



OZONATION AND BIOFILTRATION IN WATER TREATMENT

*OPERATIONAL STATUS AND
OPTIMIZATION ISSUES*

TECHNEAU

WP5.3 Operation of water treatment facilities – Optimization efforts and modelling of unit processes



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Title

OZONATION AND BIOFILTRATION IN WATER
TREATMENT - OPERATIONAL STATUS AND
OPTIMIZATION ISSUES

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Deliverable number

D 5.3.1 B

This report is:
PU = Public

Summary

Due to its low maintenance costs and effective removal of biodegradable organic matter, ozonation - biofiltration is becoming an attractive drinking water treatment method. Biofiltration is expected to become even more common in the future as efforts intensify to decrease the presence of disease-causing microorganisms and disinfection by-products in drinking water, to minimize microbial regrowth potential in distribution systems, and where operator skill levels are emphasized. This report covers operational aspects of ozonation-biofiltration processes, with special emphasis on NOM removal,

After introduction to the aim and scope, the descriptions of terms and definitions used in the report are listed (**Chapter 1**). Raw water characterisation and treatability by biofiltration, i.e. available characterisation methods and known links between raw water characteristics and treatability (AOC, BDOC etc) are described in **Chapter 2**. Operation practices are reviewed from available operational experience, based on literature and communications with existing WTPs (**Chapter 3**). Major operational challenges and bottlenecks are reviewed in **Chapter 4** including such problems as low temperature, nutrient limitation, oxygen supersaturation, presence of invertebrates and high concentration of microorganism at the biofilter effluent. Furthermore, the report is addressing pre- and post treatments (**Chapter 5**), process control and automation (**Chapter 6**), unit process operation modelling (**Chapter 7**), and treated water characterization (**Chapter 8**). Case studies at existing plants are included, at sites in the Netherlands, Finland, Norway, Germany and Latvia (**Chapter 9**). Finally, the knowledge gaps and research needs are listed (**Chapter 10**) as inputs for the design and performance of the pilot and full scale investigations.

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INTRODUCTION

1.1 Optimum operation of water treatment facilities – the best operation practices

Sub-optimal operation of water treatment facilities may seriously compromise safety, sustainability and cost-efficiency of water supply systems. Even more serious, it may compromise public health.

There is reason to believe that non-optimal operation of water treatment facilities is relatively widespread phenomenon also in Europe. The reasons for this may be several: missing focus on operational issues and challenges in general, lack of driving forces for optimization efforts, non-optimal treatment technologies and improper process design, a large number of small scale water supply systems with limited resources, under-reporting of water-borne diseases, rapid changes in raw water quality and correspondent operational difficulties, lack of robust and reliable sensors for process control purposes, lack of – or too complicated - unit process models, more long-term effects of climate change on raw water quality and quantity, e.g. increasing NOM, greater quality variations due to more severe droughts and floods, lack of operational competence and knowledge on optimization potentials and possibilities, etc.

1.2 NOM removal and biofiltration

Natural organic matter (NOM) in water is a major concern and should be removed from drinking water for a number of reasons, including that NOM:

- affects organoleptic properties of water (colour, taste and odour);
- reacts with most disinfectants used in water treatment;
- influences disinfectant demand, and disinfection process design, operation and maintenance;
- can produce disinfection by-products (DBPs) of various kinds;
- affects stability and removal of inorganic particles;
- influences heavily on coagulant demand;
- may control coagulation conditions and coagulation performance;
- affects corrosion processes;
- affects biostability and biological regrowth in distribution systems
- may form complexes with and increase mobility of most chemical substances found in nature and
- is responsible for fouling of membranes
- may block pores and thereby reduce PAC/GAC adsorption capacity
- may compete with taste and odour compounds for adsorption sites in PAC/GAC filters

NOM of microbial, animal and vegetable origin in reservoir catchments is the key factor influencing most, if not all, water treatment processes. The

character of the NOM and its involvement in water treatment processes requires greater understanding for determination of improved removal processes and interactions with other water contaminants.

Four principal methods for removal of NOM are chemical coagulation, anion exchange, nanomembrane filtration, and granular activated carbon (GAC) filtration. During last 20 years GAC is being replaced with ozonation-biofiltration process (OBP). In this process, instead of using adsorptive properties of GAC, the microbes living in the media are employed. Biological processes offer a potentially useful alternative because regular regeneration of GAC is not required and biodegradable organic matter (BOM) is preferably eliminated (Servais et al 1991).

In recent years, attention has focused on the carbonaceous organic matter that can be used by microorganisms as source of nutrients. This fraction of DOC is called biodegradable organic matter (BOM). This fraction is favourably used by bacteria as substrate promoting their growth. The growth of bacteria in distribution networks is not desirable because some of these bacteria can be potentially pathogenic for humans (e.g. *Legionella* and *Mycobacterium avium*). Bacterial growth in the network is mainly occurring in biofilm on surface of pipes. The biofilm can be environment where invertebrates can develop and pathogens are protected from disinfectant effect. To reduce these problems the BOM levels in drinking water leaving the treatment plant should be as low as possible. Water in which bacterial cells are not multiplying is called biologically stable.

1.3 Report objective and scope

The principle of biofiltration is to utilize bacteria and protozoa, which are attached to the surface of filter materials, to clean water from undesirable substance in drinking water. The biofilters are used for removal of BOM, iron, manganese, sulfate, nitrate, pesticides (Boley et al, 2006), taste and odor causing substances and algal metabolites (Nerenberg et al, 2000). This report deals mostly with the application of biofilters for the removal of natural organic matter (NOM). The report concentrates on rapid biofiltration processes. Other drinking water processes which are classified as biological, like slow sand filtration and artificial ground water recharge are not considered here.

In Europe, the application of biofiltration processes for the removal of natural organic matter has a long tradition which goes back to the 1970s. It is very often applied in Germany, France, and The Netherlands. In North America this process has gained special attention since the late 1980s. The biofilter removes biodegradable organic matter (BDOC) and therefore increase the biostability of the water and reduce the risk of DBP formation in the following chlorination process. The process is often combined with ozonation (thereafter ozonation-biofiltration) and actually, the use of ozonation requires biofilters since ozonation increases the amount of biodegradable matter in the

water. Often this biofiltration process is referred as Biologically Activated Carbon (BAC) filtration. In this report we will use the term ozonation-biofiltration processes (OBP) to emphasize the need for application of ozone before filtration.

The main objective of this report is to identify optimization potentials and the best operation practices of ozonation-biofiltration facilities for drinking water treatment, with emphasis on the removal of NOM. The main focus will be on the existing water treatment facilities.

The OBP are used for the removal of NOM from groundwater as well as surface water. In groundwater and small surface water treatment plants, ozonation-biofiltration may be used as the only unit processes. In larger plants, which are using polluted surface water, it is common that OBP are only parts of the unit processes used in the plant. Commonly, the process is combined with coagulation and particle separation (Figure 1.1). The process often includes pre-ozonation to provide primary disinfection and also to aid coagulation/flocculation in a following particle separation step. It has been reported however, that ozonation prior to coagulation may increase residual coagulant concentrations and reduce the removal efficiency of the humic fractions of NOM.

After coagulation/flocculation and sedimentation or filtration or both, an intermediate ozonation is often applied for disinfection. This is usually followed by biofiltration where granular activated carbon is normally the filter media, and a disinfection step (UV, chlorination, chloramination) after the biological filtration in order to obtain additional disinfection and inactivate the heterotrophic bacteria present in the biofilter effluent. Some biological activity occurs also in rapid filters after sedimentation. However, usually they are not designed as biofilters and are out of the scope for this report.

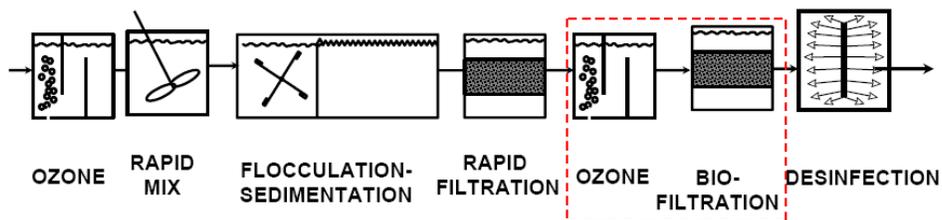


Figure 1.1 Commonly used ozonation-biofiltration units within coagulation chemical-sedimentation treatment plant train

Another well known process utilizing ozone and biofilters is the so called Mühlheim process (Bundermann, 2006) (Figure 1.2). This process includes artificial ground water recharge. This part of the process however, is not within the scope of this report.

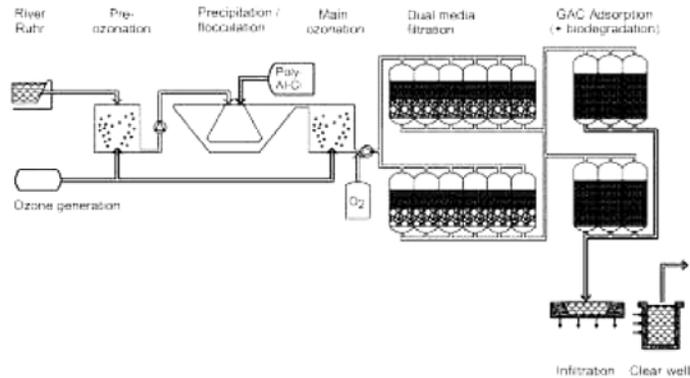


Figure 1.2 The Mülheim process: a typical process where biofiltration is taken advantage of in surface water treatment

Integration, e.g. linking raw water characteristics and water treatment, treatment process control, risk reduction, etc. is a major objective within the Techneau project. For the optimization efforts on biofiltration facilities it is therefore important to establish good links to other work areas, including climate change/changing raw water qualities, monitoring technologies, treatment technologies, risk assessment and risk management during water treatment and demonstration activities.

This report is based on literature review and consultation with operators from several water treatment plants. The major sources of information used for preparation of this report were scientific articles, textbooks, reports and proceedings from international conferences. Comprehensive literature review about biofiltration has been published by Urfer et al (1997), Gimbel et al, (2006) and Uhl (2000).

1.4 Terms and definitions

- AOC - Assimilable Organic Carbon** is part of organic carbon which is converted to biomass by specified bacteria or consortium of bacteria.
- ATP - Adenosine triphosphate**
- BDOC - Biologically Degradable Organic Carbon** is the part of DOC which is consumed by a community of natural bacteria in favourable conditions during a certain period of time (normally less than one month)
- Biofiltration** - A process in which water is filtered through media populated with microorganisms capable to treat water. Ozonation is usually used prior biofiltration and then the process is termed an **ozonation biofiltration process (OBP)**
- BOM - Biodegradable Organic Matter** is the fraction of DOC that could be converted by bacteria to energy or biomass

- CHA - Charged hydrophilics:** The fraction of NOM (DOC) that is retained by the anion exchange resin IRA-958
- Contact filtration:** A process where coagulated water is treated in a filter step without any separate flocculation, settling or flotation steps prior to the filter unit. Thus, the coagulation/destabilization processes will to a great extent occur within the filter bed in close contact with the filter media grains. Capture and storage of captured substances (NOM, particles, bacteria, viruses, protozoa, etc.) will have to take place in the filter bed. Because coagulant species like metal hydroxides may be effective adsorbents, particles, micro organisms, NOM may adsorb to precipitated coagulant species and deposits in the filter bed. This may be an additional and relevant removal mechanism in contact filtration processes.
- Conventional filtration:** A process with coagulation, a separate flocculation step, and pre separation units (sedimentation or flotation) prior to the filtration step
- CTC - 5-cyano-2, 3-ditolyl tetrazolium chloride**
- Direct filtration:** A process with coagulation, and a separate flocculation step before the water is filtered directly without any settling or flotation steps prior to the filter unit
- DOC - Dissolved Organic Carbon:** The concentration of organic carbon in a water sample after 0.45 µm prefiltration
- DOM - Dissolved Organic Matter:** The total concentration of organic substances in a water sample after 0.45 µm prefiltration
- EBCT - Empty Bed Contact Time** is the theoretical contact time associated with the volume that is displaced by the bulk media in the filter. It is calculated as total volume of the filter bed divided by the water flow through the filter
- GAC - Granular Activated Carbon**
- HPI - Hydrophilic fraction:** The fraction of NOM (DOC) that is passing, i.e. not sorbed on XAD-8 or XAD-4
- HPO - Hydrophobic fraction:** The fraction of NOM (DOC) that is retained, i.e. sorbed on XAD-8
- HPON - Hydrophobic neutrals:** The fraction of NOM (DOC) that is retained and eluted from XAD-8 with organic solvents such as acetonitrile CH₃CN
- INT - 2-(*p*-iodo-phenyl)-3-(*p*-nitrophenyl)-*s*-phenyl tetrazolium chloride**
- NEUT - Neutral hydrophilics:** The fraction of NOM (DOC) that is passing, i.e. not sorbed on DAX-8, XAD-4 and IRA-958
- NOM - Natural Organic Matter** is a chemically complex and heterogeneous mixture of organic substances produced from

vegetative decay processes. NOM is a ubiquitous constituent of all drinking waters, and is known to control the coagulation process in many cases. NOM may also interfere with adsorption and disinfection processes and cause fouling of membranes.

OBP - Ozonation-Biodegradation Process: water treatment process in which ozonation is combined with rapid filtration through media populated with bacteria.

Particle counts: is a measure of particle number and (and size) in water sample (or other liquid or gas). Particle counts are determined by (Liquid) Particle Counter. Three methods are commonly used for detecting and measuring particles (though many exist); Light Blocking, Light Scattering and the Coulter principle. Most commonly used is the Coulter principle that states that particles pulled through an orifice, concurrent with an electrical current, produce a change in impedance that is proportional to the size of the particle traversing the orifice.

RDOC - Refractory dissolved organic carbon: The fraction of DOC which are not degraded by bacteria

SEC - Size exclusion chromatography

SHA - Slightly hydrophobic acids: The fraction of NOM (DOC) that is retained, i.e. adsorbed by XAD-4

SMPs: Soluble Microbial Products are the by-products of biodegradable organic matter metabolism released by the biomass attached to the filter media.

SUVA (or SUVA₂₅₄): Specific UV Absorbance: UV absorbance at 254 nm (1/m) divided by the concentration of dissolved organic carbon (mg C/L). The unit of SUVA is commonly expressed as L mgC⁻¹ m. TSUVA is defined similarly, with DOC replaced by TOC.

TPH - Transphilic fraction: The fraction of NOM (DOC) that is passing XAD-8, but retained, i.e. sorbed on XAD-4

TPHA- Transphilic acids: The fraction of the TPH that is eluted from XAD-4 with NaOH at pH 13

TPHN- Transphilic neutrals: The fraction of TPH that is eluted from XAD-4 with organic solvents such as acetonitrile CH₃CN

Turbidity: Turbidity is cloudiness or haziness of water (or other fluid) caused by individual particles (suspended solids and colloids) that are generally invisible to the naked eye. Turbidity standard is measured in nephelometric turbidity units (NTUs). Turbidity is usually measured using an optical instrument in a laboratory called a nephelometric turbidimeter. The term Nephelometric refers to the way the

instrument estimates how light is scattered by suspended particulate material in the water.

VHA - Very hydrophobic acids: The fraction of NOM (DOC) that is retained, i.e. adsorbed by DAX-8

XAD (4 or 8): The trade names of a series of proprietary resins (manufactured by Rohm and Haas Corp.), which are useful for the characterization/fractionation of (natural) organic matter.

2 RAW WATER CHARACTERISTICS AND TREATABILITY

In this chapter available characterisation methods for NOM and known links between raw water characteristics and treatability by ozonation and biofiltration are described.

2.1 Natural organic matter (NOM)

Natural organic matter (NOM) in water contains a heterotrophic mixture of hydrophobic and hydrophilic organic compounds with a wide range of molecular weights, including non-homogeneous organic compounds such as humic substances (HS), amino acids, sugars, aliphatic and aromatic acids. NOM can be broadly divided into two fractions: humic substances (HS) and non-humic substances (non-HS), which include carbohydrates, lipids, and amino acids. HS are considered resistant to bacterial degradation, whereas non-humic substances are biodegradable and often referred as biodegradable organic matter (BOM). NOM is divided in dissolved organic carbon (DOC) and particulate organic carbon (PAC). DOC is defined operationally as material that passes a 0.2- or 0.45- μm filter. DOC consists of truly dissolved substances and macromolecules with colloid-like properties (e.g. HS). PAC is defined as material that is captured by a 0.2- or 0.45- μm filter. PAC consists of larger particles (e.g. algae, bacteria, particulate detritus, and organic matter within flocs). In addition, PAC includes inorganic particles covered by NOM.

Due to the great adsorbing and complexing abilities of NOM, it may be illustrated as a “vacuum cleaner” (Figure 2.1). This figure illustrates well the ability of NOM to mobilize and increase availability and exposure of micro-pollutants to human beings.

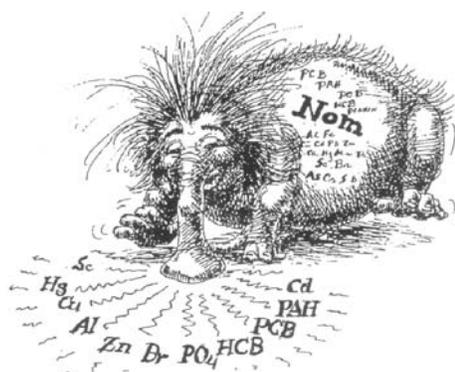


Figure 2.1 Illustration of NOM as a ‘vacuum cleaner’, due to the ability to act as a complexing agent/adsorbent for a large number of elements and substances.

The International Humic Substance Society (IHSS) has proposed a number of different NOM configurations, as illustrated in Figure 2.2. The cartoons

illustrate well the complex nature of NOM and the different understandings of NOM structure and configuration.

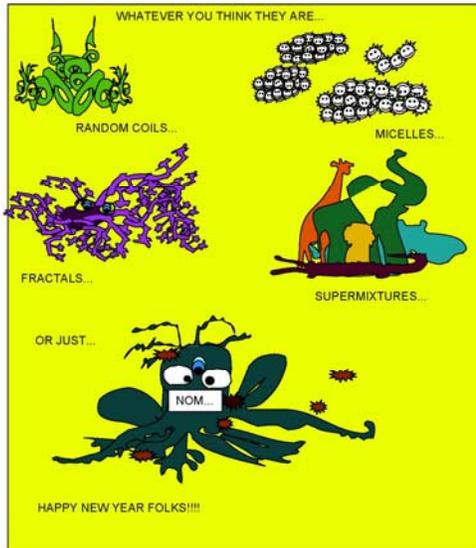


Figure 2.2 Proposed NOM configurations (IHSS, Newsletter, 2006).

2.2 Colour and UV-absorption

Visible and ultraviolet absorbance has been widely used to characterize raw waters in general, Because of the good correlation to dissolved organic carbon, colour and UV-absorption (UV-abs) are also used as surrogate parameters to DOC.

2.3 TOC and DOC

Total and dissolved organic carbon is measured indirectly from the CO_2 produced by UV-oxidation or combustion of the organic matter in a water sample.

2.4 Specific UV-absorption (SUVA)

Specific UV absorbance (SUVA or SUVA_{254}), is defined as the UV absorbance at 254 nm (1/m) divided by the concentration of dissolved organic carbon (mg C/L). The unit of SUVA is commonly expressed as L/mgC m.

Specific UV-absorption (SUVA) correlates well with the aromaticity and the hydrophobicity of the organic carbon. High hydrophobicity is associated with good treatability by coagulation. Therefore, SUVA can be used to indicate raw water treatability by coagulation and to predict the removal of organic carbon by coagulation.

2.5 Chemical and physical fractionation

Natural organic matter (NOM) may have distinctive characteristics associated with its origin (vegetation, soil, wastewater, agricultural return). For example, dissolved organic matter (DOM) from aquatic algae has a relatively large nitrogen content and low aromatic carbon and phenolic contents. On the other hand, terrestrially derived DOM has relatively low nitrogen content but large amounts of aromatic carbon and phenolic compounds. Thus, the aromatic content, which is believed to be a major reactive component, varies with different sources. The contribution of each carbon source is seasonally dependent, and the hydrological and biogeochemical processes involved in physical mixing and in the carbon cycles can alter the chemical composition and the physical structure of DOM.

Changing climatic conditions (e.g. global warming and more intensive rain events) may increase the volume of the DOM reservoir (e.g. increased amount of litter), the biodegradation rates and also the volumetric transport of DOC from a watershed to a water source. As a result, the concentration of NOM/DOM in drinking water sources may increase, in accordance with observations from Northern Europe and North America during the past 10-20 years.

From these reasons, physical and chemical fractionation of NOM/DOM may yield valuable information on characteristics and treatability. In a review paper, Chow et al (2005) present an overview of chemical and physical fractionation of DOC.

2.5.1 XAD fractionation

This fractionation is based on differences in sorption efficiency of DOC on XAD resins under acid and base conditions. Three major fractions can be isolated by using XAD-8/XAD-4 resins (acidified 0.45 μm filtered water samples at pH 2) according to the scheme illustrated in Figure 2.3:

- 1) Hydrophobic fraction - HPO (sorbed on XAD-8)
- 2) Transphilic fraction - TPH (not sorbed on XAD-8, sorbed on XAD-4)
- 3) Hydrophilic fraction - HPI (not sorbed on XAD-8 or XAD-4)

The HPO fraction is also referred to as the humic fraction, while the TPH and HPI fractions are referred to as the non-humic fractions. The two fractions that are adsorbed on XAD-8 and XAD-4, i.e. the HPO and TPH fractions, can be further fractionated into acidic and neutral fractions by using different eluting solvents:

- 1) Hydrophobic acids (HPOA) are eluted from XAD-8 with NaOH at pH 13. This fraction can be further divided into humic acids (HA) and fulvic acids (FA) which are precipitated and soluble, respectively at pH 1
- 2) Transphilic acids (TPHA) are eluted from XAD-4 with NaOH at pH 13

- 3) Hydrophobic neutrals (HPON) are eluted from XAD-8 with organic solvents such as acetonitrile CH_3CN
- 4) Transphilic neutrals (TPHN) are eluted from XAD-4 with organic solvents such as acetonitrile CH_3CN

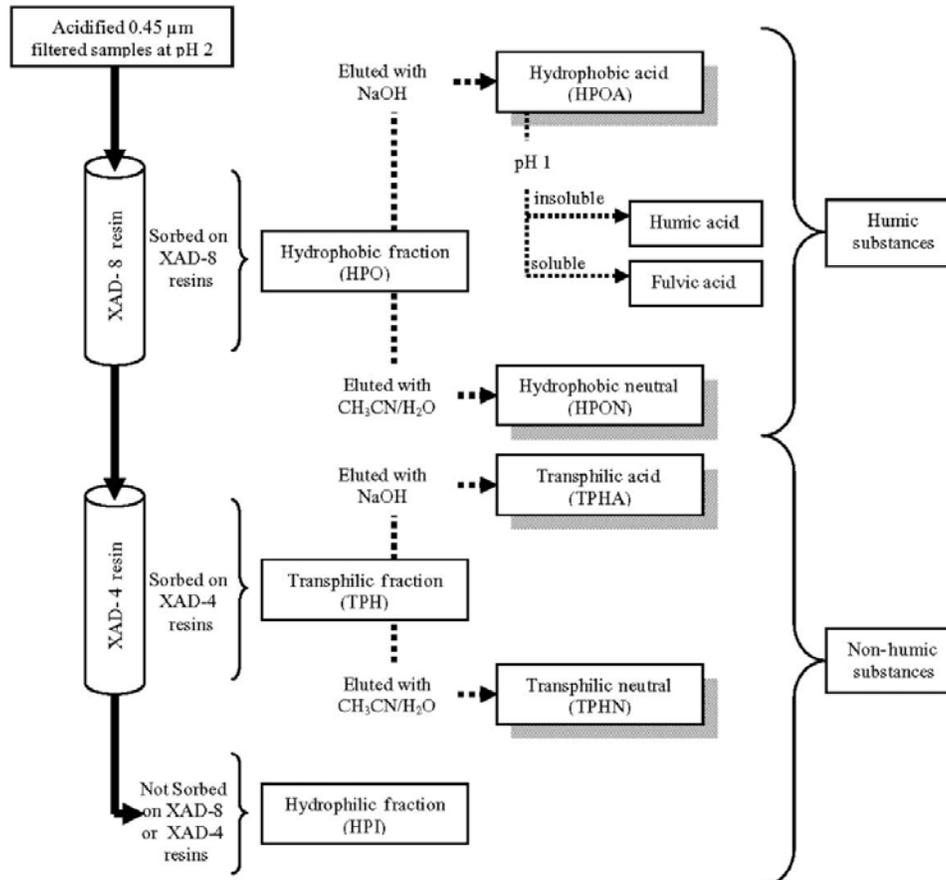


Figure 2.3. XAD-8/XAD-4 fractionation scheme (Chow et al 2005).

The HPOA fraction contains aliphatic carboxylic acids, aromatic acids and phenols, i.e. material derived from organic compounds in terrestrial plants and soils such as lignin, tannins and pigments that give rise to highly coloured humic and fulvic acids. This fraction accounts for more than 50% of DOC in most raw waters. In waters where terrestrial plants are the main source of dissolved organic matter (allochthonous sources), the humic (HPO) fraction can be as high as 80% of DOC. In contrast, in waters where the organic matter is derived from autochthonous sources like algae, bacteria and macrophytes, humic/HPO fractions as low as 33% have been detected.

The TPHA fraction is probably produced by autochthonous processes such as phytoplankton, bacteria, and microbiological degradation and will normally show significant seasonal variations. HPON consists of hydrocarbon-like

materials and amorphous polymers and appears to be a relatively stable and un-reactive fraction. TPHN seems to consist of proteins and appears nearly invariant with regard to source type (bog, lake, and river). It exhibits a strong seasonal trend.

The neutral fractions have higher C:O ratios, but lower C:N and C:H ratios than those of the acidic fractions, indicating a greater aliphatic character of the neutral fractions. The higher the proportions of oxygen, nitrogen and hydrogen, i.e. the lower the C:O, C:N and C:H ratios, the more hydrophilic and aliphatic the neutral and acid DOC fractions are.

Rapid Fractionation

A rapid and simplified fractionation technique based on measuring DOC concentrations before and after contact with the resins DAX-8, XAD-4 and IRA-958 was described by Chow et al (2000). This rapid characterization technique is specifically designed to study water treatment processes, and is based on the full-scale fractionation scheme. Based on subtractions of the DOC concentrations of subsequent resin effluents, the organic carbon concentrations of four fractions of NOM can be determined:

- Very hydrophobic acids, VHA (adsorbed by DAX-8)
- Slightly hydrophobic acids, SHA (adsorbed by XAD-4)
- Charged hydrophilics, CHA (bound to the anion exchange resin IRA-958)
- Neutral hydrophilics, NEUT (passed through all columns)

The details of the DOC calculations are presented below (Chow et al 2004):

$$\begin{aligned} \text{VHA} &= \text{Raw} - (\text{DAX-8 effluent}) \\ \text{SHA} &= (\text{DAX-8 effluent}) - (\text{XAD-4 effluent}) \\ \text{CHA} &= (\text{XAD-4 effluent}) - (\text{IRA-958 effluent}) \\ \text{NEUT} &= (\text{IRA-958 effluent}) \end{aligned}$$

Results can be presented as actual DOC concentrations of each fraction or as a relative percentage. VHA and SHA is predominantly composed of higher molecular weight humic and fulvic acids, CHA is ascribed to proteins, amino acids and anionic polysaccharides, and the NEUT fraction is ascribed to carbohydrates, aldehydes, ketones and alcohols. These are typically small molecular weight components such as polysaccharides and proteins and are often indicative of biologically derived material. Specifics of the technique and definitions have been described elsewhere (Chow et al 2004).

Fabris et al (2006) stated that the rapid fractionation technique could also be used to identify situations where treatment was not effective for DOC removal, either due to lack of optimisation or problems with normal operation.

2.5.2 Ultrafiltration and size exclusion chromatography

Particulate organic matter ($>0.45 \mu\text{m}$), such as zooplankton, algae and detritus is usually a small fraction of the total organic matter (TOM) in drinking water sources. Dissolved organic matter ($<0.45 \mu\text{m}$, i.e. DOM) is predominant in most water sources, and contains humic substances (HS) as a major class of organic compounds, viruses, amino acids, carbohydrates, fatty acids and hydrocarbons. Most studies of molecular weight fractionation focus on HS. The average molecular size of aquatic HS varies within the range of about 0.47-3.3 nm in the radius of gyration, corresponding to an apparent molecular weight (AMW) of 0.5-10 kDa.

Molecular weight fractionation is commonly achieved by ultrafiltration (UF) or size exclusion chromatography (SEC). Unlike XAD-8 and XAD-4 fractionation, UF or SEC does not require acids, bases or other types of chemicals. The alteration is minimal and the nature of DOM is preserved.

SEC can be supplied with on-line detectors (UV, fluorescence, DOC) for more specific investigations of NOM and NOM removal. However, UV is able to detect only a fraction of the NOM and is therefore not a preferred detection method for this application. The fractions collected by both UF and SEC are based on differences in molecular size rather than actual molecular weight, and the results are dependent on the operating conditions.

DOM is typically fractionated into four AMW ranges: <1 , 1-10, 10-30 and >30 kDa. In most source waters, DOM with AMW of 10 kDa or less is dominant. HS with an AMW above 10 kDa is uncommon.

Owen et al (1993) state that the 1-10, 10-30 and > 30 kDa fractions can be efficiently removed by coagulation and ozonation, while the fraction < 1 kDa is not-removable. It is likely that the small-size fractions of NOM are amenable to efficient removal by biofiltration.

2.6 Biodegradable organic matter (BOM)

BOM is mainly composed of non-humic substances, though lately it was proven that 10 to 20% of river and lake HS are biodegradable and thus also contribute to BOM. In the drinking water industry, two measurement-specific BOM subsets are widely used. Biologically degradable organic carbon (BDOC) is the fraction (10-20%) of the DOC that can be mineralised by heterotrophic bacteria (Servais et al, 1987), whereas assimilable organic carbon (AOC) is the fraction of DOC (1-10%) that can be converted to cell mass by either a single organism or a consortium of bacteria (van der Kooij et al, 1992). Another method to determine regrowth potential of bacteria in drinking water is the BRP method (biological regrowth potential). The BRP method is not primarily aimed at identifying the utilizable organic carbon concentrations, but more on the question how the bacteria originally present in the water sample can multiply.

2.6.1 Assimilable organic carbon (AOC)

This parameter was developed by van der Kooij (1992). It is based on culturing two bacterial strains (*Pseudomonas fluorescens* P-17 and *Spirillum* sp. Strain NOX) in the water under investigation and matching the maximum number of cells obtained with a calibration curve produced by using an easily assimilated nutrient such as sodium acetate. A value of no more than $10\mu\text{g L}^{-1}$ of AOC is recommended for biologically stable water. The AOC level is considered to indicate the quantity of carbon in a water sample that can easily be assimilated by bacteria. Lately several improvements which allow a significant decrease in the time requirement of the AOC analysis has been suggested (Hemmes and Egli, 2005)

2.6.2 Biological regrowth potential (BRP)

In the BRP method the sample is prepared by sterile filtration through $0.2\mu\text{m}$ pore size filters, and a nutrient salts medium is added. Bacterial proliferation is then monitored as turbidity at 12° forward scattering. From the curves of turbidity increase over time the growth rate is calculated by fitting a Monod type growth function. Usually a regrowth factor is given as a result, i.e. the turbidity plateau reached after some time is divided by the initial turbidity.

Sometimes the concentration of assimilated organic carbon is calculated from a regression of regrowth factors for water samples spiked with acetate. It is often claimed that an advantage of the BRP method is the use of a mixture of autochthonous bacteria which may be better adapted to the substrates than the specific strains applied in the AOC method. However, the amount of bacteria available from the sample may often be too small. Then bacteria from other environments (mostly from GAC filter effluents of drinking water treatment plants) may be added.

Disadvantages of the BRP method includes the fact that the inoculum needed is relatively large in comparison to the AOC method (about $5 \cdot 10^4$ cells mL^{-1}). Thus a large increase in bacterial concentration has to be found to be reflected in the regrowth factor. Furthermore, the risk of contamination of the sample with biodegradable carbon is higher during sterile filtration compared to pasteurization, and the equipment used is relatively expensive.

2.6.3 Biodegradable dissolved organic carbon (BDOC)

The sample to be analysed for BDOC is placed in contact with a native mixed biomass. Monitoring of DOC enables the degradation of the organic matter to be observed, with corresponding increase in carbon dioxide and bacterial cells. When the degradation has reached a plateau, the residual DOC value obtained is described as refractory dissolved organic carbon (RDOC). The difference between the initial DOC and the RDOC enables the BDOC to be calculated in mg L^{-1} . A 30 day incubation time is normally used. For faster results more biomass should be used in the experiments. This can be accomplished by adding sand with biomass to the sample or by filtration of

the water sample through a column (2 hours) in which stable biofilm has developed. It has been found that biologically stable waters contained less than 0.25 mg L^{-1} (Niquette et al. 2001).

The determination of total BOM is not always enough to predict removal of NOM in biofilters. The reason is that the degradation rates of DOC are often different for different waters even when the total BDOC is the same. The contact time in biofiltration is normally within the range of 5 to 20 minutes, while BDOC during the bioassay is determined over a time period of several days (the more biomass the less the length of the test). Thus, not all BDOC will be removed in the biofilter. Therefore kinetics of DOC degradation should be determined in order to predict biofilter performance. To address this problem some authors have suggested to classify BDOC in several fractions depending of their degradation rates (Carlson and Amy, 2001). Two classes are suggested: $\text{BDOC}_{\text{rapid}}$ is BDOC which is rapidly degraded (60 min) and $\text{BDOC}_{\text{slow}}$ is BDOC degraded over the period from 60 minutes to the end of the BDOC test (30 days). It was found that $\text{BDOC}_{\text{rapid}}$ is similar for several water sources, representing about 12% of DOC when a minimum ozone dose was applied ($> 1 \text{ mgO}_3 \text{ mgDOC}^{-1}$), while $\text{BDOC}_{\text{slow}}$ varied significantly from 5 to 25% depending on the water source. Most likely $\text{BDOC}_{\text{rapid}}$ will be removed in the biofilter while $\text{BDOC}_{\text{slow}}$ will enter the distribution network and will be slowly degraded by the bacteria living there.

Yavich et al (2004) using the same approach divided BDOC into 3 classes: "fast" BDOC, "slow" BDOC and "non" BDOC. Klevens *et al.* (1996) operationally defined the distinction between "fast" and "slow" BDOC by bisection the extreme tangent lines at the beginning and at the end of the biodegradation curve (Figure 2.4). From this mathematical interpretation the maximal rate of biodegradation of "fast" BDOC can be calculated, and the value was found to vary from 0.08 to $0.46 \text{ mgDOC L}^{-1} \text{ min}$ for ozonated water. Digiano *et al.* (2001) have developed method for the determination of the biodegradation kinetics of BDOC.

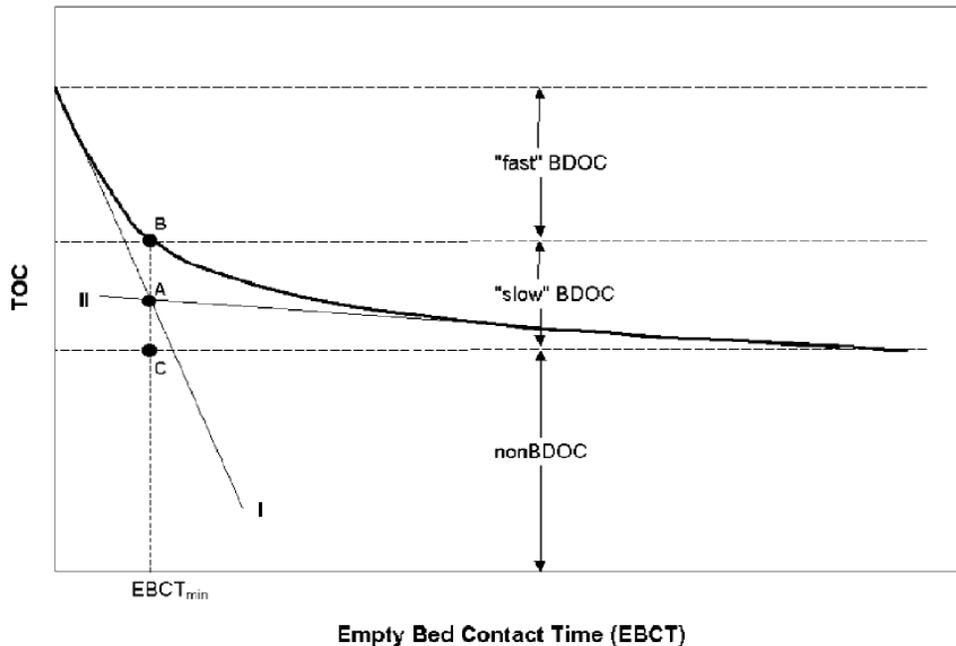


Figure 2.4 Illustration of an approach for the quantification of biodegradation rates of NOM fractions (Yavich et al 2004)

2.7 Treatability of NOM with OBP processes

2.7.1 Fractions of NOM in raw water removable by biofiltration

Ozonation-biofiltration processes will remove the biodegradable fractions of NOM. The hydrophilic NOM fractions (the high polarity and low molecular weight organic solutes) are more biodegradable and will be preferably removed by OBP. So, according to the rapid NOM fractionation procedure waters with high CHA and NEUT fractions will be more amenable for treatment by biofiltration.

Aromaticity correlates well with the specific UV absorbance (SUVA) High colour and UV adsorption indicate a high concentration of HS, which are difficult to remove with biofiltration unless pre-ozonation is applied. A SUVA of less than 2 L mgC⁻¹ m indicates that the water contains biodegradable fractions which are removable with biofiltration. One of the most important factors which regulate the microbial bioavailability of NOM is the molecular size, often less precisely expressed as the molecular weight. In general, the smaller the molecule the more rapidly it will be degraded by bacteria.

The most widely used analysis of NOM is AOC and BDOC analyses. AOC is compiled of small organic molecules therefore this fraction is effectively removed by biofiltration. BDOC represents a biodegradable pool of DOC,

however only rapidly degradable BDOC will be removed by biofiltration due to the relatively short contact time in biofilters. The removal of less biodegradable DOC can be enhanced in the presence of readily biodegradable fractions such as AOC.

DOC treatability by biofiltration depends on raw water characteristics, the conditions in the watershed, climatic conditions and season. The seasonal variation of raw water quality may influence TOC treatability and removal by ozonation-biofiltration processes. In the Croton Reservoir (New York) the amount of fast BDOC was lowest in the raw and ozonated water during winter and highest during fall (Klevens *et al.*, 1996). The relative amount of hydrophilic as well as low molecular weight compounds was higher during the warm season. These are the same fractions that increase during ozonation and are subsequently removed in the biofilters.

2.7.2 Effect of ozone on NOM treatability

When ozonation is used prior to biological filtration, and environmental conditions such as dissolved oxygen, pH and temperature are favourable, the microbiological activity is increased in the filter and the biodegradability of DOC is enhanced (Figure 2.5). Ozone addition not only increases the biodegradability of the dissolved organics, but also introduces large amounts of oxygen to the water, thus creating an excellent environment for biological growth on the filter media

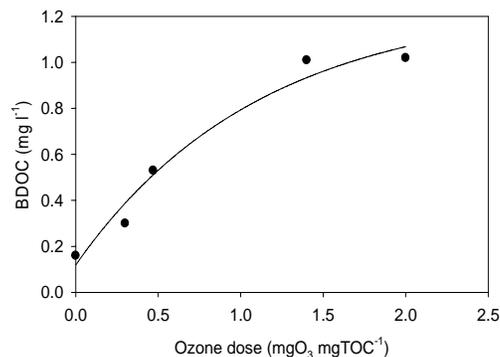


Figure 2.5. Effect of ozone dose on the formation of BDOC in humic surface water (Melin *et al.*, 2006).

Ozonation causes structural changes to NOM and in particularly to the humic fractions. The changes include a strong and rapid decrease in colour and UV-absorbance due to a loss of aromaticity and depolymerisation, a small reduction of TOC (e.g. 10% at 1 mgO₃ mgC⁻¹), a slight decrease in the high apparent molecular weight fractions, a slight increase in the smaller fractions, a significant increase of the carboxylic functions, and the formation of ozonation byproducts (OBP). These byproducts include small molecular weight compounds like aldehydes (Le Lacheur *et al.* 1993), acetones, ketoacids

(Xie and Reckhow, 1992; Le Lacheur et al 1993) and carboxylic acids (Gagnon et al 1997). In general, ozonation makes NOM more biologically available for bacteria in biofilters.

Carlson and Amy (1998) reported the following composition of filter removable BDOC:

- aldehydes (formaldehyde, acetaldehyde, glyoxal methyl glyoxal): 3%
- ketoacids (glyoxylic acid, puryvic acid, ketomalonic acid): 12%
- carboxylic acids (formic acid, acetic acid, oxalic acid): 13-15%
- unknown: 70-72%

The formation of ozonation by-products (aldehydes and ketoacids) was proportional to the DOC content and the ozone dosage (Griffini et al., 1999). The formation of BDOC and OBPs during ozonation was similar during the ozonation stage.

These compounds have different degradation rates and therefore different conditions (temperature, media, backwash etc.) are likely to influence the removal of these compounds differently. Liu et al. (2001) studied degradation of formaldehyde, glyoxal, formate and acetate in bench-scale biofilters. Glyoxal, which is a slowly biodegradable aldehyde was removed to a lesser degree in biofilters than other compounds and it was more influenced by media, low temperature and the presence of chlorine during backwash.

Yavich et al. (2004) studied the effect of ozone doses on the BDOC degradation rate and found that the degradation rate increased with increasing ozone dose. Carlson and Amy (2001) showed that there is some maximal value above which the degradation rate of BDOC_{rapid} is not increasing significantly. More research is needed to be able to do any generalisation on this issue.

2.8 Other factors affecting the removal of NOM by biofiltration

Temperature is one of the important factors affecting the removal of NOM by biofiltration. In cold climates the application of biofiltration is possible; however DOC removal efficiency will be low when the water temperature is low.

3 PLANT OPERATION PRACTICES

3.1 Ozonation

3.1.1. Use of ozone in water treatment

Since the discovery of ozone in 1840, this oxidant has been much used for water purification, due to its strong bactericide activity. However, a widespread application of ozone was only seen in a few countries in Europe, namely France, Germany and Switzerland. During the last decades however ozonation is introduced in more and more water treatment plants as a replacement of primary chlorination in order to control the formation of chlorination by-products.

Because ozone is a strong oxidant, it can be used not only for disinfection but also for removal (oxidation) of organic substances. Ozone is used for the treatment of water polluted with pesticides or other anthropogenic substances. Ozonation can be combined with UV and H₂O₂ treatment in order to increase the oxidation power. Such advanced oxidation processes (AOP) appear to be potentially effective methods for the removal of emerging substances like endocrine disruptors, pharmaceuticals, resin softeners, new disinfections by-products, etc..

Ozone can also be added prior to coagulation to improve particle removal efficiency by inducing so-called microfloculation. This potential beneficial effect of preozonation is however dependent upon several factors, including water hardness and DOC concentration. In moderate to high DOC waters the coagulant dose is set by the DOC. Ozonation converts NOM into smaller compounds, e.g. oxalic acid, that adversely affects coagulation. Thus, microfloculation will not be observed for moderate to high DOC waters. In low DOC waters the coagulant dose is set by the particles and the adsorbed organic matter. Ozonation may react with adsorbed DOC and thereby alter the amount and conformation of adsorbed organic matter. Therefore microfloculation is most likely to occur in low DOC waters (Becker and O'Melia, 2001). Thus, preozonation is effective in low DOC waters whereas it can have adverse effects in high DOC waters.

The use of ozonation can combine disinfection and NOM removal. During ozonation NOM is transformed to BOM which can be removed in the biofilters. Depending on EBCT, temperature and type and forms of BOM, 10-50% of NOM can be removed. Thus the application of biofilters will restrict the bacterial regrowth in the water distribution network and decrease the disinfection byproduct formation.

3.1.2 Ozone chemistry

Ozone (O₃) is a highly reactive gas formed by electrical discharge in the presence of oxygen (O₂). Substantial amount of energy are required to split the stable oxygen - oxygen covalent bond to form ozone, and the ozone molecule readily reverts to elemental oxygen during the oxidation reduction reactions. Ozone is more soluble than oxygen in water. Once ozone enters solution, it follows two basic reaction paths: 1) direct oxidation, which is rather slow and selective, and 2) autodecomposition to the hydroxyl radical. A low pH favours the slow, direct oxidation reaction path involving O₃, and a high pH or a high concentration of organic matter favour the autodecomposition route. A high concentration of bicarbonate or carbonate buffer reduces the rate of autodecomposition by scavenging the hydroxyl radicals. This means that ozone residuals last longer at low pH and in highly buffered waters.

3.1.3. Ozone systems

Ozone systems are typically comprised of four basic parts: ozone generators, feed gas preparation, contacting, and off-gas disposal. In ozone generators ozone is produced from air or oxygen which is passed between two electrodes covered with a dielectric. Air or oxygen is prepared on site or supplied at the plant in liquid form. The generated ozone gas is fed into the water flow. To ensure necessary disinfection efficiency the certain predetermined contact time between ozone and water is provided in the contact chambers. Since the high concentration of ozone can be toxic the ozone off-gas escaping from upper surfaces of water is collected and destroyed (e.g. treated with ultraviolet radiation) to neutralize the ozone to produce oxygen.

3.1.3 Ozone effect on biofiltration

Ozone is used prior to biofilters for two main purposes: 1) disinfection, and 2) oxidation of organic matter to produce more BOM. In practice, the ozone doses and ozone contact times are selected to ensure the inactivation of protozoan oocyst like *Cryptosporidium parvum* and *Giardia lamblia* and viruses. In USA, for example, water treatment plants serving more than 10,000 people and use surface water as their source water are required to obtain at least 99% (2-log) removal of oocysts (USEPA, 1998). The ozone dose and contact time required to obtain a given removal is mainly depended of pH and temperature.

A biofilter may also provide some additional barrier efficiency against pathogens. However, accumulation of biofilm on the filter media and a high NOM content in the water significantly impaired the removal of oocysts relative to a clean filter bed operated in the absence of NOM (Dai and Hozalski, 2002).

3.2 Biofiltration

The observation that granular activated carbon (GAC) filters were able to remove DOC far beyond the point at which the adsorption capacity is exhausted has led to the development of biofiltration processes for drinking water treatment. Due to its low maintenance costs and effective BDOC removal, biofiltration is becoming an attractive unit process. In the following subsections the mechanisms involved in DOC removal in the biofilters and the factors affecting their performances will be reviewed.

3.2.1 Mechanisms of DOC removal in biofilters

In textbooks, biofiltration is often not identified as a specific water treatment process, it is rather discussed in connection with biological activity in granular activated carbon (GAC) filters (AWWA, 1999). The shift from adsorptive processes to biological processes is well illustrated in Figure 3.1, which shows that when the GAC adsorption capacity is exhausted the biological processes, mostly occurring in the biofilm on the surface of the filter grains, start to dominate the DOC removal efficiency. Although not very significant (Kaplan and Newbold 1995), the sorption process may continue on the surface of the biofilm covering the filter media (Carlson and Silverstein 1997). This type of sorption is called biosorption.

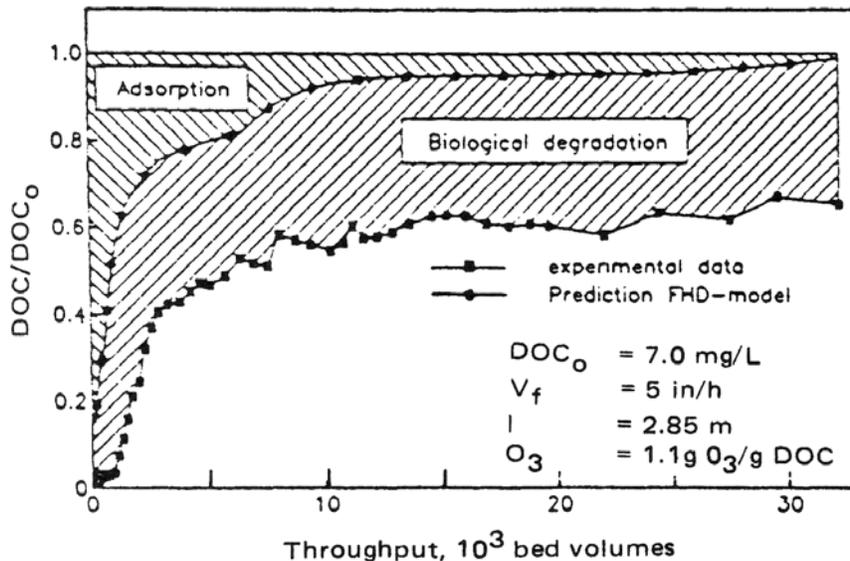


Fig. 3.1 DOC removal by adsorption and biodegradation activity during GAC filtration of an ozonated humic acid solution (Source: AWWA, 1999)

Most of the microorganisms present in biofilters, as well as in the entire water supply system, are living and growing in biofilms (Figure 3.2). A common feature of a biofilm is that the microorganisms are embedded in a matrix of extracellular polymers (EPS). Even if GAC is used as a biofilter media, the

bacteria (biofilm) can cover the outer surfaces because the pores of GAC are too small (1-100nm) for bacteria (>200 nm) to enter. Biofilms in biofilters contains bacteria, both aerobic and anaerobic, as well as protozoa which are grazing on the bacteria. The type and structure of bacteria is depended on water chemistry and composition of DOC. Bacteria which utilise readily available carbon is located in the top layer (Moll et al., 1998) while bacteria able to degrade more recalcitrant carbon are living deeper in the filter.

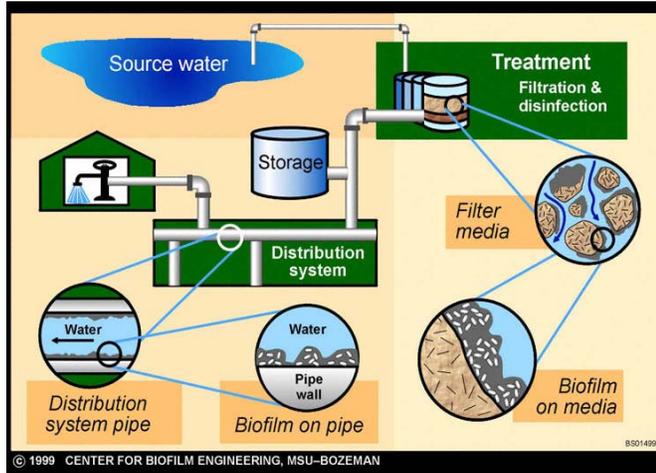


Fig. 3.2. Biofilms in drinking water supply systems(Source: www.erc.montana.edu)

The biomass levels in biofilters can be measured by analysing the lipid-bound phosphorus (total lipid phosphate) content (Findlay et al., 1989) which is then converted to carbon and bacteria number per cm^3 of media. About $1 - 1000 \mu\text{gC cm}^{-3}$ or about $10^8-10^{10} \text{ cells cm}^{-3}$ have been determined in operating biofilters (Servais et al., 1991; Magic-Knezev and van der Kooij, 2004). However, because of the fact that not all of the bacteria in biofilters are alive the total bacterial number does not always correlate well with the removal efficiency of DOC. For example, during winter periods with lowering of the temperature the number of bacteria may remain the same, but their activity is significantly decreased (Fonseca et al 2001). This is the reason why bacterial activity is measured instead of bacterial number.

The biological activity in biofilters is normally assessed by analysing the oxygen consumption, the heterotrophic plate counts, the 5-cyano-2, 3-ditolyl tetrazolium chloride (CTC), the 2-(*p*-iodo-phenyl)-3-(*p*-nitrophenyl)-*s*-phenyl tetrazolium chloride (INT) (Fonseca et al 2001), or the adenosine triphosphate (ATP) content in the biomass. Although the active biomass content is dependent on many factors and may vary over a wide range (25 to 5000 ng cm^{-3}), as shown in studies in the Netherlands (Figure 3.3), it is clear that the bacterial activity is increasing with increasing amount of readily available DOC (e.g after ozonation) and increasing temperature.

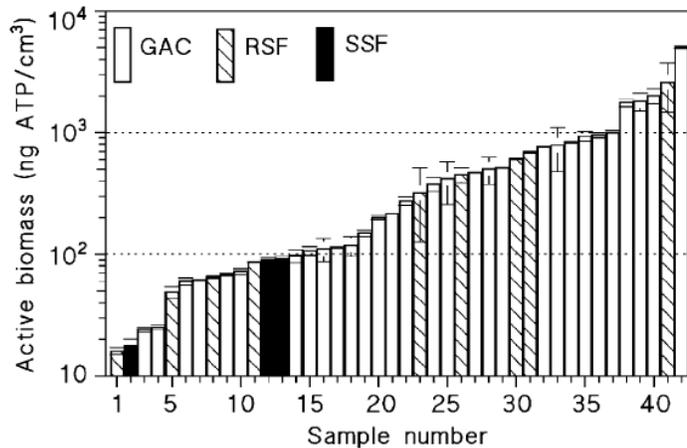


Figure 3.3. Active-biomass concentrations in GAC, RSF and SSF samples from 9 water treatment plants in the Netherlands. GAC, granular activated carbon; RFS, rapid sand filters; SSF, slow sand filters (Magic-Knezev and van der Kooij, 2004).

Biodegradation is the dominating process occurring in biofilters used for the removal of DOC from water. DOC is used by bacteria for energy production (catabolisms) and for the building of biomass material (anabolism) (Fig. 3.4). As a result, DOC is transformed into CO₂ or other organic substances. Some of these substances are readily degradable organic substances such as AOC and used by other bacteria, while others can be recalcitrant to bacteria or so called soluble microbial products (SMPs). The protozoa living in the filter consume bacteria and therefore also contribute to removal of TOC. During the operation of biofilters protozoa and metazoa are deposited on the filter media and may proliferate.

Servais et al (1994) found that the density of protozoa in activated carbon biofilters used for treatment of drinking water was about 450 individuals per mL bed volume. In the top sections of the filter beds they found about 100 individuals per mL bed volume. At 6 m bed depth the density had decreased by a factor of approximately 50. Protozoa considerably contributed to the flux of organic matter in the biofilters investigated. Net bacterial production in the filters was found to be about 20 times higher directly after startup of the filters compared to the periods of pseudosteady state. This was supported by the fact that during the initial phase bacteria in the filter effluent were much bigger in size than later. For aquatic ecosystems it is known that protozoa preferably feed on bigger bacteria. This supports the hypothesis that considerable grazing occurs during pseudo-steady state biofiltration. It is difficult to compare the biomass of protozoa and bacteria in biofilters. Calculations can only give a rough estimate. From the numbers cited above and the typical concentrations of sessile bacteria it can be calculated that bacterial biomass is expected to be in the same order or up to 100 times higher

than the biomass of protozoa. However, conversion by protozoa can be considerable.

For bacteria nutrients such as nitrogen and phosphorus are needed. Oxygen is a common electron acceptor; therefore it is needed for effective utilisation of DOC. Carbon can be degraded also in absence of phosphorus but then this process would be slower.

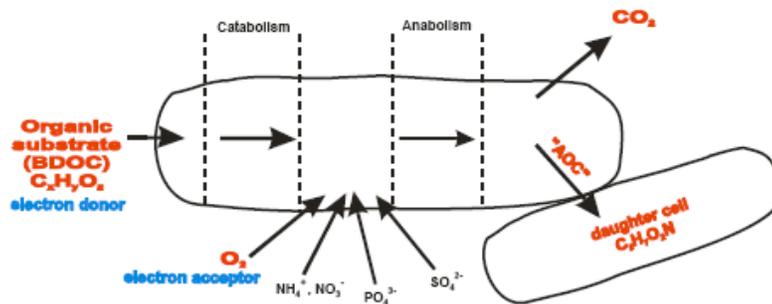


Fig. 3.4 Transformation of DOC by bacteria in process of biodegradation (Source: Uhl, 2006)

3.2.2 Biofilm establishment

When new biofilters are taken into use, they go through a colonisation phase to achieve full biodegradation activity. Biofilm growth in new biofilters is a slow process. Wang *et al.* (1995) studied the amount of phospholipids on the top of anthracite-sand filters receiving ozonated water. It took 4-5 months before a steady-state level of about 80 nmol lipid-P g⁻¹ media was measured. In GAC filters, a 50% increase in biomass was still observed between 100 and 200 days of operation.

However, achieving steady-state biological removal of organic matter does not take as long time as steady-state biofilm formation. The amount of biofilm and the removal of organic matter do not necessarily correlate (Wang *et al.*, 1995; Liu *et al.*, 2001). In the anthracite-sand filter described above, a steady-state removal of different organic parameters was achieved within 100 days although the biomass continued to increase after this time. The amount of biomass is not necessarily related to the activity of the biomass. However, the amount of biomass may be an important factor for buffering of the biofilter performance against changes in operational conditions like filter loading rate or water temperature. For example, Liu *et al.* (2001) observed less than expected reduction in the removal efficiency when temperature of the raw water was decreased. Their explanation was that accumulated, but less active biomass located deeper in the filter compensated the reduced biodegradation rates at the lower temperature. At high temperatures, the biodegradation of

easily biodegradable organic matter occurred at the top of the biofilters while at lower temperatures it happened along the whole filter bed.

Najm *et al* (2005) compared the performance of GAC-sand biofilters with fresh and used GAC. Even after seven months of operation, the amount of biomass on the surface of the fresh GAC filter was 1.0-1.5 log lower compared to filter media that had been used for about four years (the levels were around 10^6 cfu g⁻¹). After one year of operation, the amount of biomass was more similar. However, from the start of the sampling period already (i.e. 3 months after replacing the media), the removal of BOM (DOC, AOC and aldehydes) was similar in each filter.

Servais *et al* (1994) observed that pilot-scale GAC filters required 100 days before the fixed bacterial biomass levels were at the same level as in biofilters that contained GAC from a BAC filter that had been operative for several years. However they did not measure BDOC very often during the colonisation phase so it is not known whether maximum BDOC removal was achieved earlier.

Griffini *et al* (1999) studied four different GAC filters and a biolite filter at 20 min EBCT. In the GAC filters, the HPCs increased in the biofilter effluents during the first four weeks until they stabilized at levels of $0.7-1.5 \times 10^5$ cfu mL⁻¹. For the biolite filter, approx. 100 days were required before similar levels were observed in the filter effluent. Formaldehyde and acetaldehyde removal started after 21-28 days of operation and over 90% removal was observed after 42 days of operation for the GAC filters. In the biolite filter however, the removal started at day 84 and reached over 90% removal after day 110. A similar pattern was observed for glyoxal and methyl glyoxal, but 73 days were required to reach 80% removal in the GAC filters, and 122 days in the biolite filter. For ketoacids, 90% removal was observed after 64 days for the GAC filters, and 100 days for the biolite filter. In case of DOC removal, adsorption into GAC is the major removal mechanism during the first week of operation. For non-adsorbing biolite, DOC removal started to increase after 87 days of operation. Steady-state removal levels were observed after approximately 120 days of operation.

For anthracite-sand and GAC-sand biofilters fed by four OBPs (formaldehyde, glyoxal, formate, acetate), time periods of 20 to 40 days were required for the biofilters to reach (pseudo) steady-state removal at 20 °C (Liu *et al* 2001).

3.2.3 Filtration rates

Servais *et al.* (1994) operated three pilot-scale BAC filters at different filtration rates (6, 12 and 18 m h⁻¹) but at the same EBCT (10 min). They did not observe significant differences in BDOC removal in the different filters, indicating that filtration rate does not affect biological activity and therefore EBCT is the main design parameter. These filters had similar levels of fixed and effluent

biomass concentrations, indicating that the increased shear forces at higher filtration rates did not affect microbial colonisation and growth.

Carlson and Amy (1998) operated two anthracite biofilters at filtration rates of 5.0 and 9.7 m h⁻¹ with similar EBCT (10 and 11 min). The filters had the same BOM removal efficiency, but the filter with the higher filtration rate accumulated more biomass than expected from the differences in loading rates. The biomass concentration profile appeared to be a function of the hydraulic loading rate. At a higher loading rate the amount of biomass was higher in the top of the filter and penetrated deeper into the filter bed. It appears that with higher filtration rates, less carbon was needed for cell maintenance and the carbon utilization yield coefficient was higher. Therefore, for a given media depth there was an optimum filtration rate which gave optimum biomass accumulation. A short term reduction in filtration rate from 9.7 m h⁻¹ to 5.0 m h⁻¹ did not result in better removal efficiency and the DOC removal became BOM limited. A short term increase in filtration rate (from 9.7 to 17.5 m h⁻¹) decreased the DOC removal efficiency and the removal became biomass limited. These results show that for optimal performance, biofilters should be operated as close to steady-state conditions as possible.

Filtration rate may be important for particle removal and higher effluent turbidity and higher head loss have been reported with higher filtration rates (LeChevallier *et al.*, 1992).

Griffini *et al.* (1999) observed that in steady-state, the removal of aldehydes and ketoacids mostly occurred in the top sections of the biofilter after 2.3 min EBCT. Little additional removal occurred in the rest of the biofilter. In case of BDOC, samples taken at different biofilter depths (i.e. increasing EBCT) showed that the majority of the BDOC removal occurred in the top sections of the biofilter with some additional removal in the lower levels (Figure 3.5). A 10-15 min EBCT would have been enough in this case.

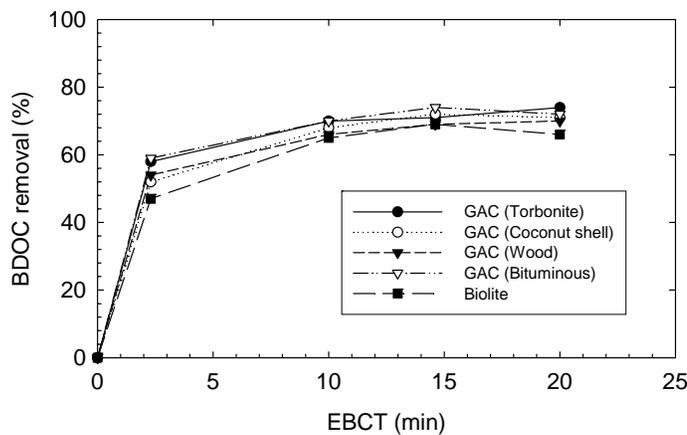


Figure 3.5. BDOC removal in biofilters with samples taken along the filter column, i.e. at different EBCTs (Griffini *et al.*, 1999).

3.2.4 Backwash practice

A successful biofiltration requires that the amount of biomass on the media is carefully controlled and maintained during the maturation period and during filter backwashing. Because biological filter beds accumulate both biological and non-biological particles, removal of these particles during backwashing is an important aspect of biological filter operation. For biological filters, an effective backwashing strategy involves removing excess biomass while not totally removing the attached biomass required for the start of the next run (Ahmad *et al*, 1998).

Important factors for the backwash of biofilters:

- bed expansion
- water flow rate
- air scour
- chlorinated water wash
- collapse pulsing air

Bacteria are hydrophobic and are therefore more strongly associated with surfaces than inorganic particles. In addition, bacteria are fixed to media surfaces by extracellular polymers in biofilms. Therefore bacteria are more difficult to remove from filter beds by backwash than inorganic materials. In order to control biofilm growth longer backwash times and/or stronger backwash strategies than for conventional depth filters may be required (Ahmad and Amirtharajah, 1998).

In North America, backwashing of filters is often performed by chlorinated water. Therefore, many American projects have studied the effect of chlorinated backwash on biofilter performance. The results show that controlling biofilm growth using chlorinated backwash water is a doubtful practice. Backwash with chlorinated water impairs the performance of a biofilter at least during the early filter cycle (Miltner *et al* 1995; Wang *et al* 1995; Ahmad *et al* 1998). However, chlorinated backwash does not completely remove biological activity and the biological activity recovers towards the end of the filter cycle. Of course, the effect of chlorinated backwash on biological activity depends on chlorine concentration and in addition the negative effect of chlorine is higher at low operating temperatures (Liu *et al.*, 2001).

Ahmad *et al* (1998) carried out pilot studies using anthracite-sand filters as biofilters. The biofilters received coagulated-sedimented river water which was ozonated with 1 mgO₃ mgDOC⁻¹. A comparison was made between collapse-pulsing air scour followed by water wash at 25% bed expansion and filters backwashed with water only (15, 25 and 50% bed expansion). The air scour resulted in higher turbidity in the ripening stage and higher initial head loss during filter run. After ripening, the differences in effluent turbidity were low and the filter run lengths were similar with 0-3 hrs differences. Run lengths with filtration rate of 10 m h⁻¹ were 20-24 hrs.

Heterotrophic plate counts were higher in the early stages of the filter run in the filter backwashed with water only. The authors take this as an indication of higher biological activity and that less biofilm was lost during the backwash with water only. Towards the end of the filter runs the HPCs were similar. However, AOC and NPOC removals were not statistically different, i.e. the backwash strategy did not significantly affect the removal of biodegradable material (Ahmad et al 1998). In bench scale bioreactors, air scour did not generally affect BOM removal (Liu et al 2001).

Emelko et al (2006) carried out full-scale studies using GAC-sand and anthracite sand filters during cold water (1-3°C) and warm water (21-25°C) periods at the Mannheim water treatment plant (Kitchener, Ontario). The results were compared to normal operation of the plant, which was air scour followed by chlorinated low-rate water wash and then a chlorinated high-rate water wash. Presence or absence of air scour did not affect TOC removal. During the warm water periods oxalate removal was also similar with both backwash strategies. During cold periods, higher removal of oxalate was observed when air scour was used. Filter effluent turbidities, initial head loss and head loss rates were similar with both backwash strategies. The duration of the filter-to-waste period (i.e. during filter ripening with turbidity levels above 0.2 NTU) as well as the peak turbidity during ripening increased in the GAC-sand filter during the warm water season when air scour was not used. These effects were not observed during the cold periods or in the anthracite-sand filter. Collapse-pulsing was tested only with GAC-sand filter and no differences in TOC or oxalate removal at 9-12 °C was observed. Filter ripening time, maximum turbidity during ripening and turbidity removal were unaffected by collapse pulsing. However, collapse-pulsing resulted in significantly shorter filter run times due to a breakthrough of turbidity (replacing time based backwash after 100 hrs).

Miltner et al (1995) observed no effect of backwash conditions on BOM removal in anthracite-sand biofilters backwashed with water only at 50% expansion. No clear trends in the amount of biomass present in the top sections of the filter could either be observed. Filters were backwashed when head loss exceeded 150 cm.

At the Sweeney water treatment plant (Wilmington, USA) BAC-sand filters treating flocculated-sedimented and ozonated river water are backwashed after a maximum run time of about 100 hrs provided no turbidity breakthrough or excessive headloss is detected (Najm et al 2005). However, when a low turbidity surface water is treated by ozonation and biofiltration, filter run times of several weeks can be expected (Melin and Ødegaard, 1999). Similarly, a pressurized pilot-scale GAC filter treating ozonated low turbidity groundwater (0.2 NTU) was backwashed once in four weeks (Tan and Johnson, 2001). Figure 3.6 shows the sum of the aldehydes and ketoacids concentration along an up-flow expanded clay aggregate (Filtralite) filter sampled and analysed one day, and three weeks after backwash. The results show that there was a loss of biological activity as a result of filter backwash

in the inlet of the biofilter, but that the total performance of the biofilter was not affected at 22 min EBCT. However, effluent sampling of ozonation byproducts showed initial lower removal efficiency of some ketoacids after backwashing when the filters were operated at EBCTs of 11-15 min (Melin and Ødegaard, 1999).

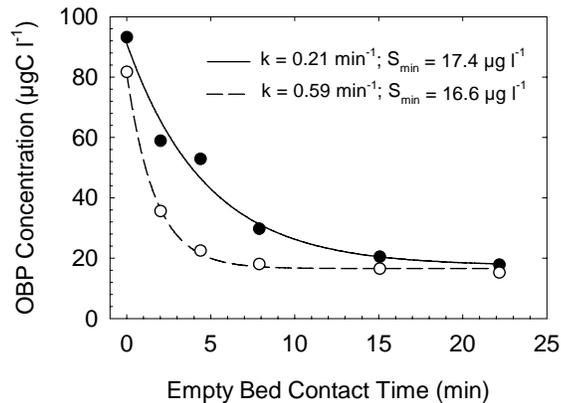


Figure 3.6. Ozonation by-product profiles along a biofilter column one (●) and 21 (○) days after backwash (Melin and Ødegaard, 1999).

At the Choisy-le-Roi treatment plant (Paris), BAC filters were washed every 50-100 hrs in summer and after several days in winter. The backwash is 2 min air scour at 30 m h⁻¹, air scour and water backwashing (10 m h⁻¹) until the water level reaches the washing throughs, and finally water backwash alone at 25 m h⁻¹ for 20 min. Servais et al (1991) took fixed biomass samples before and after backwash. Although the backwash reduced the stratification of biomass, it did not completely remove it. Analysis of the bacteria in backwash water indicated that only 4-8% of the fixed biomass was lost during backwashing. The effluent bacterial counts were also similar before and after backwash. According to simulations carried out by Hozalski and Bouwer (2001b), such a low loss of biomass will not affect the removal of BOM in biofilters.

From the results from Choisy-le-Roi, a carbon mass balance was calculated for the BAC filter (Table 3.1). The results indicate a bacterial yield of 0.29 gC gC⁻¹. It should also be noted that less than 1% of the produced biomass is lost during backwash and that 8% is lost in the filter effluent. Therefore, to maintain a steady-state biomass the protozoan grazing must play an important role.

Table 3.1. Carbon mass balance for full-scale BAC (media depth 1 m, filtration rate 5 m h⁻¹, backwash interval 50 hrs) (Servais et al., 1991).

Process	Carbon mass rate (gC m ⁻³ h ⁻¹)
BDOC uptake	1.2
NBDOC adsorption	0.6
Bacterial production	0.35
Bacterial respiration	0.85
Bacterial biomass exported during backwash	0.005
Bacterial biomass in filter effluent	0.028

Although many studies report either no loss of biodegradation or a slight temporary loss after backwash, some contrary results have also been published. In BAC-sand filters receiving ozonated river water pretreated by coagulation and sedimentation, Niquette et al (1998) observed that removal of BOM actually improved in the BAC layer after backwashing. They used air scour in their pilot plant with backwash intervals of 36-48 hrs. The authors suspected that this could be related to particle accumulation which may act as a diffusion barrier for the biodegradable compounds. Carlson et al (1996) observed that floc loading affected the interstitial volume significantly on the top of the anthracite filter reducing the actual contact time in the filter and the DOC removal in the top sections of the filter. However, the filter effluent DOC was unaffected at 11 min EBCT. The filters were backwashed with unchlorinated water.

Too long backwash intervals or ineffective backwashing may allow a large population of bacteria or particles to remain on the media and can lead to media clumping and mudball formation after long periods of operation. With pilot-scale Filtralite upflow filters, and filter run times of three to four weeks, media clumping in the inlet was observed which made backwash difficult even with air scour (Melin and Ødegaard, 1999). Therefore, long filter runs may not be advisable even if the head loss appears to be low in the biofilter.

Backwash strategy may also influence attrition of the filter material and also the filter material affects the amount of attrition. In collapse pulsing backwashing, stronger filter materials like sand and anthracite release fewer particles than activated carbon. In addition, after a certain time, there is no release of particles from sand and anthracite (Humby and Fitzpatrick, 1996). During a 100 hrs test equivalent to approximately 3 years of daily backwashing, two different activated carbons lost 2.1-3.5% and 4.5-7.3% of the media during backwash depending on method of calculation. Although losses were highest in the beginning of test, the GACs lost particles during the entire duration of the test while the anthracite stopped losing particles after about 70 hrs. Mikola et al (1999) carried out full-scale studies with backwashing of two-stage GAC filters that had been in operation for five months. The analysis of filter effluent after backwashing showed presence of carbon fines even one hour from backwashing when air scour was used. When backwash was carried out with only water, the release of carbon fines was minimal,

GAC effluent may contain carbon fines which have shown to contain both heterotrophic plate count bacteria and coliform bacteria (Camper et al 1986). Bacteria attached to GAC are more resistant to disinfection and may survive final chlorination. The fines were present throughout the filter cycle and therefore not strongly connected to backwash. However, it was not clearly stated what was the function of the GAC filters in the treatment plants sampled, but it seemed that they were used as adsorption filters rather than biofilters. Although pathogenic bacteria can attach and survive in pure GAC beds, the attachment is inhibited and their levels declined faster in colonised GAC filters (Camber et al 1985). This is probably due to the fact that they cannot compete with indigenous bacteria for nutrients. Therefore it is not likely that pathogenic bacteria accumulate in biofilters. Tan and Johnson (2001) detected no coliforms in an ozonated/biofiltered water, and LeChevallier et al (1992) reported that "coliform bacteria were rarely observed in biologically treated effluents".

3.2.5 *Effect of hydraulic transients*

An increase of anthracite-sand filter velocity from 10 to 13 m h⁻¹ 1 hr after the maximum turbidity ripening peak resulted in increased effluent turbidity and breakthrough within 3 hrs of additional run time (Ahmad *et al.*, 1998). In contrast, it took around 6 hrs before breakthrough happened in a non-biological (pre-chlorinated) filter with normal run times about 24 hrs. The hydraulic transient did not affect effluent HPC level or AOC removal. Larger changes (from 9.7 to 17.5 m h⁻¹ can cause significant reduction in BOM removal efficiency (Carlson and Amy, 1998).

3.2.6 *Biofilter media*

The type of biofilter media affects the amount of biomass accumulating in the biofilter (Table 3.2). For comparison, Table 3.2 also shows anthracite filters which received chlorinated water or was backwashed with chlorinated water. Although normalising biofilm amount for media weight is not the best alternative since it does not take into account different densities or diameters of media grains, the results show the highest biofilm accumulation in the GAC filters. However, there are differences between different GAC which are probably due to the pore size distribution. Niquette et al (1998) estimated the weighted mean of the biomass present in BAC-sand filters to about 14 µgC cm⁻³ while in anthracite-sand filter it was 3.0 µgC cm⁻³. Anthracite-sand removed also less OBPs at 12 min EBCT.

Table 3.2. Biomass accumulation on biofilter media after 95 days (Wang et al., 1995).

Media	Biomass nmol lipid-P g ⁻¹ media	
	Mean	Standard deviation
Anthracite prechlorinated	2.0	0.5
Anthracite backwashed with chlorinated water	6.0	0.6
Anthracite	55.0	1.7
Sand	90.6	1.3
GAC1 (bituminous coal)	305	9
GAC2 (lignite coal)	465	9
GAC3 (wood)	382	11

The amount of biofilm present in biofilters does not directly compare with biofilter performance. The steady-state removal of AOC-NOX was not statistically different in the anthracite, sand and GAC filters in Table 3.2 although on average GAC1 and GAC2 had the highest AOC removal (Wang et al 1995). Similarly, Liu et al (2001) found no correlation between BOM removal and biomass. Biofilm accumulation is highest in the top sections of the biofilter (in downflow filters) and the amount of biofilm decreases with the filter depth (Wang et al 1995), which correlates with high initial removal of BOM in the top layers of the biofilter.

Table 3.3 shows a comparison of four GACs manufactured from different materials and from a non-adsorbing media (biolite) during parallel testing (Griffini et al. 1999). The removal efficiencies are from periods when the biofilters appeared to have reached steady-state removal. The influent NBD OC and BDOC concentrations were 1.37-1.62 mg L⁻¹ and 0.32-0.59 mg L⁻¹, respectively. Some GACs maintained high adsorption during the test period as can be seen from the non-biodegradable DOC removal (NBD OC). The GAC filters achieved slightly better BDOC removal compared to the biolite filter.

Table 3.3. Average removal efficiencies of filter materials in pilot-scale studies between 122 and 290 days of operation at 20 min EBCT (Griffini et al., 1999).

Filter material	DOC removal (%)	BDOC removal (%)	NBD OC removal (%)
GAC (Torbonite)	59	74	50
GAC (Coconut shell)	40	71	24
GAC (Wood)	30	70	13
GAC (Bituminous)	53	72	40
Biolite	18	66	5

GAC filters are more likely to tolerate adverse conditions (low temperature, chlorine in backwash water) than sand-anthracite filters (Liu et al, 2001). A full-scale study showed that although there was little difference during warm water period between GAC-sand and anthracite sand filters, GAC-sand filters had better removal efficiencies during cold water period (Emelko et al 2006).

Najm et al (2005) made full-scale tests between lignite and bituminous GAC between 3 and 13 months of operation at EBCTs varying 10-60 min. There was no statistical difference between the biofilters considering DOC, AOC, aldehyde or turbidity removal. The biomass accumulation was also similar. The authors concluded that lignite would be advantageous since it is lighter and therefore requires lower pumping rates during backwash. If the influent contains residual dissolved ozone, GAC or anthracite is an advantage because ozone is rapidly decomposed in the media. With ozone residual of 0.4 mg L⁻¹, no ozone was detected at first sampling point at about 2 min EBCT in biofilters (Niquette et al 1998).

Melin et al (2006) compared a three-media (anthracite-sand-CaCO₃) filter with a filter with CaCO₃ as the only filter material. When the filters received water with residual dissolved ozone, the ozone was removed in the first few centimetres in the anthracite layer while ozone penetrated almost 60 cm of the CaCO₃ filter. This is not a problem in treatment plants with open filters because they are operated with no dissolved ozone in the biofilter influent. However, in plants having closed filters the ozone contactor may be operated with high residual ozone.

Another advantage of GAC is that during start-up some of the organic matter (BDOC) is adsorbed while the biological activity is building up (Carlson et al 1994; Servais et al 1994; Melin et al 2002). The length of the adsorption phase can vary quite a lot depending on the type of GAC (Carlson et al 1994).

4 OPERATIONAL CHALLENGES AND BOTTLENECKS

4.1 Low temperatures

Microbial growth and degradation rates depend on water temperature. In northern climates, low water temperatures during winter may represent a potential problem for BOM removal, especially in biofilters having a short EBCT.

During long term pilot-scale treatment of surface water with temperatures ranging from 5 to 30 °C, Nishijima *et al*, (1998) did not observe significant reduction of biodegradable organic matter at the lowest temperatures in an ozonation-BAC process operated with 15 min EBCT. Melin *et al* (2002) observed seasonal changes in TOC removal efficiency in a treatment plant treating humic surface water with water temperatures ranging from 2 to 14°C. The TOC removal efficiency did not, however, completely correlate with the water temperature and changes in the TOC removal might have been due to seasonal changes in NOM composition. BDOC analyses did not reveal any clear trend raw water temperatures of 3 to 10°C and 54% removal was observed at 3°C. The biofilter had EBCT of 26-30 min which might have helped to mitigate the temperature effect.

Emelko *et al* (2006) did not observe statistically significant differences in TOC removals in full-scale GAC-sand or anthracite-sand filters at cold (1-3 °C) or warm (21-24 °C) water periods. During cold water periods, GAC filters had better TOC removal (23%) compared with anthracite (14%). The filter loadings were slightly lower during the cold water periods (5.8 m h⁻¹) compared to the warm water periods (4.2 m h⁻¹). When they analysed a major OBP, oxalate, its removal efficiency was reduced from 92% to 56-72% in the GAC-sand filters during the cold water period, and it was not removed at all in the anthracite-sand filters. GAC might therefore be a better filter media if the treatment plant receives cold water.

In biofilters fed by a combination of formaldehyde, glyoxal, formate and acetate the removal rates were similar at 5 and 20°C, especially in BAC filters (Liu *et al* 2001). The temperature effect on degradation rate is “mitigated” to a certain degree in a biofilter, i.e. while at high temperatures degradation occurs mainly in the top layers of the filter, BOM removal occurs throughout the entire filter at lower temperatures. Niquette *et al* (1998) observed that at 10-12 °C some OPBs were degraded within 6 min EBCT in BAC-sand filters while at 0.5 °C they were degraded evenly along the filter depth with 12 min EBCT.

Servais *et al* (1991; 1994) reported that at low temperatures (< 15 °C) there was less stratification of fixed biomass in the BAC filter while stratification

was clear at higher temperatures. At high temperatures, the degradation activity is probably so high that less biomass is able to grow at lower levels of the biofilter.

4.2 Limiting nutrients

Bacteria require nutrients and vitamins for growth. A recommended ratio between the major nutrients C:N:P is 100:10:1 (in mg L⁻¹). In some cases, especially if the raw water contains a high amount of organic carbon, the inorganic nutrients can become limiting. Limitation of phosphorus for microbial growth in drinking water has been observed in Japan, China, Finland, and Latvia (Sathasivan et al 1997; Lehtola et al 2001). Because phosphorus is effectively removed during chemical coagulation this nutrient can become limiting (Lehtola *et al.*, 2002; Rubulis and Juhna, 2002). In such cases removal of organic carbon in biofilters may not be effective. Trials with dosage of phosphorus prior to biofiltration have been reported. However, there was no increase in TOC or AOC removal, but instead a significant increase in heterotrophic bacteria in the biofilter effluent was observed (Vahala et al 1998). It was not analysed, though, whether water treated in biofilter actually was nutrient limited.

However, it seems possible to get significant biological activity in biofilters even with low nutrient waters. Melin et al (2002) operated an ozonation-biofiltration plant treating humic surface water which had total phosphorus concentrations of 1.1 to 2.0 µg L⁻¹ and the batch tests showed that growth and DOC degradation was phosphorus limited. Despite this, the obtained removal efficiencies of TOC (average 23%) and BDOC (average 55%) were not especially low. The effect of low phosphorus concentration is however not well understood.

4.3 Oxygen supersaturation

If oxygen is used for ozone production, the treated water may become supersaturated by dissolved oxygen. Although there is evidence that extremely high dissolved oxygen concentrations (≥ 42 mg L⁻¹) is inhibitory for microorganisms (Mikell et al 1984) this seems not to be a problem for microbial activity in practice.

Tan and Johnson (2001) reported that an average DO concentration of 19 mg L⁻¹ was measured in the effluent of a pressurized BAC filter treating ground water, and ozonated with a system based on pure oxygen without apparent problems in the biological activity. The biofilter had 58% AOC removal and high levels of HPC in the effluent.

4.4 Invertebrates

The food chain is developing in biofilters, thus if not properly controlled invertebrates such as nematodes, gastrotrichs, rotifers, oligochaetes and anellids may proliferate in the outlet from biofilters. The source of these

organisms is often unknown and difficult to identify. Invertebrates have been found in biofilters even in cases where groundwater which did not contain these organisms was used.

Presence of these “worm like” organisms in drinking water can cause complaints from consumers. From a hygienic point of view they are not considered dangerous, because even if bacteria and viruses can adhere or be ingested by the invertebrates, there is no evidence of pathologies associated with their recovery (Gray 1994). In practice, the maximal concentration should not exceed about 3–4 nematodes/L in drinkable water and 20 nematodes/L in raw water. If this limit is exceeded, it is useful to implement engineering containment measures.

The chlorination of filters during backwash is not an effective approach for predators control because residual chlorine is rapidly decreased in the filter bed due to reactions with the filter material, the organic matter (biofilm) covering it, and the organic deposits. To control invertebrates (oligochaetes) a backwashing routine which involves reduction of the amount of oxygen during filter shutdown was proposed (Beaudet et al, submitted). The biomass present in the filters consumes the dissolved oxygen, resulting in the development of anoxic and anaerobic conditions and consequently the death of oligochaetes. Then they are removed by a proper filter backwashing technique following the shutdown. The problems associated with this approach are bacterial by-products formation (e.g toxins) and temporary lower water quality (e.g. ammonium) after start up of the filter after the standby.

4.5 High concentration of microorganisms in the effluent of biofilters

Due to bacterial growth in the biofilter the concentration of microorganisms in biofilter effluents can be high, with levels reaching 10^5 - 10^6 cell mL^{-1} . To reduce the bacterial levels some plants have introduced ultrafiltration processes after biofiltration. This is an effective, however expensive process.

Another approach is to change the operation conditions of the filters by increasing the bed depth and the EBCT accordingly. This can bring about a sharp reduction of the numbers of organisms in the filtrate. On the other hand, increasing the EBCT by decreasing the flow rate did not decrease but sometimes increase the plate count (van der Kooij 1983). The problem remains to be solved.

5 Pre- and posttreatments

5.1 No pre-treatment

Many small ozonation-biofiltration plants are operated without any pretreatment, and ozonation is used for color removal and disinfection. This is especially a possible case when the raw water is low in turbidity.

5.2 Pre-treatment

5.2.1 *Artificial groundwater recharge*

The bacterial regrowth potential is usually low if artificial groundwater is used as the raw water source. This process is very effective in removing AOC and BDOC, due to long retention times and high bacterial activity in the aquifer. However, for further removal of NOM with biofiltration, an oxidation/ozonation step is always needed.

5.2.2 *Coagulation, flocculation and sedimentation*

To decrease the particle load to biofilters, coagulation, flocculation and sedimentation can be employed as pretreatment (see references in Uhl, 2000). During these processes not only fine suspended particles are removed, but also DOC. Part of the removed organic matter is biodegradable, resulting in a considerable reduction of AOC or BRP as well.

A study by Volk and LeChevallier (2002) showed conflicting results. A 12-month assimilable organic carbon (AOC) survey of 64 conventional water treatment plants was conducted to assess mechanisms affecting effluent AOC levels. The effect of conventional treatment steps (i.e., oxidation, coagulation, filtration, disinfection) on the formation or removal of biodegradable organic matter (BOM) was also investigated at six of the sites. BOM levels were increased or decreased by various factors including the type of filter media, the type of coagulant used, and the level of watershed protection of the plant's source water.

Contrary to conventional filtration processes, however, the enhanced coagulation process aimed at maximizing the removal of DOC employs elevated coagulant concentrations and strict pH control. In the U.S. enhanced coagulation is considered as one of the best available technologies for TOC removal. Enhanced coagulation removes primarily the high molecular weight and hydrophobic (humic) fractions of NOM. The biofiltration processes on the other hand removes primarily the more hydrophilic and low molecular weight fractions of NOM. Thus a biofiltration process with enhanced coagulation as pre-treatment is an interesting process combination.

Several types of coagulants and flocculants are available. It has been reported that polyaluminum chloride may have an inhibitory effect on the growth of *P. fluorescens* P17 in the AOC assay. Thus, special care has to be taken when investigating AOC in samples from coagulation/ flocculation processes.

5.2.3 Oxidation before biofiltration

Ozonation is an oxidation process. The reaction of ozone with natural organic matter can take place via the molecular ozone pathway and the free radical pathway. Both pathways result in the formation of higher oxidized organic compounds with lower molecular weight and better biodegradability. With increase of ozone dose BRP and AOC is increasing. At low specific ozone doses (i.e. up to 1.5 mg O₃ mg⁻¹ DOC) a linear relationship between ozone dose and AOC formation is observed. For higher ozone doses AOC formation levels off and reaches a plateau concentration (Figure 5.1). This may be due to the effect that at further addition of ozone a part of the low-molecular weight compounds are oxidized to carbon dioxide and, in parallel, new AOC is formed from remaining higher molecular weight compounds. When applying ozonation in drinking water treatment, special care has to be taken to secure that the following treatment steps are well designed for the removal of easily biodegradable compounds.

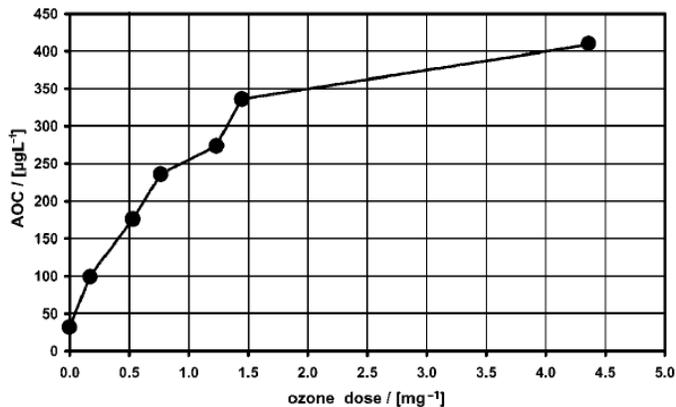


Figure 5.1. Effect of the ozone dose on AOC formation (Uhl, 2000)

The application of specific ozone dose levels of 0.5 to 1.0 g O₃/g DOC is quite common prior to biofiltration. However, if the water contains high amounts of refractory DOC, more advanced oxidation processes (AOP) could be used. AOP implies the combination of UV-light or H₂O₂ with ozone to promote the formation of highly reactive OH-radicals, which would enhance the oxidation of refractory DOC. Another reason for the use of AOP is to reduce the formation of bromate during ozonation. However, compared to ozonation, AOP processes are relatively costly.

5.2.4 *Multistage ozonation biofiltration*

Nishijima et al (2003) studied a multi-stage ozonation-biological treatment process for DOC removal from drinking water. Waters with different types of DOC were used, i.e. a reservoir water for drinking water supply, a secondary effluent from a municipal wastewater treatment plant and a solution of humic substances extracted from leaf mold. The multi-stage ozonation-biological treatment process was compared with a conventional single-stage ozonation-biological treatment process. The amount of DOC removed during biological treatment was defined as the amount of BDOC in the influent to the biological treatment step. DOC removal in the multi-stage ozonation-biological treatment was higher than that in the conventional single-stage ozonation-biological treatment with the same total ozonation time for the reservoir water and the humic substance solution. Moreover, three- or four-stage ozonation for 5 min followed by biological treatment (total ozonation time 15 or 20 min) showed higher removal of DOC than the single-stage ozonation (60 min) and biological treatment. The higher DOC removal in the multi-stage treatment was due to the production of BDOC by ozonation. The long-term ozonation was not effective to produce BDOC because most of the ozone was utilized to oxidize BDOC produced in the early stage of ozonation. In the multi-stage treatment, ozonation was effective to decompose refractory DOC and to produce BDOC because BDOC was removed by the biological treatment. However, the multi-stage ozonation-biological treatment was not effective for the secondary effluent. The reason seems to be high concentration of ozone scavengers in that water and low reactivity of DOC for ozone.

Fahmi et al (2003) compared ozonation-biological treatment with single and multi-stage advanced oxidation process (AOP)-biological treatment for drinking water treatment, especially for waters containing DOC considered less susceptible to oxidation by ozone. Minaga reservoir water and the secondary effluent from a municipal wastewater treatment plant were used as dissolved organic matter (DOM) solutions. DOC removals after 60 min AOP-biological treatment were 62% and 41% in the Minaga reservoir water and the secondary effluent, respectively, whereas those in the ozonation-biological treatment were only 40% and 15%, respectively. The results indicate that the single-stage AOP-biological treatment could improve DOC removal in comparison with the single-stage ozonation-biological treatment. This is because the AOP mineralized both biodegradable dissolved organic carbon (BDOC) produced in the early stage of oxidation and non-biodegradable dissolved organic carbon (NBDOC), whereas only BDOC was mineralized by further ozonation and NBDOC was not oxidized in the ozonation-biological treatment. The multi-stage treatment could not improve DOC removal in comparison with the single-stage treatment in the ozonation-biological treatment for the secondary effluent containing DOC less susceptible to oxidation by ozone. However, the multi-stage AOP-biological treatment significantly reduced DOC and achieved 71% of DOC removal by 4 times repetition of 15 min oxidation, whereas DOC removal was 41% in the single-stage AOP-biological treatment for the same oxidation time. The

improvement of DOC removal by the multi-stage AOP-biological treatment was due to BDOC removal both during biological treatment and as a result of the direct mineralization.

5.2.5 Chlorination

Chlorination, normally applied for disinfection purposes, is an oxidation process like ozonation, and results in an increase in biodegradability of organic matter (see references by Uhl, 2000). It was shown that an increasing chlorine dose (molar ratios of Cl : C of 0.1 and 1.0 were investigated) also increased the portion of DOC oxidized by bacteria from approximately 2% in the unchlorinated solution to about 6% at a Cl : C-ratio of 1.0. Moreover, it was shown that chlorination not only produced low-molecular weight compounds which could be degraded by bacteria. The low molecular weight compounds were removed after chlorination using gel chromatography, and the regrowth experiment was redone. This revealed that even high molecular weight compounds were more susceptible to utilization by bacteria after chlorination. LeChevalier et al (1992) observed that ozonation-biofiltration reduced the increase in AOC from chlorination.

5.3 Post-treatment

5.3.1 Biofiltration prior to other methods of water treatment

Biofouling control is considered as a major challenge in operating membrane systems. Hu et al (2005) studied the feasibility of using biofiltration as a pretreatment process to control RO membrane biofouling. Biofiltration was found to be a viable way of assimilable organic carbon (AOC) and dissolved organic carbon (DOC) removal, with removal efficiencies of 40-49% and 35-45% at an empty bed contact time (EBCT) of 30 min. It was also found that biofiltration as a pretreatment step reduced the rate of biofouling. It took only about 72 h for biofouling to have a significant impact on the performance of the RO membrane, when the system was operated without biofiltration as pretreatment. A five times increase in operational time to more than 300 h was observed when biofiltration pretreatment was used. This study illustrated well the suitability of biofiltration as an economical and simple way of biofouling control in RO membranes.

Biofiltration partially removes DOC from water therefore it decreases by-product formation potential and is used before the disinfection.

6 PROCESS CONTROL AND AUTOMATION

Raw water quality and flow in a water treatment plant is changing with time. To produce a constant water quality the water treatment processes should be controlled by adjusting to these changes. This process can be done manually and automatically. The automatic process control requires instrumentation including water quality sensors which are able to measure water quality continuously. The degree of instrumentation and automation in water treatment plants today varies significantly. Many plants are fully automated while others employ minimal monitoring and control technologies and rely on manual operation.

The application of modern process control and automation systems represents several advantages including: improvements in treated water quality and operation efficiency, increased safety and robustness, significant savings in energy and labour costs, and reduced maintenance cost.

6.1 Principles of automation and control

6.1.1 Automated control system components

The principles of advanced automated control system functions are shown in Figure 6.1. Depending on the objective of the automation (e.g. flow control, quality control) different sensors (e.g. flow meter, residual ozone sensor) continuously monitor changes in treatment process conditions and performance. The data acquired by the sensors are fed into the control system, which estimates an error between values measured by the sensor and the optimal values (i.e. set points) which are set by the operator or a mathematical algorithm. Then the control system turns on the actuators (e.g. variable speed drive pumps, valves, ozone dosing system) which adjust the operating conditions of the plant towards the optimal operation conditions (set points).

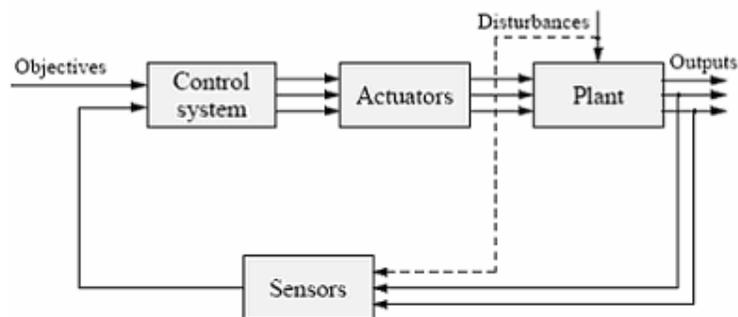


Figure 6.1. Principles and elements of an automated control system

There are two control levels: local and central. The local controllers can be mounted on the instrument or located in a special panel nearby the instrument. The local controllers in the panels are microcomputers including programmable logic controllers (PLC) with input/output (I/O) subsystems for monitoring and controlling processes and equipment. Remote terminal units (RTU) are other type of local controllers but they are located in remote sites and linked to a central control system by phone, radio or internet (Figure 6.2). Local controllers (PLC and RTU) are linked in the unified central control system (SCADA or DCS) which is operated by the software. Distributed control systems (DCS) and supervisory control and data acquisition (SCADA) systems are the two most widely used central control systems in drinking water treatment plants.

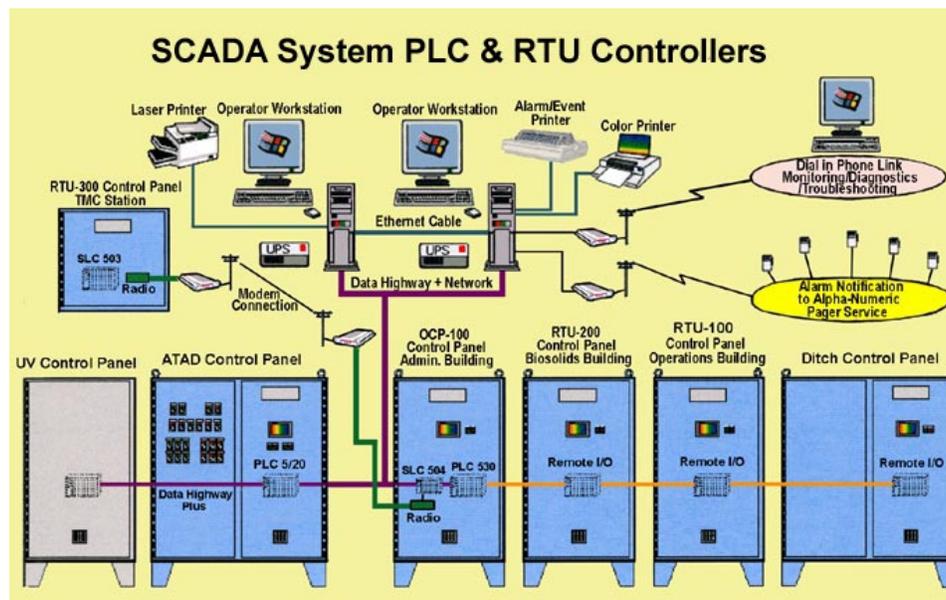


Figure 6.2. Automated control system (<http://www.princeton-indiana.com>)

6.1.2 Control types and algorithms

There are three types of automatic controls that are commonly implemented in the water treatment plants:

- sequential controls
- feedforward controls
- feedback controls

Sequential control is using a timer to switch devices on or off. Thus, only two state devices such as fixed speed motors and open-closed valves can be used with this type of control.

Feedforward control measure one or more process input parameters and calculates the necessary final control element settings based on these values. Ratio control is the most commonly used water treatment application of

feedforward control. This type of control is usually used for feeding chemical to water. This is done by measuring the water flow, calculating the amount of chemical that is needed to obtain the desirable ratio (dose), and feeding them into the water flow. This control is used if the water quality is not changing significantly with time.

Feedback control (or closed loop control) measures the value of controlled process variable and compares this with desirable setpoints. The feedback controller then adjusts the variables to minimize the error and maintain the process at the setpoint conditions.

A commonly used control algorithms for adjustment based on calculation of the error are proportional control, proportional plus integral (PI) control or proportional plus integral plus derivative (PID) control.

A proportional control simply adjusts the controlled device in a manner proportional to the measured process variable. However, this control does not always provide stability of water treatment process if there are rapid variations of the parameters with time. In this case PI controls are preferably used. PI control algorithms integrate changes in parameter values over time allowing for a more stable control. If there is a significant time lag of measured parameter values in response to adjustment (e.g. pH, temperature) PID control is used which basically adds a derivative control term added to PI control.

More advanced process control algorithms include adaptive gain control, nonlinear control, model based control and fuzzy logic control. To increase the accuracy of on-line measurements sensors can be placed in multiple points.

6.2 State of automation and control of biofilters

6.2.1 Biofiltration plant control strategies

Biofiltration consist of two principle water treatment processes: ozonation and filtration. As a rule of thumb, automation and control is done separately for each process. The most widely used monitoring parameters (by sensors) and controlled parameters (by actuators) are presented in Table 6.1.

Table 6.1 Monitored and controlled parameters in ozonation-biofiltration plants

	Filtration	Ozonation
Monitored parameters	Level, differential pressure, valve positions, flow, turbidity, particle counts	Ozone feed, flow, ozone residual, turbidity
Controlled parameters	Flow, level, backwash valves, backwash pumps, filter backwash pumps, surface wash pumps, air scour blowers	Ozone feed

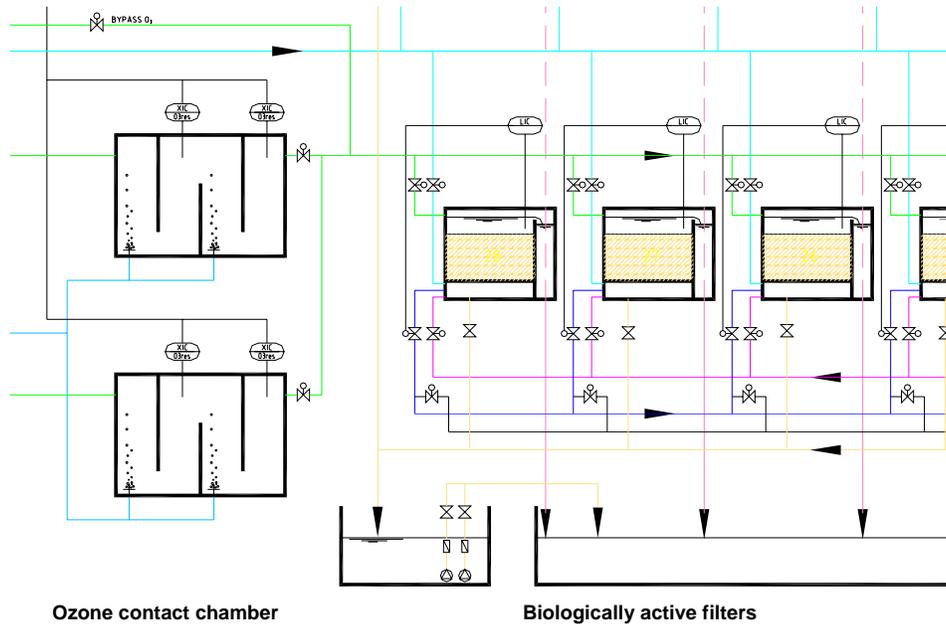


Figure 6.3. Ozonation biofiltration plant with sensors

Figure 6.3 shows a ozonation-biofiltration process scheme with a traditional placement of the sensors. Residual ozone sensors are located in two points in the contact chamber. Feed back control is being used here. Measurements of ozone are linked through the SCADA system to the ozone dosing system that automatically adjust the ozone feeding valve to compensate the difference between the set point and the measured value of residual ozone. In order to increase the accuracy of the on-line measurements, sensors can be placed in two or more points. This also allows the measurement of the kinetics of ozone consumption in the contact chamber. Biologically active filters are equipped with a level measuring sensor at the upper part of the filter, and with turbidity and pH sensors (not shown) in the outlet. The sensor is set to the certain water level at which signal from the sensor initiates the SCADA system to close the water inflow valves and start filter backwashing.

6.2.2 Instrumentation of biofilters

Instrumentation of biofilters includes sensors and actuators. The most widely used are residual ozone sensors, water level sensors, turbidity, temperature and pH meters. Lately sensors for measuring concentration of organic matter are also used, e.g. UV monitors that use UV absorption as a surrogate for organic carbon concentration. Often 254 nm wavelength is used in stead of absorption at 410 nm because of increased sensitivity of the measurements. Sensor validation is essential to the success of process monitoring (Olsson et al 2006). Any faults have to be identified and corrected before the sensor signal can be used for parameters monitoring or close loop control. The sensor data may be corrupted by abnormal noise, they may be drift in data, or the signal is too much biased. Many of these faults can be identified. There

are model-based approaches to sensor fault identification and there are data driven approach, for example via multivariate statistics.

7 PROCESS MODELING

A mathematical model is a systematic attempt to translate conceptual understanding of the real-world system into mathematical terms. Thus, a model is a valuable tool for testing our understanding of how a system works. From a practical point of view, models can be used for design of water treatment units, including biofilters, because with their help parameters of the treatment train (filter surface area, depth) can be calculated more accurately. Models can also be used to improve operation of water treatment processes. By simulating different scenarios models enable operators to adjust operational parameters like loading rates, ozone doses, and backwashing conditions, etc. to optimal levels. Models can also be applied to improve automation and control of biofilters because they allow predictions and the use of computers to simulate different patterns of control actions (e.g. ozone dose) to find the most appropriate one (Ingildsen and Olsson, 2001). Up-to-date, this kind of automation is more frequently used for operation of wastewater treatment plants.

Roughly models can be divided in **mechanistic** and **empirical** (even though all models contain certain elements of both). Mechanistic models strive to understand and mathematically describe the mechanisms behind the processes occurring in a given system (e.g. biofilters). Empirical model are simpler and often obtained by fitting mathematical equations to a set of experimental data (“black box” approach).

For ozonation-biofiltration plants models are developed both for ozonation and biological filtration. Both mechanistic and empirical models are available for ozone disinfection efficiency, ozone decay rate, and formation of AOC and bromate (van der Helm, 2006). This report is however focused on models describing the removal efficiency of NOM in ozonation-biofiltration processes.

7.1 Mechanistic models

The DOC removal in biofilters is mainly carried out by the biofilm bacteria, hence the development of mechanistic models are linked to the development of biofilm models (Figure 7.1). In general, all models describe the efficiency of BOM removal at different depths of biofilters depending on biomass, contact time and temperature (Figure 7.2).

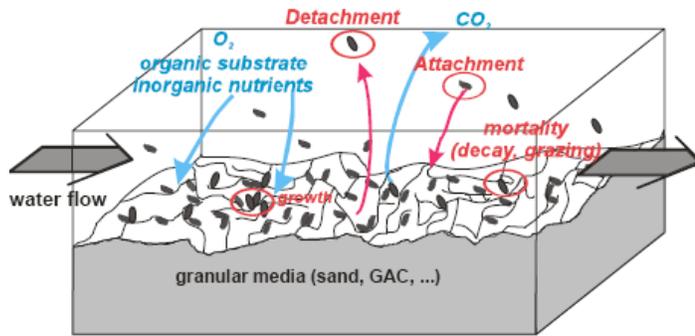


Figure 7.1 Illustration of biofilm processes occurring on the surfaces of the granular media of biofilters (Source: Uhl, 2006)

In the models, phenomena such as substrate utilisation, bacteria attachment rate, bacterial growth, bacteria mortality due to die-off and protozoa grazing are included.

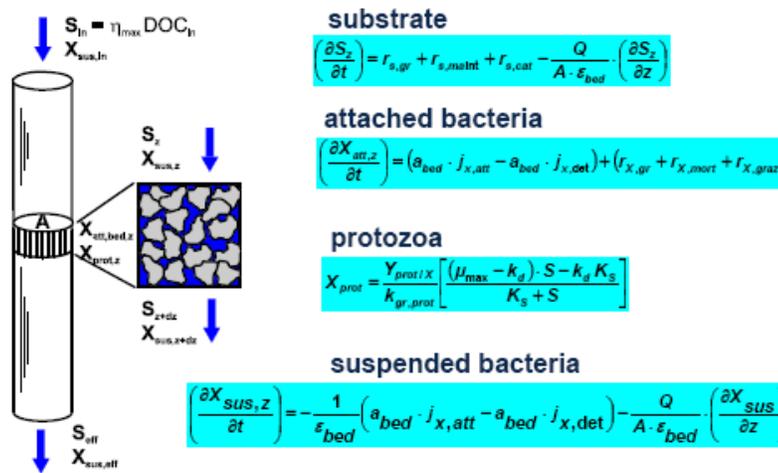


Figure 7.2 Models describing removal of DOC in a biofilter column due to various phenomena occurring in the filter (Source: Uhl, 2006)

The major factors influencing microbial conversion of DOC are (Uhl, 2006):

- biomass concentration
- energy yield of redox-reaction
- available carbon source
- available nitrogen source
- available phosphorous source
- temperature
- pH
- surface properties (attachment of microorganisms; adsorption of substrate; abiotic catalytic activity)

The most commonly used mechanistic models are presented in Table 7.1. Two models are described in more detail below.

The CHABROL steady-state model applies kinetic modelling of the utilization of dissolved organic matter by fixed and suspended bacteria in granular medium filters. This model provides as an output the vertical distribution of fixed and suspended bacterial biomass and the degradation of BDOC. The results are predicted from initial values of BDOC and temperatures at the biofilter inlet. The following processes are taken into account in this model:

Interactions with DOC

DOC is hydrolysed by enzymes into substrate which is rapidly utilised by bacteria according to the Michaelis-Menten equation (a hyperbolic relation between substrate concentration and its uptake rate). Only a part of DOC is hydrolysed. This part is determined from the BDOC test.

Interactions with solid support

Adsorption (rapid and slow) and desorption of bacteria are considered. The rate of adsorption of bacteria to the filter media is proportional to the number of bacteria in the liquid and the vacant adsorption sites on the media. Desorption is assumed to be first order with respect to the concentration of adsorption sites. Bacteria growth on the media is assumed to be proportional to their uptake of substrate.

Mortality and grazing of bacteria

Mortality of bacteria are considered both as a result of grazing by protozoa predators and die-off. Part of the bacterial material subject to mortality is considered to recycle DOC, and the remaining part is assumed to be mineralised.

The BIOFILT model is a numerical non-steady state model which was developed to simulate the non-steady-state behavior of biofilters used for drinking water treatment. The models simulate the substrate (BOM) and biomass (both attached and suspended) profiles in a biofilter as a function of time. One of the innovative features of BIOFILT compared to previous biofilm models is the ability to simulate the effects on substrate removal of a sudden loss in attached biomass or biofilm due to filter backwash.

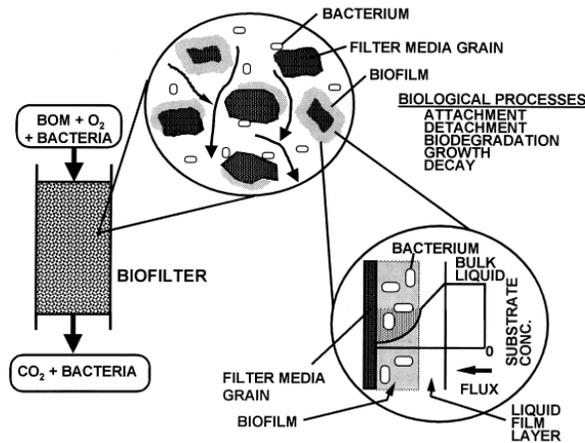


Figure 7.1. Schematic diagram that illustrates the processes occurring in a biofilter (backwash cycle excluded) (Hozalski and Bouwer, 2001)

Table 7.1 Major mechanistic models for NOM removal in biofilters

Model name	Output parameters	Number of parameters*	Verification	Reference
-	biofilm thickness, substrate flux into biofilm, bulk substrate concentration	8	-	Rittmann and McCarty, 1980
-	concentration of rapidly and slowly BDOC with filter depth	10	-	Wang and Summers, 1995
-	Biofilm thickness, concentration of suspended microbes, bulk substrate concentration	Dependent of model formulation	-	Wanner and Gjuer, 1986
BIOFILT	Non-steady-state BDOC removal, biomass	13	Full-scale (California, Mannheim, Canada)	Hozalski and Bouwer, 2001
CHABROL	BDOC removal at different depth, bacterial biomass in water in biofilm	15	In pilot and full-scale in France and Canada	Billen et al 1992
-	AOC removal	-	-	Dugan 1998
-	BDOC effluent concentration, biomass profiles, and the residual capacity of adsorption.	19	An experimental column	Liang and Chiang, 2006

* parameters that require in-depth scientific investigation (e.g. substrate diffusivity constant) are counted

Dugan (1998) has developed a model which takes into account the effect of ozone and pH on removal of AOC.

Liang and Chiang (2006) have developed a non-steady-state numerical model to differentiate the adsorption and biodegradation quantities of a biofilter. The mechanisms considered in this model included adsorption, biodegradation, convection and diffusion. Simulations were performed to evaluate the effects of some parameters such as specific biodegradation rates and diffusivities on adsorption and biodegradation performances for the removal of dissolved organic matter from water.

7.2 Empirical models

7.2.1 Ozonation models

Ozone is effective in removing colour from humic water and Figure 7.2 shows the results from batch tests carried out with different source waters. When the removal efficiency is plotted against colour specific ozone dose ($\text{mgO}_3\text{mgPt}^{-1}$), similar removal efficiencies are achieved regardless of water source and quality (Figure 7.2. b). The following model was developed to estimate the colour removal (Ødegaard, 1996):

$$\frac{C}{C_0} = \frac{1}{1 + 20 \cdot D / C_0}$$

where C_0 is colour in the raw water (mgPt L^{-1}), C is colour after ozonation (mgPt L^{-1}) and D is the ozone dose ($\text{mgO}_3 \text{L}^{-1}$).

From Figure 7.2 b it appears that the obtained colour removal efficiency for the ozonation/biofiltration process is around 80%, which is achieved at doses around $0.15\text{-}0.20 \text{ mgO}_3 \text{ mgPt}^{-1}$. By increasing the ozone dose above this level, only little additional removal was obtained. The ozonation stage is responsible for the major part of the colour removal and little additional removal in the biofilter has normally been observed (Melin and Ødegaard, 1999; Melin et al 2006).

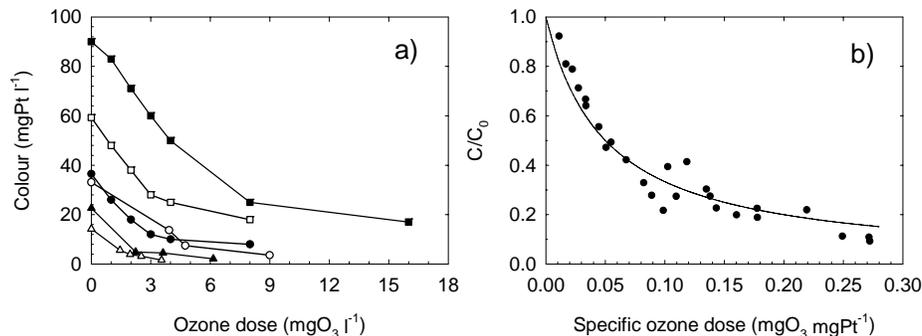


Figure 7.2. Colour removal in different source waters (a) and removal efficiency as a function of colour specific ozone dose (b). The solid line presents the model prediction (Ødegaard et al, 2006).

The removal of colour is due to modifications of the NOM structure rather than oxidation of organic matter into CO₂. Typically, TOC removal in the ozonation stage is less than 10% with the ozone doses required for about 80% colour removal (Melin and Ødegaard, 1999; Melin et al 2002), although with high dosages higher TOC removals has been observed in the ozonation stage.

7.2.2 Biofiltration models

Empirical biofiltration models usually are equations or guidelines to find a particular parameter needed for design or operation of the biofilter. Attempts have been made to model the removal of BDOC by empirically fitting first order rate reactions to data for biofilter performance. According to Uhl (2000) such an empirical approach is very well suited to describe removal of DOC and AOC in biofilters. For process analysis, the removal efficiency $\eta_z(x)$ for a parameter x at a given bed depth z is defined as:

$$\eta_z(x) = \frac{X_{inf} - X_z}{X_{inf}}$$

where the index *inf* indicated the value of the parameter at the influent, e.g. BDOC or AOC.

A maximal removal efficiency η_{max} can be expressed and defined. If a first order reaction is assumed in the interstitial water of the bed, the removal efficiency as a function of bed depth can be calculated as:

$$\frac{\eta_z}{\eta_{max}} = 1 - e^{-(k_{app} \times \epsilon_{bed}) EBCT}$$

where k_{app} is the apparent rate constant for a first order reaction, ϵ_{bed} is the interstitial porosity and EBCT is the empty bed contact time from influent to bed depth z .

The parameters should be determined experimentally. Results from the application of such a model are shown in Figure 7.3.

Huck (1999) linked the most important parameters (e.g. filter media, temperature, BOM composition), and developed an equation that essentially defined the depth of a biofilter column required to achieve a given BOM removal. The reactor depth was transformed into a dimensionless empty bed contact time, termed X^* which is calculated from EBCT, specific surface area, diffusivity of substrate in the biofilm, half velocity constant in the Monod expression, maximum specific rate of substrate utilisation and biofilm density.

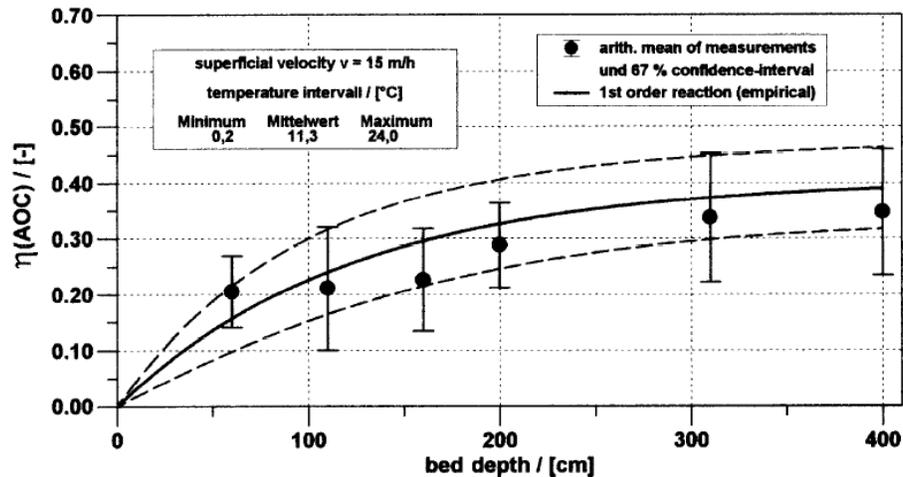


Figure 7.3. AOC removal efficiency as a function of bed depth, with empirical modelling of first order reactions (Uhl, 2000)

7.3 Application of models for operation and maintenance

Although mechanistic models can predict accurately the performance of biofilters, the main limitation of these models is that they require a lot of parameters (usually around 10 - 20) to be calibrated for every new water treatment plant where they are intended to be used. Often, this calibration requires intensive scientific investigations.

The main limitation of empirical models is that they can only be used to estimate the performance of similar systems as long as the operating conditions are within the range of the evaluated data. They are not as accurate as mechanistic models. However, the number of required parameters is lower, and usually the determination of parameter values can be done by laboratory staff at the water treatment plant. For instance, a simple attached growth reactor can easily be set up and used for optimising biofilters. This allows for measurements of the rate of BOM decomposition, from which major parameters such as EBCT and filtration rate can be determined (Digiano et al 2001).

8 TREATED WATER CHARACTERISTICS

The overall objective of biofiltration is to remove TOC and provide biostability of the drinking water. Even though TOC and DBPs are regulated by water quality standards in Europe and USA, BOM to secure biostability of drinking water is not regulated.

8.1 Regulations on organic substances in drinking water

In the European Union (EU), drinking water quality is regulated by Council Directive 98/83/EC. The total amount of organic substances in drinking water is reflected by two parameters: TOC and oxidability (sometimes termed Chemical Oxygen Demand or COD).

Indirect control of organic carbon in water is done by limiting the colour level (normally due to humic substances) and the THMs (formed from reactions of organic matter with disinfectants).

There is no maximum allowable concentration limits for colour and TOC set in the Drinking Water Directive (DWD). TOC should be analysed only for systems producing more than 10 000m³/day, and “no abnormal changes” should occur over time. Colour should be “acceptable to consumers, and no abnormal change” is allowed. If TOC measurements are not possible (e.g. due to lack of TOC detection apparatus), organic matter could be evaluated by measuring oxidability. This is an old test in which substances in a water sample are oxidised with permanganate (for 10 minutes in acid conditions at 100 °C). Consumption of permanganate is expressed as mgO₂/L. The maximal value should be less than 5 mg O₂/L according to the DWD. The limitation of this method is that not all organic substances are oxidised with permanganate, thus this test does not always show the true concentrations of total organic matter.

Council Directive 98/83/EC is however regulating by-product formation from reactions between TOC and disinfectants. There are many by-products of chlorination and ozonation for which the chemical structure is known (Figure 8.1). A significant part however, is still not identified. A maximal allowed concentration of 150µg/L is set for one group of DBPs, namely the trihalomethanes (THMs) which include compounds such as chloroform, bromoform, dibromochloromethane, and bromodichloromethane.

EU countries are allowed to set stricter requirements and/or include more parameters in their national drinking water standards. Several Member States have introduced stricter requirements for THMs, and maximum numerical values for colour and TOC to replace the unprecise terms “acceptable to consumers and no abnormal change”.

In the United States the Environmental Protection Agency (USEPA) has issued specific regulations regarding TOC in drinking water, i.e. the Disinfectants/Disinfection By-products Rule, D/DBPR (1st stage). Under this rule, utilities are required to remove predetermined amounts of TOC as a way to reduce DBP precursors and DBP formation (Table 8.1). The required amount of TOC to be removed as defined by the B/DBP rule (Table 8.1) is affected by the raw water TOC concentration and raw water alkalinity. The treatment goals and requirements are based on evaluations of DBP formation potentials and economical and practical reasons.

Table 8.1 Total organic carbon (TOC) removal goals (USEPA)

TOC in raw water (mg/L)	TOC removal goals (%) for different raw water alkalinity levels (mg/L CaCO ₃)		
	0-60 mg/l	60-120 mg/l	>120mg/l
2.0-4.0	35	25	15
4.0-8.0	45	35	25
>8.0	50	40	30

The 2nd stage Disinfectants/Disinfection By-products Rule (USEPA) also regulates two groups of disinfection by-products which are originating from organic matter: Total THMs, and a group of five haloacetic acids (HAA5). The maximum THMs value of 80 µg/L is stricter than that in the EU Directive 98/83/EC, and the maximum value for HHA5 is 60 µg/L.

8.2 Disinfection byproducts - DBPs

One of the reasons for removal of TOC with biofiltration is the need for reduction of DBP formation. THMs and HHA5 contribute less than half of the total halogenated organic matter found in drinking water (Figure 8.1). The other substances are not yet identified or the potential risk to humans is still unknown. Due to their aromatic character, the humic and fulvic acid fractions of NOM are considered main precursors of DBPs. These humic NOM fractions can be effectively removed by biofiltration only if they are converted to biodegradable organic matter (BOM) during preozonation/pre-oxidation.

Several mathematical models have been developed (Table 8.2) to predict DBPs formation potentials. In general, THMs formation is depended on TOC, UV-abs, chlorine dose, pH and temperature.

Table 8.2 Models for prediction of DBPs formation

Model	Reference
$HHA5 = 4.8 \times 10^4 [OH^-]^{0.35} (C_0[1-\exp\{-kT\}])^{0.43} (UV_{254})^{0.34}$	Sung <i>et al</i> (2000)
$TTHMs = a [OH^-]^j (C_0[1-\exp\{-kT\}])^m (UV_{254})^n (\text{algae})^p$	Sung <i>et al</i> (2000)
$TTHMs = b (DOC)^{b1} (Cl_2)^{b2} (Br^-)^{b3} (\text{temperature})^{b4} (pH)^{b5} (\text{Time})^{b6}$	Westerhoff <i>et al</i> (2000)

During ozonation and chlorination several by-products, both organic (Figure 8.1) and inorganic (e.g. bromate) can be formed. However, biofilters is able to effectively remove most of the DBPs formed from preozonation (Speitel et al 1993; Kirists et al 2001)

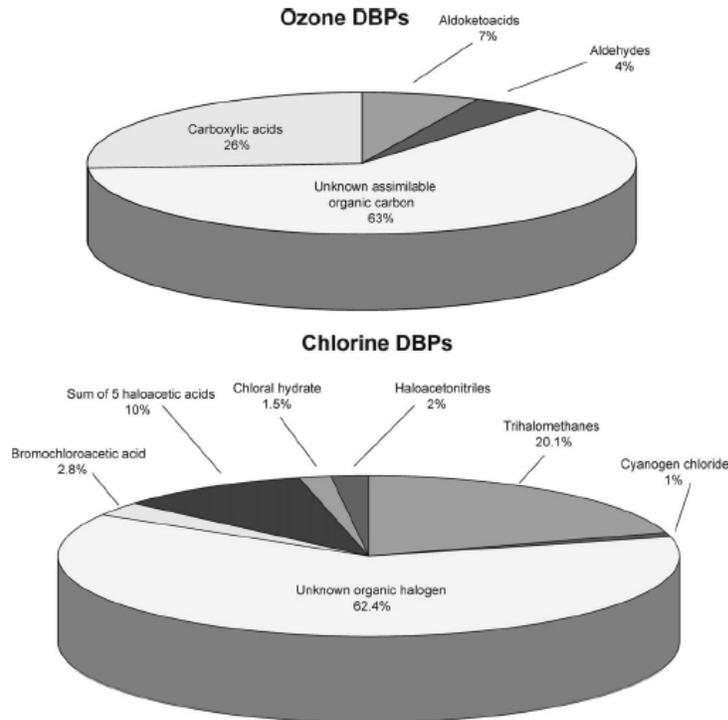


Figure 8.1. Amounts of DBPs in drinking water from ozonation (upper) and chlorination as proportions of the total assimilable organic carbon (ozonation), and of the total organic halogen in chlorinated drinking water (Richardson 2003).

8.3 BOM in treated water

Efficient removal of BOM (AOC or BDOC) is important to provide biostability of drinking waters. Under optimal operation conditions ozonation-biofiltration processes provide sufficient removal to eliminate bacterial regrowth problems in distribution networks. Operating problems may arise if the preozone dose is too high, and slowly degradable organic matter - BDOC_{slow} are formed (**Chapter 2**). These fractions of NOM are not significantly removed by biofilters, because of the limited contact time. Thus they may enter the distribution network and produce regrowth problems. Normally, the removal efficiency for TOC during biofiltration varies in the range of 15-30% (Table 8.3), for AOC in the range of 40 to 80% (Table 8.4), and for BDOC in range 25-80% (Table 8.5).

Table 8.3. Reported TOC removals in ozonation-biofiltration processes

Media	Influent water	O ₃ dose (mgO ₃ mgTOC ⁻¹)	EBCT (min)	TOC _{in} (mg L ⁻¹)	Removal (%)	Reference
GAC	SW-O ₃	0	10	3.18	14	Klevens <i>et al</i> , (1996)
GAC	SW-O ₃	0.5	5	3.18	21	Klevens <i>et al</i> , (1996)
GAC	SW-O ₃	0.5	10	3.18	23	Klevens <i>et al</i> , (1996)
AS	SW-O ₃ -Co-Se	0.8	9.2	1.1-2.2	16	Wang <i>et al</i> , (1995)
Sand	"	0.8	9.2	1.1-2.2	20	Wang <i>et al</i> , (1995)
GAC (bituminous coal)	"	0.8	9.2	1.1-2.2	29	Wang <i>et al</i> , (1995)
GAC (lignite coal)	"	0.8	9.2	1.1-2.2	27	Wang <i>et al</i> , (1995)
GAC (wood)	"	0.8	9.2	1.1-2.2	21	Wang <i>et al</i> , (1995)
GAC	Se-O ₃	0.78	14	3.85	19	Carlson <i>et al</i> , (1994)
GAC	GW-O ₃	3.8	9	5.2	23	Tan and Johnson (2001)
GAC	SW-MF-O ₃	2.5	15	2.8-4.2	36	Nishijima <i>et al</i> , (1998)
GAC	SW-Co-Fi-O ₃	NG	10	1.73	14-15	Servais <i>et al</i> , (1994)
Filtralite	SW-O ₃	1.1	16	5.0	23	Melin and Ødegaard (1999)
Filtralite	SW-O ₃	1.7	27	4.7	31	Melin and Ødegaard (1999)
GAC-sand	SW-Co-Se-O ₃	NG	24	NG	23	Emelko <i>et al</i> , (2006)
GAC	SW-O ₃ -Co-Se-Fi-O ₃	1-2 mgO ₃ L ⁻¹	7-12	1.70-2.95	21	Servais <i>et al</i> , (1991)
GAC	SW-Cl-Co-Se-Fi-O ₃	0.7-1.6	20	1.7-2.2	30-59 ¹	Griffini <i>et al</i> , (1999)
Biolite	SW-Cl-Co-Se-Fi-O ₃	0.7-1.6	20	1.7-2.2	18	Griffini <i>et al</i> , (1999)
Coke	SW-RiBF-O ₃	1	24	2.8	17	Wricke <i>et al</i> , (1996)
GAC-phonolith-CaCO ₃	SW-O ₃	1.5	28	2.4-3.9	23	Melin <i>et al</i> , (2002)

1) Range for 4 different types of GAC

Table 8.4. Reported AOC removals in ozonation-biofiltration processes

Media	Influent water	O ₃ dose (mgO ₃ mgTOC ⁻¹)	EBCT (min)	AOC _{in} (µg L ⁻¹)	Removal (%)	Reference
AS ¹	SW-O ₃ -Co- Se	0.8	9.2	NG ²	39 ³	Wang <i>et al</i> , (1995)
Sand	"	0.8	9.2	NG	43 ³	Wang <i>et al</i> , (1995)
GAC1	"	0.8	9.2	NG	51 ³	Wang <i>et al</i> , (1995)
GAC2	"	0.8	9.2	NG	47 ³	Wang <i>et al</i> , (1995)
GAC3	"	0.8	9.2	NG	42 ³	Wang <i>et al</i> , (1995)
GAC	GW-O ₃	3.8	9	598	58	Tan and Johnson (2001)
GAC-Sand	SW-O ₃ -Co- Se	2.5-2.8 mg L ⁻¹	10	368	80	LeChevalier <i>et al</i> , (1992)

1) AS = anthracite-sand

2) NG = Not given

3) Only AOC-NOX included; P17 values very low

Table 8.5. Reported BDOC removals in ozonation-biofiltration processes

Media	Influent water	O ₃ dose (mgO ₃ mgTOC ⁻¹)	EBCT (min)	BDOC _{in} (mg L ⁻¹)	Removal (%)	Reference
GAC	SW-O ₃	0	10	0.47	26	Klevens <i>et al</i> , (1996)
GAC	SW-O ₃	0.5	5	1.05	39	Klevens <i>et al</i> , (1996)
GAC	SW-O ₃	0.5	10	1.05	46	Klevens <i>et al</i> , (1996)
GAC	SW-Co- Fi-O ₃	NG	10	0.41	49-56	Servais <i>et al</i> , (1994)
GAC	SW-O ₃ - Co-Se-Fi- O ₃	1-2 mgO ₃ L ⁻¹	7-12	0.40- 0.90	40	Servais <i>et al</i> , (1991)
GAC	SW-Cl- Co-Se-Fi- O ₃	0.7-1.6	20	0.32- 0.59	70-74 ¹	Griffini <i>et al</i> , (1999)
Biolite	SW-Cl- Co-Se-Fi- O ₃	0.7-1.6	20	0.32- 0.59	66	Griffini <i>et al</i> , (1999)
Coke	SW-RiBF- O ₃	1	24	0.75	47	Wricke <i>et al</i> , (1996)
GAC- phonolith- CaCO ₃	SW-O ₃	1.5	28	0.75-1.2	55	Melin <i>et al</i> , (2002)

1) range for four different types of GACs

8.4 Bacterial endotoxins

Biofilters contains high concentrations of bacteria that under certain circumstances may produce several by-products. Gram-negative bacteria, which represent most bacteria living in water, have cell walls made principally of lipopolysaccharide and lipoprotein complexes. During cell lysis (death), lipopolysaccharide endotoxins are released. Thus, bringing biofilters on-line after a few days out of service may lead to the presence of elevated concentration of bacterial endotoxins (see references in Niquette et al 1998).

8.5 Microorganisms and turbidity in treated water

For surface water treatment plants turbidity is used as a tool to control treatment plant performance and to secure optimum operation. Efficient control of filter effluent turbidity and/or particle count is also part of the pathogen control strategy. In case of biofiltration, considerable amounts of heterotrophic bacteria are released from the filters. The heterotrophic bacteria count (HPC) leaving the filter can be high, ranging from 10^2 to 10^6 CFU mL⁻¹ (Table 8.6). Possible health-related effects of HPC have been discussed extensively. It is agreed however, that the potential effects do not justify inclusion of HPC as a parameter in the Drinking Water Directive. Nevertheless some countries have taken additional precaution and have set a maximum limit of 100 CFU mL⁻¹ in their national standards.

Table 8.6. Effluent steady-state HPC levels in biofilters receiving ozonated water.

Media	HPC (bacteria/CFU mL ⁻¹)	Method	Reference
GAC	In order of 10^5		Tan and Johnson (2001)
GAC-sand	250-650	R2A, 7 days, 20°C	Najm <i>et al</i> , (2005)
GAC	$8 \times 10^4 - 15 \times 10^4$	Epifluorescence microscopy	Servais <i>et al</i> , (1994)
GAC	$1.1 \times 10^5 - 4.0 \times 10^5$	Epifluorescence microscopy	Servais <i>et al</i> , (1991)
GAC-sand	$1.2 / 7.5 \times 10^4$	R2A, 7 days, 20°C	LaChevalier <i>et al</i> , (1992)
Anthracite-sand-garnet	1.4×10^4	R2A, 7 days, 20°C	LaChevalier <i>et al</i> , (1992)
GAC	$0.7-1.0 \times 10^5$	R2A, 7 days, 20°C	Griffini <i>et al</i> , (1999)

Servais *et al*, (1994) observed the highest bacteria counts in GAC effluents during colonisation of the biofilters. More bacteria are exported, relative to the biomass fixed to the GAC during the early stages of biofilter operation, as compared to the later stages when biological equilibrium is reached (after 150 days). A gradual development of the population of bacterial grazers is one possible explanation.

Once steady-state is established, the carbon conversion efficiency is remarkable. Elimination of about 0.25 mgC L⁻¹ as BDOC resulted in the release of less than 0.0015 mgC L⁻¹ (0.5×10^5 bacteria mL⁻¹) as bacterial biomass in the effluent (Servais et al 1994).

In general biofilters are effective in removing turbidity entering from the raw water source (Goldgrabe et al 1993). However, the research has shown that due to changes of the surface properties of the media (e.g. biofilm development), the potential for *Cryptosporidium parvum* oocyst breakthrough is increasing with time (Dai and Hozalski, 2002). Proper pretreatment, e.g. coagulation prior to biological treatment is therefore critical for effective removal of oocysts.

To decrease HPC number in the biofilter effluent, chlorine is usually used as a final water treatment step. However, chlorination can oxidise BDOC_{slow} (see **Chapter 2**) and therefore decrease the biostability of the water. To mitigate this problem some biofiltration plants have introduced an additional ultrafiltration step to reduce the number of bacteria entering the distribution network.

Emelko et al 2006 reported that the biofilter effluent turbidity was always below 0.1 NTU in a full-scale plant treating surface water with turbidity in the range of 0.9-13.7 NTU by flocculation, sedimentation, ozonation and parallel GAC-sand and anthracite-sand filters. This shows that biofiltration can have a high standard of particle removal.

8.6 Elevated oxygen levels in treated water

To increase production efficiency several full-scale plants are producing ozone from oxygen instead of air. This can result in high levels of oxygen in the treatment plant effluent. This may affect microbial growth and corrosion in the distribution networks.

9 CASE STUDIES

9.1 Leiduin water treatment plant, Amsterdam Water Supply (Netherlands)

Operation of the Leiduin water treatment plant has been reviewed by van der Hoek et al (2000). The raw water source is the River Rhine. The water is pretreated by coagulation, sedimentation and rapid sand filtration and subsequently transported to the dune area west of Amsterdam where it is infiltrated. After a residence time of approximately 100 days the water is abstracted and collected in an open basin. This recharged water is treated by rapid sand filtration, ozonation, pellet softening, two-stage biological activated carbon filtration and slow sand filtration. No final disinfection is applied. The ozonation-biofiltration step was installed in 1995. One of the main reasons for this was the presence of organics such as pesticides, and the need for additional reduction of DOC and AOC. In addition, additional disinfection capacity was provided. Also, the ozonation-biofiltration step has increased the run time of the slow sand filters.

Table 9.1. Leiduin water treatment plant (van der Hoek et al., 2000).

Production capacity	70 million m ³ yr ⁻¹ 8000 m ³ h ⁻¹		
Surface infiltration	Residence time ca. 100 days (60-400 days)		
Rapid sand filtration	4 m h ⁻¹		
Ozonation	0.75-0.9 mgL ⁻¹ 0.38-0.45 mgO ₃ mgDOC ⁻¹ (min-max 0.5-2.0 mgL ⁻¹)	Contact time 20 min (min 15 min)	
Pellet softening			
BAC (two filters in series)	EBCT 20 + 20 min	Filtration rate 7 m h ⁻¹ (max 10 m h ⁻¹)	Norit ROW 0.8S Regenerated once in two years
Slow sand filtration	0.25 m h ⁻¹ Max 0.5 m h ⁻¹	Covered	Running time at least 2 years

The plant has two GAC filters in series. After a total operational period of 2 years, the first stage filter is regenerated, and put back into service as the second stage filter. After 1 year of operation the adsorption capacity is exhausted (DOC removal around 70%) and this filter is switched to the first stage mode and operated for another year. During this time the DOC removal in this filter is 20-30%. The influent DOC into the 1st stage BAC filter is 2.0±0.2 mg L⁻¹ and the effluent DOC is 1.6±0.2 mg L⁻¹. The GAC in the filters is regenerated sequentially so that every 2-3 weeks a filter bed is sent to the carbon supplier for regeneration (the plant has 40 filters in total). The DOC of the combined second stage BAC filter effluent is around 1 mg L⁻¹.

With an ozone dose of 0.9 mg L⁻¹, and with average hydraulic load, the CT-value is 2 mg min L⁻¹. After ozonation, AOC increases to 50-150 µg L⁻¹. Table 9.2 shows the biological stability of the treated water.

Table 9.2. Biological stability of the finished water (average ± standard deviation) before and after the introduction of ozonation-biofiltration, i.e. biological activated carbon filtration - BACF (van der Hoek et al, 2000).

	Before introduction of BACF	After introduction of BACF
AOC (µg L ⁻¹)	5.1 ± 1.1 (n=8)	5.4 ± 1.7 (n=20)
BFR (pg ATP cm ⁻² day ⁻¹)	0.25 ± 0.03 (n=16)	0.84 ± 0.14 (n=8) ¹ 0.25 ± 0.03 (n=9) ²

¹March 1997–August 1997.

²September 1997–January 1998.

9.2 Pitkäkoski and Vanhakaupunki WTPs, Helsinki Water (Finland)

Helsinki water has two water treatment plants that use the same treatment processes, Pitkäkoski and Vanhakaupunki treatment plants (Figure 9.1). The water is taken from Lake Päijänne and transported through a 120 km long tunnel to the treatment plants. The humic substances are precipitated with ferric sulphate and the particle separation is carried out by settling and sand filtration. After pH-adjustment with lime water, primary disinfection is done with ozone and most of the organic residual is then removed by two-stage activated carbon filtration. Bacteria from the biological carbon filter effluents are destroyed with UV-disinfection. In addition, some combined chlorine is used to reduce bacterial growth in the distribution system. For corrosion control, carbon dioxide is added and the pH-value is adjusted to 8.5 with lime. Some major operational characteristics are presented in Table 9.3.

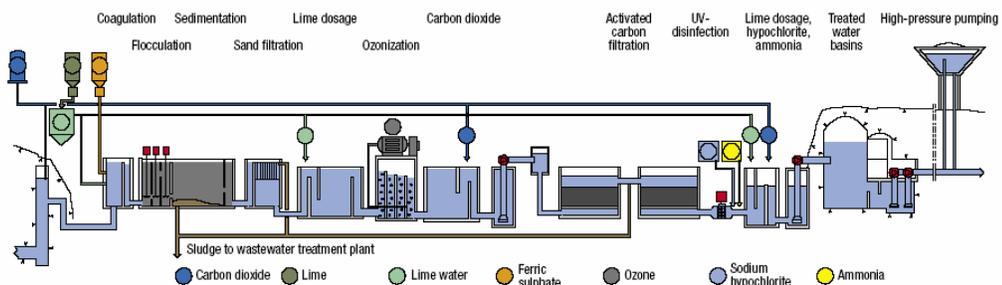


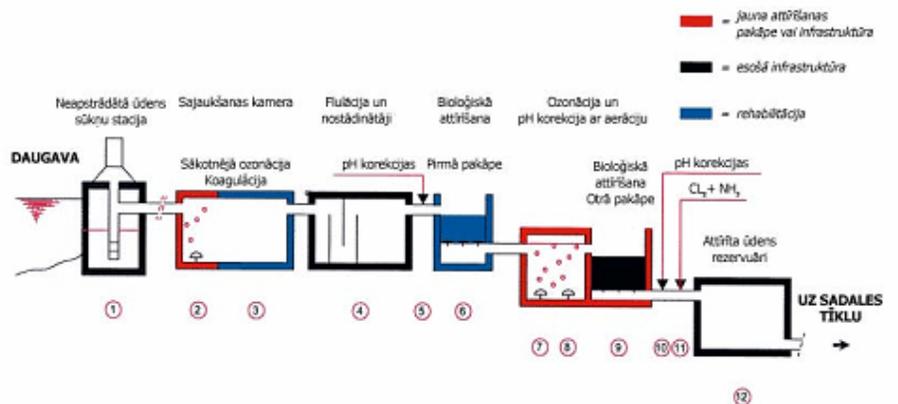
Figure 9.1. Schematic of the treatment process in the water treatment plants in Helsinki.

Table 9.3. Operation characteristics (mean values) for Helsinki Water treatment plants.

	Pitkääkoski	Vanhakaupunki
Mean water production (m ³ h ⁻¹)	4680	4320
Coagulant	Ferric sulphate	Ferric sulphate
Flocculation	Vertical agitators with vertical axis	Propeller agitators
Sedimentation surface load (m h ⁻¹)	0.36-0.37	0.87
Sand filters filtration rate (m h ⁻¹)	2.5-2.7	3.0
Sand filter backwash interval (weeks)	1	1
Ozone target concentration (mgL ⁻¹)	0.4	0.4
Contact time (min)	12	12
BAC filtration rate (m h ⁻¹)	7.8	10.3
Backwash interval 1st stage (weeks)	1	1
Backwash interval 1st stage (weeks)	0.5	0.5

9.3 Daugava water treatment plant, Riga (Latvia)

Daugava water treatment plant is the largest plant in Latvia supplying about 50% of the drinking water for Riga city. The plant takes water from the River Daugava (from the reservoir of Riga hydroelectric power station). Upstream the intake several villages and cities are located (in Russia, Belarussia and Latvia), therefore the raw water is influenced by wastewater discharges. To reduce the pollution risk of the drinking water the treatment process was upgraded with two stage ozonation in 2001. The watershed of the River Daugava is largely covered with swamps and forests; therefore, water in the river contains a high amount of humic substances (NOM). During a study period the concentration of DOC in the river varied between 9 and 16 mg/L. To remove DOC and increase biostability, biofilters have been installed after the main ozonation. Ozon is also used prior to coagulation.



- 1- water intake; 2- ozonation; 3-coagulation (alum), 4-sedimentation, 5-pH correction, 6-filtration, 7-ozonation, 8-airation, 9- biofiltration, 10- pH correction, 11-final chlorination, 12- clean water reservoir

Figure 9.2. Scheme of the Daugava water treatment plant in Riga. Newly added treatment steps are shown in red.

About 50% of the DOC is removed by coagulation, any further removal of DOC in the biofilters is not occurring, however (Juhna and Rubulis, 2004). During ozonation humic substances are oxidized which results in a significant reduction of UV_{abs} and an increase of the BDOC value to 1-2 mg/L. The DOC concentration after treatment is in range of 2-8 mg/L, which is considered as high. As a result of this, chlorine consumption is high and there are bacterial regrowth problems in the distribution network.

9.4 Klungset water treatment plant, Fauske (Norway)

Klungset water treatment plant was the first public water treatment plant in Norway to apply ozonation and biofiltration (Melin et al, in preparation). The design flow is $144 \text{ m}^3 \text{ h}^{-1}$. The raw water from a small reservoir is filtered through a 0.3 mm sieve, after which ozone is introduced by a side stream injector recirculating the water from the contact tanks. After the contact tanks, the water is treated in biological filters containing GAC. Ozonation and biofiltration is carried out in three parallel independent lines. Final disinfection is carried out by UV (Figure 9.3).

The plant has an automatic control system to adjust the ozone dose based on ozone concentration after the contact tanks. However, for proper operation the ozone sensors had to be calibrated weekly and this turned out to be a very time consuming task. Large variations in ozone dose were also observed, which might have been due to improper control systems. From these reasons, this control system is not presently used and the operator sets manually the desired ozone dose, which is then automatically maintained according to the water flow. The operator sets the ozone dose based on an estimation of raw water colour. The ozone dose is normally $3.5\text{-}5.0 \text{ g m}^{-3}$.

Backwashing of biofilters occurs either based on time, turbidity breakthrough or head loss. Normally, the biofilters are backwashed after 21 days of operation. Water from the clear water tank is used for backwashing which lasts 9 minutes. The backwash flow 52 L s^{-1} which gives a backwash water volume of about 1.3 times the biofilter tank volume. During filter ripening the effluent water goes to waste. The ripening takes about 25 min and is finished when the turbidity of the water is below 1.5 NTU.

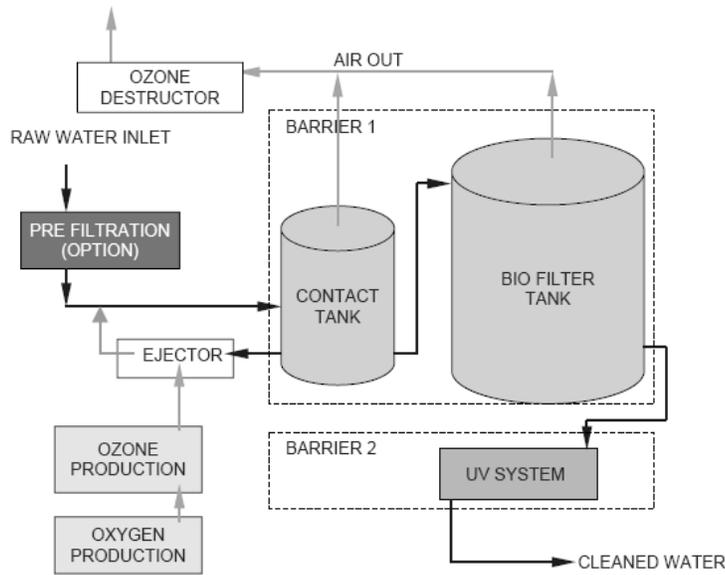


Figure 9.3. Process scheme of the Klungset WTP (Ried and Mielcke, 2006).

9.5 Styrum-East water treatment plant, Mülheim (Germany)

The Styrum-East WTP is based on the “Mülheim process” which was developed in 1970s (Figure 9.4). The plant receives water from the River Ruhr. The water is pre-treated by slow sand filtration, and artificial ground water recharge with soil infiltration for 1-2 days. The water is then extracted from collecting wells and ozonated. The ozonated water is treated by biological double layer filtration and BAC filtration. The final disinfection is carried out by UV.

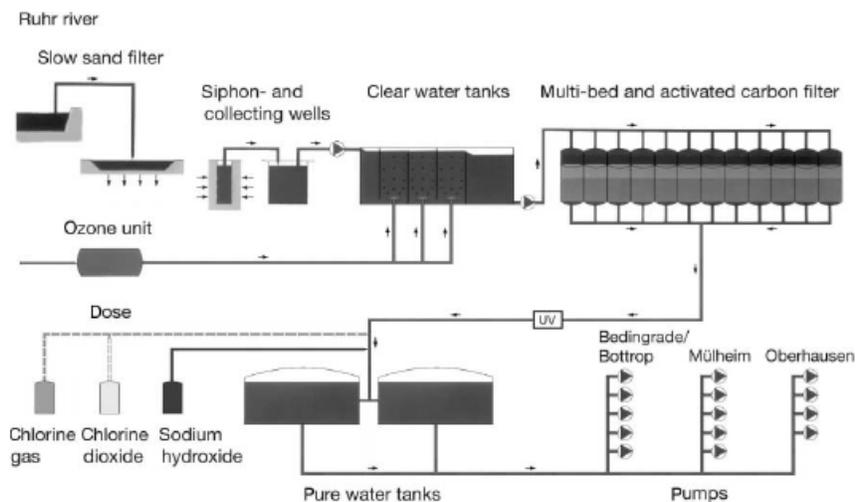


Figure 9.4. Process scheme of the Styrum-East WTP (Bundermann, 2006).

10 KNOWLEDGE GAPS AND RESEARCH NEEDS

Most of the studies reviewed here have concentrated on ozonation and biofiltration after coagulation and particle separation. However, especially many small treatment plants use ozone as the only pre-treatment before biofiltration. More studies should be carried out to determine optimal design and operation of ozonation/biofiltration plants as an independent unit process (e.g. backwash routines, optimal ozone doses, etc.)

The microbiology and kinetics of phosphorus limited biofiltration is not well understood at the moment.

More studies should be carried out in order to study the effects of seasonal changes in raw water quality and adverse conditions on biofilter operation and design (e.g. NOM content and NOM composition, low temperature, P-limitation, changes in raw water turbidity).

There is need for rapid monitoring equipment or suitable surrogates for monitoring BOM removal in biofilters.

There is a need for better understanding of the effects of particle accumulation on biofilter performance, including the effects of backwash routines.

The role of dead bacteria from disinfected biofilter effluents as substrate for micro-organisms in distribution systems is not comprehensively studied.

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