D - cell diameter

Comment : The morphology of different algal species asserts the need of different algae counting procedures. *Microcystis aeruginosa* is counted as single cells and as colonies. Bundles of filamentous species most often disintegrate during the preparation of the sample; an attempt is made to count all the algae that may form such bundle structures. *Scenedesmus* spp. of 8 or 12 cells are counted as 2 or 3 *Scenedesmus* entities. Similarly other disintegrated colonial species are counted as separate entities.

Fig. 1.4 Average monthly algal count for the WRK III reservoir (integrated over the total reservoir depth of 20 m), for different algal taxa in the period 1988-92.

Fig. 1.5 Average yearly algae count for different taxa in the WRK III reservoir for the period 1988-92.

Reservoir management involving chemicals application (affecting nutrients availability, pH, etc.) and water mixing within the reservoir, as well as climatic conditions, influence the phytoplankton composition as for example in the PWN and WRK reservoirs where higher

concentrations of *Oscillatoria* spp. were found in the period of 1990-92. This is consistent with other experiences under Dutch climatic circumstances [25]. This suggests that the current raw water quality criteria are insufficient to avoid prolonged cyanobacteria blooms. Cyanobacterial dominance refers to a high biomass concentration, which is correlated (indirectly) to the nutrient level. This is the case in particular with *Oscillatoria* spp., while for *Microcystis* spp. it is the limiting nutrient that most likely will determine the magnitude of its bloom.

1.3 DISCUSSION

1.3.1 Algae and water treatment - problems and remedies

1.3.1.a Reservoir management

To cope with algae efficiently requires a combination of detailed water quality monitoring, reservoir water quality management, and water treatment measures. Appropriate and especially well timed microbiological analysis (i.e. done immediately after sampling and not postponed for weeks, which often is the practice), accompanied with other physio-chemical analysis provides a better tool for prediction of algae concentration and species variations. Although treatment plant operators are found to be increasingly aware of the importance of the prediction capacity, it often is neglected or insufficiently addressed. Delays between sampling and analysis typically result in poor treatment process operation. On the other hand, it is generally very effective to reduce the algal load ahead of the treatment through reservoir water quality management and pretreatment as it appears that no treatment step on its own is able to ensure adequate removal rates. A range of reservoir management techniques are applied in this context with varying degrees of success: phosphorus removal (either by precipitation with coagulant, or by diverting phosphorus contributing flows), artificial destratification by mixing of the water column, use of algicides (mainly copper sulphate), sonic disruption of algal cells, suction dredging, and aeration of the hypolimnetic layer [4]. Reservoir management significantly diminishes the algae load, but the appropriate approach is reservoir and algae specific.

The successful application of better reservoir design is the case presented in Fig. 1.6. The use of the shallow 3 m deep PWN reservoir was temporarily supplemented by the 20 m deep WRK III reservoir, resulting in drastic reduction of the exposure to sun-light and subsequent poor algae growth. Examples of successful pre-treatment include the coagulation with ferrous sulphate and settling of water entering the water reservoir of GWA, the application of ferrous sulphate in Andels Meuse by DZH and in the WRK III reservoir (which also includes periodical mixing of the water column). On the other hand, such measures have also been reported to yield adverse effects. In the Wahnachtalsperrenverband in Siegburg, Germany, phosphorus reduction in the raw water reservoirs led to a rapid increase in diatom level at the expense of other algae, and resulted in poorer overall treatment performance during periods of their blooms.

Fig. 1.6 Average algal volume for different taxa in the reservoirs of PWN (3 m deep) and WRK III (20 m deep), (1987 data unavailable).

1.3.1.b Treatment process

Cyanobacteria and in particular *Microcystis* spp. and *Oscillatoria* spp. are known to pass readily through treatment and end up in drinking water in relatively large numbers. A critical feature of the *Microcystis* spp. is their ability to form colonies, as well as to exist in a single cell form, whilst *Oscillatoria* spp. can exist as solitary or intermingled filaments. Small algae pass through filters more readily than larger ones so that when blooms of small algae, such as *Microcystis* spp. occur, their counts in treated water are high especially towards the end of the bloom period when colonies disintegrate naturally, or as a consequence of algicides. Up to 5.3×10^4 cells/mL of the former and 4.5×10^4 cells/mL of the latter species have been recorded by the Severn Trent Water Authority in 1985 and 1986 in the United Kingdom, for filtered water [4]. *Microcystis* spp. cells can rupture and leak intra-cellular substances during turbulent aqueduct transport from the reservoir to the treatment site (rupture rate between 61-72%) [28]. Similar rupture and colony break-up may be expected during initial pumping stages in water treatment plants, representative for Dutch operation circumstances. In other words, water treatment plant design may in itself aggravate the algae related problems.

In general, the treatment approach in the studied plants is complex and aims at the overall removal optimization of different pollutants, including suspended solids, dissolved and colloidal matter, organic micropollutants, heavy metals, algae, etc.; this varies depending on the raw water source as well as the final product user (Table 1.5). Coagulation and flocculation, and rapid sand filtration are applied in all plants, while granular activated carbon filtration is used in three cases (GWA, WRK and PWN). Dissolved air flotation is applied at DZH and EWR, while sedimentation is practised at WRK III and PWN. Slow sand filtration is practised only at the GWA.

Table 1.5 The treatment processes in the five studied treatment plants.

MANAGEMENT &TREATMENT TECHNOLOGY APPLIED	GWA 30Mm ³ /y Weesperkarspel	WRK III 110 Mm ³ /y Prinses Juliana	PWN 35 Mm ³ /y Andijk	DZH 70 Mm ³ /y Bergambacht + Scheveningen	EWR 15 Mm ³ /y Lindenberg
pre-coagulation	###	###		###	
pre-settling	###			###	
reservoir	###	###	###	###	
reservoir mixing		###			
pre-filtration	###			###	
straining 35µm			###		
sieves 200µm		###			
pre-chlorination		#P#	###		
transport Cl ₂				#P#	
ozone	###				
softening	###				
coagulation/ flocculation	ooo	###	###	###	###
sedimentation	ooo	###	###		
flotation (DAF)				###	###
rapid sand filters	ooo	###	###	###	###
activated carbon	#o#	###	###		
slow sand filters	###				
straining 30µm			###		
post-chlorination	#P#				
ClO ₂ disinfection			###		

- PRESENTLY IN OPERATION

ooo - PREVIOUSLY IN OPERATION

#P# - PERIODICALLY APPLIED

#o# - REPLACED WITH (IN OPERATION NOW)

Algal blooms and typically those of cyanobacteria, pose the most serious treatment problems. These include : (i) interference with the coagulation and flocculation process, (ii) increased disinfectant demand, (iii) trihalomethane production, (iv) frequent filter blocking, and (v) increased use of backwash water (Table 1.6). This translates into higher production cost. Process modifications to address these problems can also be expensive or have side effects. As no algae standards or objectives exist, water companies tend to deal with the problem on the basis of 'as much as possible' removal, within the technological context and production cost bracket that is considered commonly acceptable. This approach is usually indiscriminatory and poorly focused. Local guide levels exist such as those at the Wahnachtalsperrenverband in Siegburg, Germany [29], (0.1 µg/L chlorophyll-*a* in treated water).

Table 1.6 Present and anticipated situation in the five studied Dutch water enterprises. Most significant alga species encountered, related treatment and distribution problems, and remedies (illustrations from [30], except for GWA photo material of μ -algae deposits on GAC filter).

Company (% prod. of total surf. water in NL)	Raw water source, treatment plant and process	Characteristic algal species in water resources	Present situation with respect to raw water source and treatment	Anticipated development
PWN (8 %)	IJssel Lake-Andijk -Straining 35 μ m -Cl ₂ 6 g/m ³ -FeClSO ₄ 16 g/m ³ -Ca(OH) ₂ 30.5 g/m ³ -Polyelect. -0.5 g/m ³ -SED, v=3.5 m/h -RSF, v=10 m/h -GAC, v=13.5 m/h -Straining 35 μ m -ClO ₂	<i>Microcystis</i> spp. (Cyanophyta) <i>Oscillatoria</i> spp. <i>Melosira</i> spp. (Diatomeae) <i>Stephanodiscus</i> spp. <i>Asterionella</i> spp. <i>Coelastrum</i> spp. (Chlorophyta) <i>Oocystis</i> spp. <i>Cryptophyceae</i> spp.	Serious current operational difficulties: ■ 100% coagulant and coagulant aid dose increase (up to 40mg/l Fe ³⁺ and 1.0 mg/l polyelectrolyte) ■ trihalomethane formation ■ increased backwash water quantity ■ algae passing through treatment	■ In long term only partial improvement of present eutrophication of raw water resource expected ■ Partial improvement of situation due to deepening of reservoir before treatment
WRK (25 %)	IJssel Lake-Princess Juliana, Andijk -Screens 200 μ m -Fe ₂ (SO ₄) ₃ 20 g/m ³ -Polyelect.-0.2 g/m ³ -Ca(OH) ₂ -SED (lamella) -RSF, v=10m/h -GAC (partial)	<i>Microcystis</i> spp. (Cyanophyta) <i>Oscillatoria</i> spp. <i>Aphanizomenon</i> spp. <i>Melosira</i> spp. (Diatomeae) <i>Stephanodiscus</i> spp. <i>Coelastrum</i> spp. (Chlorophyta) <i>Scenedesmus</i> spp.	Serious current operational difficulties: ■ 100% coagulant and coagulant aid dose increase (up to 40 g/m ³ Fe ³⁺ and 1.0 g/m ³ polyelectrolyte) ■ trihalomethanes formation ■ increased backwash water quantity ■ algae passing through treatment ■ increased MFI and AOC values and possible infiltration wells clogging	■ In long term only partial improvement of present eutrophication of raw water resource expected ■ Partial improvement of situation due to mixing and coagulant application in reservoir
GWA (7 %)	Loenderveen Lake-Weesperkarspel -Ozone - 2-5 g/m ³ -Softening -FeCl ₃ (1992) -Polyelect.-(1992) -SED(lamella, 1992) -PAC (1992) -RSF (1992) -GAC -SSF	<i>Microcystis</i> spp. (Cyanophyta) <i>Oscillatoria</i> spp. <i>Anabaena</i> spp. <i>Nitzschia</i> spp. (Diatomeae) <i>Asterionella</i> spp. μ -algae <i>Cryptophyceae</i> spp. <i>Chrysophyceae</i> spp.	Operational difficulties: ■ surface clogging of GAC filters by phyto- and zooplankton cake layer	■ New surface water source of inferior quality (contributing to eutrophication) to be used to double production capacity ■ Reduction of algae load over GAC filters due to: -direct filtration mode of filtration plant at intake (1-3 g Fe ³⁺ /m ³) -reduced ozone doses at beginning of process
DZH (16 %)	Andels Meuse-Bergambacht + Scheveningen -RSF -PAC- 2-4 g/m ³ or -Al ₂ (SO ₄) ₃ 4-6 g/m ³ -DAF, t=7-14 min -RSF, v=10 m/h	<i>Microcystis</i> spp. (Cyanophyta) <i>Melosira</i> spp. (Diatomeae) <i>Cryptophyceae</i> spp. <i>Chrysophyceae</i> spp.	Operational difficulties: ■ algae and cellular products passing through treatment ■ increased AOC values, resulting in reduced pipe flow due to increased k-values of pipes	■ Unchanged situation as long as application of phosphate precipitation in Andels Meuse is allowed ■ New direct filtration plant for algae removal in Brakel under investigation
EWR (3 %)	Rhineland polder water- Lindenbergh -FeClSO ₄ - 8 g/m ³ -DAF, t=15 min -RSF, v=5 m/h	<i>Microcystis</i> spp. (Cyanophyta) <i>Oscillatoria</i> spp. <i>Stephanodiscus</i> spp. (Diatomeae)	Product water quality problems: ■ algae passing through treatment in large quantities, especially during periodical blooms	■ Complete change of present raw water source with one of better quality, possibly Andels Meuse

DAF - dissolved air flotation;

GAC - granular activated carbon

PAC - powder activated carbon

Nevertheless, their implementation on a wider scale and at different circumstances is not always feasible. It has been argued that the discussed chlorophyll-*a* guide level is too stringent in view of the very low assimilable organic carbon (AOC) value associated with it [31]. Namely, the proposed 0.1 µg/L chlorophyll-*a* guideline value should imply 3 µg/L AOC, which is lower than the already ambitious recommended 10 µg/L AOC required for biologically stable water (proposed by van der Kooij [32]).

Research in the past two decades was mostly directed at optimising conventional treatment processes, commonly centred around sedimentation and filtration. Agglomeration (coagulation/flocculation) has received the largest attention, as it is the prerequisite for any efficient particles (and algae) removal regardless of the subsequent down-stream removal process (sedimentation, flotation or filtration). Efficient water conditioning with the purpose of improving the coagulation/flocculation process was approached in a similar manner. However, it was soon realised that it is typically impossible to achieve adequate algae removal when they are present in concentrations that are typical for some eutrophic reservoirs (periodically up to 10^5 - 10^6 cells/mL). Obtained removal rates strongly depend on prevailing plankton type and range from 50-99.9%. However, even a rate of 99.9% removal can still be inadequate for raw water of such poor quality [29].

1.3.2 Dissolved air flotation (DAF) - a viable solution option

Because of the natural tendency of algae to float dissolved air flotation (DAF) attracts attention as a (pre)treatment option prior to final filtration. Because DAF requires a preceding particle agglomeration (coagulation/flocculation) step, it performs equally well as common flocculation/sedimentation with respect to DOC, UV, true colour and trihalomethane precursor removal [33, 34]. On the other hand DAF tends to be more efficient than sedimentation for removal of turbidity and particulates formed by coagulation [35]. Experience from newly built flotation facilities for drinking water treatment (e.g. Birmingham Frankley, UK; Millwood New Castle, USA; Pietarsaari, Finland), and from adapted existing plants, where e.g. sedimentation was replaced by flotation followed by rapid sand filtration document this [36, 37]. It is generally confirmed that algal removal efficiency can be substantially enhanced by flocculation and flotation upstream of the final filtration. The same applies for algae conditioning, i.e. bringing the algae into a state which results in their more efficient down-stream process removal, by application of oxidants, e.g. ozone [38], or KMnO_4 [39].

Janssens [40] has proposed a tentative diagram to be used for process selection based on raw water turbidity and algal content (Fig.1.7). If all other quality requirements are fulfilled regarding DOC, hardness, colour, etc., flotation is the best available (pre)treatment technology for treatment of raw water with chlorophyll-*a* values of typically > 10 µg/L. This is the case in all of the five studied companies for shorter or longer periods of the year (Table 1.1). In addition, under the Dutch circumstances the occurrence of *M. aeruginosa* (Table 1.4) favours use of DAF, because this species is characterised by buoyancy [3], which renders them more susceptible to removal by the rising air bubbles in DAF.

Fig. 1.7 Typical selection diagram for a water treatment process based on turbidity and chlorophyll-*a* concentration (from Janssens [40]).

Although this diagram can be very useful, it does not consider a range of factors which may be of substantial impact on the choice of process technology. Such factors include algae morphology and physiology (different species exhibit different response to treatment) [41], the source and character of turbidity, temperature, colloidal and particulate matter concentration, and possibly other factors as well. Furthermore, the choice of technology is largely a matter of cost-benefit analysis, suggesting that this diagram can be considered only as indicative.

A typical layout of a DAF treatment plant is presented in Fig. 1.8. It consists of a tank in which two zones are distinguished : (i) the contact zone, and (ii) the separation zone.

Fig. 1.8 Schematic presentation of a DAF treatment plant [42].

In DAF, air is introduced into the contact zone via a pressurised (approximately 500 kPa) recycled stream of treated water (5-10% of total flow). The change of pressure from e.g. 500 kPa to atmospheric results in the production of numerous microscopic (10-100 μm) air bubbles which attach to the particles (flocs). The commonly large ratio between air bubble and particle concentration of 10^2 - 10^3 , encountered in normal surface water treatment, is suggested responsible for the high DAF efficiency [42]. The particle-bubble agglomerates rise to the surface in the separation zone, where they are easily skimmed off as a light foam.

The following advantages of DAF are proposed [43]:

- small surface needed for unit and equipment;
- low civil engineering cost requirements;
- typical reduction of 10-20% in chemicals consumption as compared to conventional sedimentation;
- high dry solids content of the sludge (4-6%), compared to sedimentation (2-4%);
- lower water losses (1%, compared to 3% for sedimentation) due to production of sludge with a higher solids content;
- less affected by flow variations;
- suitable for intermittent use, reaching steady quality after approximately 60 min;
- operates in cold climates with minimum flotation time less than 10 min, and less sensitive to temperature variations than sedimentation;
- high algae removal efficiency.

Disadvantages include higher operational costs and the requirement to have skilled operating personnel. Nevertheless, for treating water with high algal count and relatively low turbidity DAF appears to be the most cost-effective technology. Water with high turbidity, on the other hand, is also treatable to a certain limit and with appropriate process modifications.

1.3.3 Agglomeration (coagulation/flocculation) of algae

The single collector collision efficiency DAF kinetic model proposed by Malley and Edzwald [45] recognises two sets of variables that define process efficiency :

- pre-treatment variables ahead of the flotation tank, which influence the particle-bubble attachment efficiency, the particle concentration and the particle-bubble collision efficiency;
- design and operation variables within the flotation tank, which influence the bubble size and concentration.

The agglomeration (coagulation/flocculation) step is a critical part of the DAF; it therefore deserves to receive priority in comprehensive research approach. Such comprehensive optimisation should take into account the down-stream filtration. Neither conventional sedimentation, nor advanced DAF are able to remove on their own the number of algae encountered in eutrophied surface water. Model calculations suggest that the particle destabilization degree is more important than the floc size [42]. pH and coagulant dose that

produce floc particles with little or no charge, generally result in favourable flotation conditions. Particles (algae) must be destabilized for good particle attachment to bubbles, as algae stability is caused mainly by : (i) the electrostatic repulsive force due to the negative surface charge (at pH 2.5-11.5), and (ii) the steric hindrance due to the water layer associated with hydrophilic surface and the adsorbed EOM at the cell surface [42]. Bernhardt and Clasen [45] conclude that algal cells can be considered to behave as inert particles and that the classical coagulation theory therefore is applicable; algae could be destabilized, aggregated and thus brought into filterable form, by adsorption coagulation or sweep coagulation. This can be achieved by charge neutralization by means of adsorption of positively charged organic or inorganic polyelectrolytes, or by metal hydrolysates when sweep coagulation is applicable. Agglomeration conditions that produce particles with little or no charge generally result in efficient DAF [42]. The selection of inorganic or organic coagulants as sole coagulants or coagulant aids, pH, and other coagulation variables therefore need further study.

The kinetic variables (G and t values) for efficient mixing and coagulation, as well as flocculation (i.e. creating particle collision opportunities) similarly need to be optimised. Unlike mixing and coagulation which take place almost instantly after coagulant addition (floc formation starts within 10^{-2} s up to a few seconds), the floc formation requires lower mixing energy input (low G) during longer time periods (5-20 min), enabling particles to collide. In practice, floc formation and floc break-up occur simultaneously, making application of mechanistic models complicated. However, floc size and strength determine overall efficiency in the floc removal step. While efficient sedimentation requires larger, heavier and easily settleable flocs, the floc size of 10 μm to few tens of micrometer has been shown to be optimal for flotation [42, 44]; similar to filtration, flotation efficiency worsens for particle size of 1 μm and is one to two orders of magnitude lower for particles $< 1 \mu\text{m}$. Floc growth spurs initially by the presence of small but numerous primary particles, and thereafter it is determined by the collision of 'micro-aggregates'. In case of adsorption coagulation, as floc size increases, the number of polyelectrolyte bridges per particle decreases proportionally to floc size and the floc becomes weaker and more susceptible to shear by hydrodynamic forces.

Algal cell aggregates in particular appear to be susceptible to shear and hence turbulence. A shear -free transport of flocculated algae to further treatment is required. A critical location in DAF is the injection of the recirculated water through the injection nozzles or needle valves. Depending on the configuration of the DAF unit, flocs may be locally subjected to high shear. This suggests that longer flocculation times to produce larger flocs are questionable, if flocs break up anyway near the nozzles. Some authors [35] conclude that the flocculation time therefore does not affect flotation performance (based on turbidity measurements); good results can be obtained without flocculation or slow mixing, with flocs formation limited to the rapid mixing period of about 2 minutes. Others [46] suggest minimum flocculation time of 5-6 minutes. On the other hand, flocculation time e.g. at the Scheveningen flotation plant of DZH is in the range of 20-40 min, while in the Lindenberg flotation plant of EWR it is 25 min.

Optimal agglomeration finally depends very much on the dominant algal species. Cyanobacteria appear in different sizes, shapes and forms, and differ in their EOM composition, which is known to influence the agglomeration process to a great extent. It can be expected that different species and seasonal conditions require different sets of optimal

agglomeration conditions for subsequent flotation. This calls for further study of the influence of morphological and physiological characteristics of alga species on the agglomeration and down-stream removal efficiency.

Particle (floc) characterisation such as automated particle count or computer image analysis becomes an essential tool in the assessment of the coagulation/flocculation and DAF process assessment.

1.3.4 Conditioning of algae

Recent advances in DAF operation pertain to the decrease of the flocculation and flotation time, and other process improvements related to the critical agglomeration (coagulation/flocculation) step. In addition, it was found that oxidants may under certain conditions (depending on raw water quality, coagulant dosage, location of dosing, etc.) improve coagulation and flocculation and hence down-stream floc removal efficiency. The positive experiences, especially with ozone [38, 47, 48] and KMnO_4 [39] in direct filtration, warrant further research on these oxidants prior to aggregation and DAF.

Although their use is not yet widespread, the application of oxidants as flocculation aid is becoming a viable option. In this context, the term conditioning seems appropriate to express the effect of oxidants on the algae structure and the resulting flocculation benefits. Namely, in addition to disinfection, secondary benefits arise from their effect on the surface chemistry of colloidal and suspended particles. As a consequence ozone can cause microfloculation (spontaneous flocculation), and hence coagulant dose reduction, improved water quality (measured as turbidity), and reduction in filter bed head-loss [47]. The mechanism of the induced flocculation is not yet fully understood; several mechanisms are proposed by Langlais et al. [49]: (i) reactions with algae, (ii) reduction of particle stability due to loss of adsorbed organic matter from natural particles, (iii) polymerization of natural organic matter (NOM), and (iv) break-up of metal-organic complexes yielding in situ iron production of metal coagulant.

The commonly recommended oxidant in DAF has been ozone, whether as a classical conditioner or for ozoflotation. Ozone reacts with the algal outer cells layer (EOM) and the algal cells themselves; the extent of oxidation depends on the algal species, making its effect seasonally dependent [38, 47]. Similarly, overdosing can break-up colonies, and damage cell structure. As one consequence, DOC and AOC levels may increase, as well as the toxins concentration [50, 51]. The optimization of the ozone dose for efficient aggregation and flotation, and the understanding of the process kinetics require further study. As an alternative KMnO_4 is not commonly applied as a disinfectant and little is known about its optimal use. Its efficiency as a conditioner is related to its oxidative activity and the coagulating effect of the hydrous manganese dioxide floc which is formed when the permanganate is reduced [39, 52].

Physical conditioning commonly consists of straining and its efficiency depends on algal size and shape. Microstraining is widely applied with mesh size usually in the range of 35 μm . Mesh sizes as low as 5 μm are commercially available, their application depends largely on

local raw water plankton and suspended matter concentration and composition. The experience in the five studied Dutch enterprises shows that microstrainers can substantially reduce the algal loads on the subsequent treatment step [19]. However, algae blooms often create problems in the operation of the microstrainers resulting in deteriorating effluent quality. Single cells of *Microcystis* spp. and *Oscillatoria* spp., although with distinctly different morphological features, have both been noted to pass through this treatment as in case of the WRK III and PWN treatment plant, causing increased algae counts in the final effluent. The current understanding of the role of this treatment step in the optimisation of the overall treatment scheme can benefit from further pilot studies.

Finally, the use of other physical conditioners such as UV light has not been considered to our knowledge. Its applicability as an efficient disinfectant is yet gaining increasing recognition [53]. UV light is not considered to be an efficient algicide as high dosages ($\sim 5000 \text{ J/m}^2$) and long exposure times (several minutes) are probably necessary. Yet, its effect on the viability of algae and hence on aggregation and down-stream flotation, should position it as a potentially interesting research topic.

1.4 CONCLUSIONS

The problems related to the periodical high concentrations of algae, are assessed for five Dutch surface water treatment locations. Cyanobacteria, especially *M. aeruginosa* and *O. agardhii* are recognised as the most significant problem causing algae. Examples of efficient reservoir management including deep reservoir operation (WRK III and PWN since recently), coagulation/flocculation and settling within the reservoir (WRK III and DZH), periodical destratification mixing (WRK III), phosphate removal by coagulation/flocculation and settling prior to the reservoir (GWA), can substantially diminish the algae load on subsequent treatment. Water treatment plant managers and operators should carefully consider such positive experiences and investigate their applicability and feasibility in their circumstances. Nevertheless, high algae concentrations, and especially blooms, can still cause numerous treatment problems. This forces water companies to adapt short-term and long-term process modifications, which raises production costs. This raises the issue of further cost-benefit algae treatment optimisation, as well as the viability and applicability of advanced treatment technologies. Increase of the knowledge basis on these issues should improve treatment process operation and raise the confidence in the application of new more efficient technologies. Existing algae related treatment problems and recent research results suggest that there is still a large operational window for increasing the efficiency of existing treatment capacities, based on the cost-benefit principle.

Considering the natural tendency of algae to float, and the positive experiences elsewhere, dissolved air flotation emerges as an attractive treatment technology, as alternative to conventional sedimentation. It features a high rate, high efficiency, low chemicals consumption, and high versatility and adaptability. On the other hand, discrepancies exist between literature and practice, which suggests that optimisation would benefit from DAF further research. The fact that in the nineties many treatment plants in Western Europe will require an increase in production capacity (in our study GWA, DZH and GWG) due to

increased water consumption, or to upgrade the process efficiency due to more stringent water quality regulations, compound this opinion. DAF is also an attractive option to be considered as pre-treatment in combination with emerging membrane technology in case of high algal loads.

Research in the Netherlands should preferably focus on the process optimization of a comprehensive DAF-based treatment scheme for the removal of cyanobacteria species, notably *Microcystis* spp. and *Oscillatoria* spp. An integrated approach is preferred for this purpose, which includes the up-stream agglomeration (coagulation/flocculation) of particles, as well as down-stream filtration. The emphasis is to be placed on the agglomeration phase which is considered critical, whether of particles (algae) in the coagulation/flocculation stage or of particles (algae) and bubbles in the DAF stage. The effects of pre-treatment and conditioning steps on overall process efficiency should be assessed. Positive and negative side effects of the treatment must be assessed, especially with regard to the level of DOC and its AOC fraction in the effluent, because of the possibility of cell disruption by the oxidants. The same applies to the toxin levels in the effluent, which are considered a second priority, but are more difficult to quantify. Finally, the influence of algal morphology and physiology on the process mechanisms need to be investigated.

The evaluation of the process efficiency should be related to the kinetic parameters and rely on more comprehensive particle characterisation. This should include comparison of particle (floc) size frequency and volume distribution for raw water, for water subject to aggregation (coagulation/flocculation), and for treated water. Bubble size characterisation (size distribution) and particle charge related phenomena are also to be considered. This would enable characterisation of process efficiency in a qualitative and quantitative manner, and further clarify important aspects of process kinetics.

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both in the smaller ($< 50 \mu\text{m}$) and larger ($> 50 \mu\text{m}$) particle size ranges. A low flocculation G value of 10 s^{-1} resulted in similar and better algae removal efficiency by DAF than the value of $G=70 \text{ s}^{-1}$, producing a floc size distribution with larger and lighter flocs, favouring efficient DAF. The interaction between the flocculation G and t values was found as the only significant factor affecting the DAF process efficiency, each of them separately having no statistically significant effects on removal efficiency. Thus, apart from the coagulation related charge effects, the particle (floc) size and density which are determined by the flocculation process, play a highly important role in determining the down-stream process efficiency. Under optimal process conditions for DAF and sedimentation no statistically significant efficiency difference was observed for the removal of particles (algae), while the achieved turbidity removal was significantly better by sedimentation than by DAF.

Application of relatively low doses of cationic polyelectrolyte (Superfloc C-573) as coagulant aid (0.5 for DAF and 1.0 mg Superfloc C-573/L for sedimentation) in combination with previously optimised coagulant doses and flocculation conditions, increased process efficiency of both processes (without down-stream filtration) up to 99%. Together with an improved removal efficiency achieved by coagulation at low pH (pH 6 vs. pH 8), this suggests adsorption coagulation and charge neutralisation are the main coagulation mechanisms that support the dosage dependant sweep coagulation and improve the overall particle (algae) removal.

Results from bench scale experiments with model water and laboratory cultured *M. aeruginosa* showed that sedimentation or DAF alone is insufficient to cope with moderate to high initial concentrations of this species. Maximum particle (algae) removal efficiency achieved under optimal process conditions was 71% for DAF (3 mg Fe(III)/L, $G=10$ or 70 s^{-1} , $t=10 \text{ min}$), and 87% for sedimentation (10 mg Fe(III)/L, $G=30 \text{ s}^{-1}$, $t=30 \text{ min}$). An integrated approach including the up-stream agglomeration (coagulation/flocculation) step, as well as the down-stream filtration step is essential. Adding a non-optimised filtration step in the experimental set-up increased removal efficiency in both cases, for DAF up to 90% and for sedimentation up to 95%. The filtration efficiency in this case (small scale filters connected to the jar test apparatus) was generally influenced by the flocculation G value and the resultant floc size/density ratio. It is also speculated that given the relatively low model water TOC of $\approx 2.5 \text{ mg/L}$, the residual coagulant (after DAF or sedimentation) is utilised in the filter in a "direct filtration" mode. Thus, the relatively low residual coagulant concentrations (from the point of view of direct filtration) result in relatively low filtration efficiencies. The filtration efficiency itself increased with the residual coagulant increase. Additionally, this situation may be negatively influenced by the (small) scale of the filters - filter wall effects and the non-ripened state of the filter, a situation which will be improved if a fully optimised filtration step is applied.

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APPENDIX 3.1 Composition of the algae cultivating medium (Jaworski) used for semi-continuous culturing of *M. aeruginosa*).

ELEMENTS :	CONCENTRATION (µM) :	TRACE ELEMENTS :	CONCENTRATION (µM) :
Ca	84.67	B	40.0
N	1038.00	Mn	7.00
K	91.10	Fe	5.60
P	191.70	Mo	0.81
Mg	203.00		
S	203.00		
C	200.00		
Na	1213.00		

VITAMINS :	CONCENTRATION (mg/L) :	COMPLEXING AGENTS:	CONCENTRATION (µM) :
B-12	0.04	EDTA Fe Na	5.60
B-2	0.04	EDTA Na	6.04
Biotin	0.04		

CULTURING PROCEDURE : The nutrients were added into 1 L of demineralised water at pH 7, followed by autoclave for a period of 20 min at 120°C. The vitamins solution which had been previously sterilised by filtration at pH 7, was added to the medium afterwards. The incubation of algae (80 mL of *Microcystis aeruginosa* suspension at the end of the logarithmic growth phase) was done in 5 L glass vessels (Duran 50, Schott Mainz, Jena, Germany) at 20°C. The algae suspension was aerated with compressed air passed through a sterile 0.2 µm filter, while continuous mixing was applied at the bottom of the vessel with a magnetic stirrer. The light cycle was continuous at light intensity of ± 1,000 lux. For this purpose a vertically placed T.L. lamp Osram-Cool White L18 W20 was used, placed at a distance of 0.2 m from the walls of the vessel. The stationary algae growth phase was reached after an incubation period of 14-17 days, after which 2 L of the algae suspension were sampled in sterile conditions and used in experiments. The abstracted 2 L of algae suspension were substituted with fresh medium and incubated under the same conditions for 7-10 days, after which algae were sampled for experiments again. This completed the culturing cycle, after which a new culture was started-up.

coagulant alone. The KMnO_4 conditioning option proved significantly less efficient than the ozone conditioning, especially in terms of particle removal. The relatively high standard deviation of the efficiency results, as well as the occasional change of the permanganate demand, suggested that the organic matter concentration and composition, play an important role in defining the permanganate demand, its chemical effect and the resulting DAF efficiency. The optimal flocculation Gt range was related to the flocculation and removal of the colloidal MnO_2 and was 60,000 (typically $G=70 \text{ s}^{-1}$, $t=15 \text{ min}$).

The Mn and coagulant concentrations in the DAF effluent at the optimal permanganate dose were higher than their MAC values. This suggested that cationic polyelectrolytes could reduce the electrostatic repulsion between the oppositely charged Mn and Fe colloids. The DAF particle and turbidity removal efficiency in the case of cationic polyelectrolyte Superfloc C-573 coagulant aid application resulted in significantly better efficiency than was achieved in the case of iron coagulant only, or in the case of the combination of KMnO_4 conditioning and iron coagulant. A particle and turbidity removal efficiency of 95% and 86% respectively, were achieved with 0.5 mg Superfloc C-573 as coagulant aid, applied in combination with permanganate and Fe(III) coagulant. A 5-25% improvement of turbidity removal efficiency was also noted in comparison with the iron coagulant and cationic polyelectrolyte combination. The observed improvement was achieved via improved removal of both the Fe and Mn residuals. The option of lowering the residual Mn by the reducing agent Na_2SO_3 proved potentially viable. In any case, other studies proved that residual MnO_2 can be efficiently removed in a final filtration step.

Appendix 4.2 synthesizes the DAF particle and turbidity removal efficiency achieved under different treatment combinations.

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size fraction resulted in a relative floc density decrease and a consecutive increase in DAF efficiency. While for sedimentation the floc size - density relation directly affects the efficiency, bubble size and bubble concentration are an additional factor of influence for DAF. Particle and bubble characterisation can further allow calculation of the floc size - density ratio (ρ) and the absolute floc density (ρ_d), important parameters in DAF and sedimentation model equations.

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negligible or even adverse effects on process efficiency.

The single collector collision efficiency DAF kinetic model can be a valuable tool in the design of DAF plant facilities, as well as for DAF experimental design, since it defines a range of coagulation/flocculation and DAF process parameters that affect the DAF process efficiency. Physical verification of the model, however, largely relies on the accurate prediction of α_{pb} . Determination of this lump coefficient largely depends on the coagulation/flocculation process. The role of raw water quality factors affecting the coagulation/flocculation and DAF process efficiency, such as NOM presence, concentration and composition, the presence of surface active agents, etc. must also be considered. The modelling assumption of treating particle-bubble collision as the only relevant mechanism for their aggregation, while neglecting other proposed mechanisms such as entrapment of bubbles within a condensing network of flocs and growth of bubbles from nuclei within flocs is not fully justified. However, these mechanisms are complex and difficult to model, in view of their dependence of the water quality.

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