



WATER TREATMENT BY ENHANCED COAGUATION

*OPERATIONAL STATUS AND
OPTIMIZATION ISSUES*

TECHNEAU

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



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Water treatment by enhanced coagulation – Operational status and optimization issues

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1 Introduction

1.1 Optimum operation of water treatment facilities – best operation practices

Sub-optimal operation of water treatment facilities is a relatively widespread phenomenon. The reasons for this may be several, including missing focus on operational issues and challenges in general; lack of driving forces for optimization efforts; suboptimal 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 operation 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.

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.

For these reasons, it is important to identify major operational challenges, optimization potentials and needs, and best operation practices of water treatment facilities.

1.2 NOM removal and enhanced coagulation

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, thus reducing their disinfection power;
- influences disinfectant demand, and disinfection process design, operation and maintenance;
- produces 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;
- forms complexes with and increase mobility of most chemical substances found in nature;
- fouls membranes;
- reduces adsorption capacity of GAC/PAC by pore blocking
- competes with taste and odour for adsorption sites in GAC/PAC

Bursill et al (2002) stated that NOM of microbial, animal and vegetable origin in reservoir catchments is the key factor influencing most, if not all water treatment processes. With respect to coagulation process operation NOM is a key parameter because it will control coagulation processes in most cases (Edzwald and Tobiason, 1999). 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.

Contrary to conventional coagulation processes aimed primarily at turbidity removal, enhanced coagulation implies the use of elevated coagulant dosages and strict control of pH. The implications of this shift in treatment target and operating conditions includes elevated sludge production rates, increased solids load to subsequent separation processes (i.e. settling, flotation and/or filtration units), use of inorganic acids for pH-control, increased focus on operation and optimization issues, possible conflicts in optimum conditions for various target parameters like turbidity, NOM and micro organisms. In addition, coagulated NOM will form loose flocs and lead to early filter breakthroughs, i.e. shortened filter runs compared with conventional coagulation and filtration processes for the removal of turbidity. Therefore, NOM is a key target parameter in this report.

Despite the important impacts of NOM on water quality and treatment issues and the trends towards increasing NOM levels that are observed in many countries, the EU drinking water directives do not reflect the importance of NOM-related parameters and NOM removal issues to the same extent as the American and Australian regulations. However, several European countries have adopted specific and strict national regulations with respect to NOM and NOM control.

1.3 Report scope and objective

This report will address major operational issues, operational challenges and bottlenecks with the goal to identify optimization issues and best operation practises of enhanced coagulation-filtration processes.

Special emphasis is given to NOM and NOM removal. This is mainly due to the fact that NOM controls the coagulation and filtration processes in many cases. As part of this focusing, NOM treatability and characterisation methods are covered as well (**Chapter 2**).

After some initial descriptions of terms and definitions used, the report covers raw water characterisation and treatability by coagulation, i.e. available characterisation methods and known links between raw water characteristics and treatability (SUVA, NOM fractions, etc) (**Chapter 2**).

Plant operation practices are reviewed from available operational experience, based on literature and communications with existing WTPs (**Chapter 3**). For coagulation, the major operational parameters covered include coagulant types and coagulant dose requirements, mixing, residual coagulant, pH-control, etc. With respect to operation of separation, the operational parameters include pre flocculation, hydraulic loading, solids load, sludge removal, backwashing, etc. for the three filtration concept evaluated: 1) Conventional filtration (with pre sedimentation or pre flotation), 2) Direct filtration (with flocculation units but without any pre separation), and 3) Contact filtration (without separate flocculation or pre separation steps).

Major operational challenges and bottlenecks are reviewed (**Chapter 4**): Low temperatures, pH, use of polymers as coagulant, flocculant and/or filter aids, loose flocs/short filter runs/early breakthrough, filter ripening, filter backwashing, effects of and handling of return flows, etc.

Furthermore, the report addresses 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 Norway, Finland and Latvia (**Chapter 9**). Finally, the report addresses knowledge gaps and research needs (**Chapter 10**), which are considered valuable input for the design and performance of the pilot and full scale investigations.

The main objective of this report is to identify best operation practices of enhanced coagulation facilities for drinking water treatment in general and NOM removal in specific. The main focus will be on the existing water treatment facilities.

Integration, e.g. linking raw water characteristics and water treatability, treatment process control, risk reduction, etc. is a major objective within the Techneau project. For the optimization efforts on enhanced coagulation and filtration 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 reviews, operational experiences and consultations with operators from several treatment plants.

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
- BDOC - Biologically Degradable Organic Carbon** is the fraction of DOC that 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 to 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. Thus, adsorption 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

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

- 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:** A 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
- 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, which are useful for the characterization/fractionation of NOM.

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

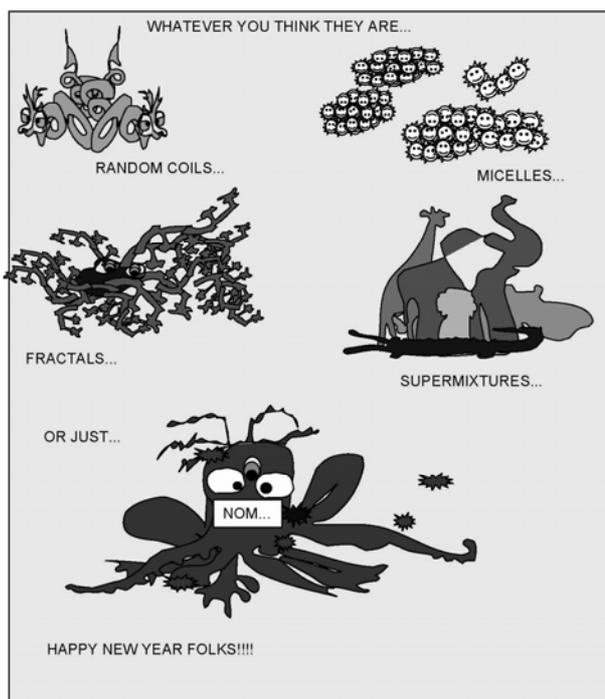


Figure 2.2 Proposed NOM configurations (IHSS, Newsletter, 2006).

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 $\text{L}/\text{mgC m}$.

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 characterisation

Natural organic matter (NOM) may have distinctive characteristics associated with its origins (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 (Skjelkvale, 1995, Eikebrokk et al 2004).

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) presents 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 in 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

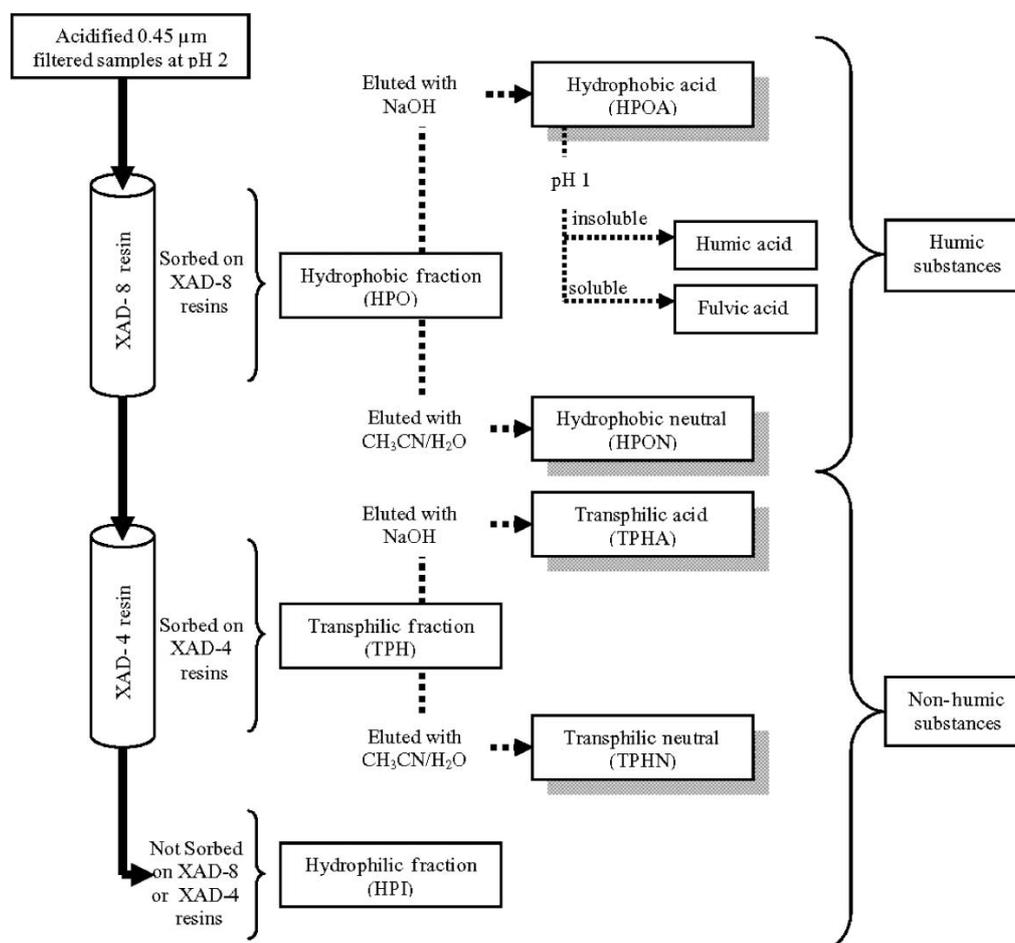


Figure 2.3. XAD-8/XAD-4 fractionation scheme (Chow et al 2005) adapted and modified from Aiken et al (1992) and Croué et al (1999).

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 the 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 the 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 reported by Croué et al (1994) and Bolto et al (1999). 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 (Leenheer, 1981, van

Leeuwen et al 2004, Buchanan et al 2005). Specifics of the technique and definitions have been described elsewhere (Chow et al 2004).

The order for ease of removal by coagulation is generally VHA > CHA=SHA > NEUT (Leeuwen et al 2002; Chow et al 2004). It was demonstrated that the removal of particular fractions was dependent upon treatment conditions such as applied coagulant dose, pH, etc. and that treatment conditions can be optimized based on the character of the organic matter present in the raw water (Chow et al 2004). A wide range of alum doses (30-180 mg/L of alum) was used to simulate situations of underdosing, conventional alum treatment, enhanced coagulation treatment, and extreme coagulant overdosing. The results show that the NEU fraction was hardly removed by alum coagulation. The CHA fraction was readily removed in the underdosing and in the extreme overdosing situations. In the intermediate dose range covering conventional to enhanced coagulation, both the SHA and the VHA fractions were preferably removed. It was stated that the rapid fractionation technique could be applied as a tool in the operation of the treatment plant and for guidance treatment operators to control and monitor the treatment process in the most effective way for NOM removal.

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 μm), 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 μm , i.e. DOM) is predominant in most water sources, and contains humic substances (HS) as a major class of organic compounds, in addition to 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 (Thurman et al 1982).

Molecular weight fractionation is commonly achieved by ultrafiltration (UF) or size exclusion chromatography (SEC). Unlike XAD-8 and XAD-4 fractionation, UF and SEC do 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, while the fraction < 1kDa is less amenable to removal by coagulation.

2.6 Biodegradable organic matter (BOM)

In recent years, attention has focused on the carbonaceous organic matter that can be used by microorganisms as a source of nutrients. This fraction of DOC is called biodegradable organic matter (BOM).

BOM is mainly composed of non-humic substances, though lately it was reported that 10 to 20% of the humic substances in rivers and lakes can be 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). The bacterial regrowth potential can also be determined by 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.

Excessive growth of bacteria in the distribution network is not desirable because of the sludge this may produce. In addition, some of these bacteria are potentially pathogenic (e.g. *Legionella* and *Mycobacterium avium*). Bacterial growth in the network is mainly occurring in biofilms on the pipe surfaces. The biofilm is an environment where invertebrates can develop and where pathogens may be protected from the effects of disinfectants. To reduce these problems, the BOM levels in drinking water leaving the treatment plant should be as low as possible. Water in which bacteria cell are not multiplying is called biologically stable.

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. An AOC value of 10 $\mu\text{g L}^{-1}$ or less is recommended for a 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 μm 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 a corresponding increase in the levels of 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).

2.6.4 *Dissolved organic nitrogen (DON)*

Both low and high molecular weight molecules containing organic nitrogen (e.g., simple amino acids, algal-derived humic substances) have been observed to implicate as disinfection by-product precursors and membrane foulants. In spite of this, relatively minor attention has focused on dissolved organic nitrogen (DON) in water. Westerhoff et al (2006) studied the occurrence of DON in raw and finished drinking waters and provided information on its chemical characteristics and reactivity towards metal hydroxides and oxidants/disinfectants. They developed an accurate DON quantification method for drinking water, quantified DON removal by coagulation and activated carbon, and defined the role of DON in DBP formation.

2.6.5 *Interaction between inorganic colloids and NOM*

Organic matter has a great influence over the fate of inorganic colloids in water (Wilkinson et al 1997). The chemical nature and structure of NOM will be an important factor in determining whether colloids will be stabilised or destabilised by NOM. Fulvic acids are likely to be responsible for coating and imparting a negative charge to colloids. If the adsorbed organic coating produced and increase in absolute surface potential, it will act to stabilise colloids in the water column. On other hand, colloid organic carbon, especially chain-like structures can aggregate inorganic colloids through the formation of bridges. The importance of each process depends on nature and concentration of organic matter, as well as on other factors (e.g. origin of NOM, temperature, water treatment process).

2.7 **NOM properties and treatability by coagulation**

2.7.1 *Coagulation mechanisms*

In conventional turbidity removal processes, Pernitsky and Edzwald (2006) state that coagulation using metal based coagulants (Al, Fe) was considered as a destabilization process of colloidal particles, involving two primary coagulation mechanisms: 1) charge neutralization of the negatively charged colloidal particles by adsorption of positively charged coagulant species, and 2) enmeshment of colloids in precipitated $\text{Me}(\text{OH})_3$ solids, i.e. $\text{Al}(\text{OH})_3$, $\text{Fe}(\text{OH})_3$ (Amirtharajah and Mills 1982; Bache et al 1999; Lartiges et al 1997).

The presence of NOM affects greatly the chemistry of coagulation. Coagulants remove dissolved NOM by complexation reactions followed by a phase change, i.e. the NOM is removed from solution by forming a solid or adsorbing onto a solid. The primary NOM-coagulation mechanisms are: 1) complexation of NOM with dissolved metal coagulant species (Al or Fe), leading to direct precipitation of a Me-NOM solid phase, 2) complexation of NOM with dissolved coagulant species, leading to adsorption of this complexed material onto precipitated $\text{Me}(\text{OH})_3$ solids, and 3) direct

adsorption of NOM onto the surface of precipitated $\text{Me}(\text{OH})_3$ solids. In addition, NOM can coat inorganic particles, altering their coagulation behaviour (Wilkinson et al 1997).

The coagulation of waters containing both particles and NOM becomes more complicated, involving several important factors: 1) the dissolved coagulant species present upon coagulant addition, 2) the presence of precipitated metal hydroxide solids, 3) the concentration of particles and NOM, 4) the chemical properties of these contaminants and their reactivity with dissolved coagulant species, and 5) the pH of coagulation, which is affected by the chemistry of the coagulant and the alkalinity of the water. The presence of other charged constituents like calcium and sulphate have also been shown to affect coagulation under certain circumstances (Letterman and Vanderbrook 1983; O'Melia et al 2000). From this, Pernitsky and Edzwald (2006) proposed the conceptual view of the coagulation reactions shown in Figure 2.4.

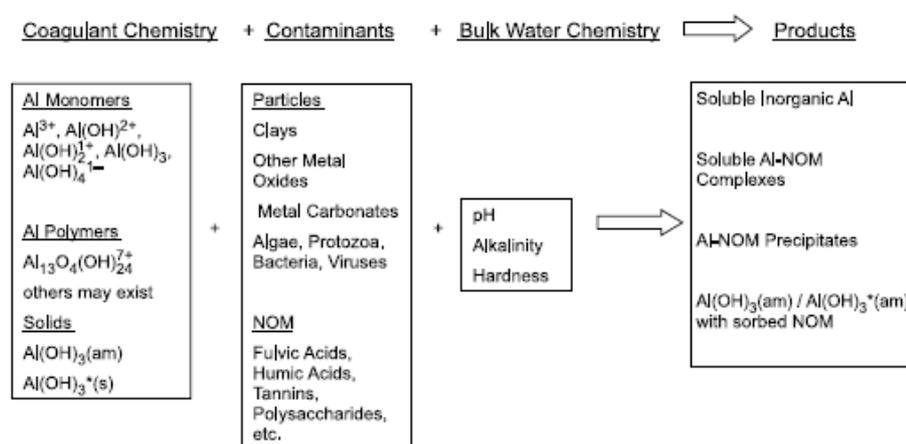


Figure 2.4 A conceptual view of coagulation reactions (Pernitsky and Edzwald, 2006).

2.7.2 SUVA, NOM-fractions and treatability

According to Edzwald and Tobiason (1999) SUVA values of 4 and higher indicate that NOM controls the coagulation process, and that good NOM removal can be expected (>50 %). In this case, NOM is dominated by high molecular weight, hydrophobic humic acid fractions.

For SUVA ratios in the range of 2-4, NOM is normally dominated by a mixture of hydrophobic and hydrophilic fractions of different molecular weights, humic and fulvic acids, as well as other NOM (algae and algae residues, etc). Here NOM influences coagulation, and fair to good DOC removals can be expected (25-50 %).

For raw waters with SUVA below 2, NOM is normally dominated by mostly non-humic, low molecular weight and substances with low hydrophobicity. NOM has little influence on coagulation performance, and poor DOC removal can be expected (< 25 %).

Table 2.1. SUVA as an indicator of raw water treatability by coagulation (Edzwald and Tobiason, 1999)

SUVA	Composition	Coagulation	DOC Removals
> 4	Mostly Aquatic Humics, High Hydrophobicity, High MW	NOM Controls, Good DOC Removals	> 50 % for Alum, Little Greater for Ferric
2-4	Mixture of Aquatic Humics and Other NOM, Mixture of Hydrophobic and Hydrophilic, Mixture of MWs	NOM Influences, DOC Removals Should be Fair to Good	25-50 % for Alum, Little Greater for Ferric
< 2	Mostly Non-Humics, Low Hydrophobicity, Low MW	NOM has Little Influence, Poor DOC Removals	< 25 % for alum, Little Greater for Ferric

From their IRC database analysis, Archer and Singer (2006) proposed an enhanced coagulation matrix with TOC removal requirements based on raw water SUVA (Table 2.2).

Table 2.2 Proposed enhanced coagulation matrix with TOC removals based on raw water TSUVA from IRC database analysis (Archer and Singer, 2006).

Raw water TSUVA (L/m · mg)	TOC removal (%)
> 1.0-2.0	35
> 2.0-3.0	40
> 3.0-4.0	40
> 4.0	55
TSUVA = UV-abs/TOC	

The different NOM fractions exhibit different properties in terms of treatability by coagulation. It has been reported that high molecular weight, hydrophobic NOM fractions can be removed efficiently while low molecular weight hydrophilic fractions are not removed. Furthermore, the different fractions exhibit different coagulant demand and show different chlorine and ozone reactivity and by product formation.

An illustrating example is presented in Figure 2.5, showing percentage removals of the different DOC-fractions VHA, SHA, CHA, and NEUT in two Australian raw waters by alum coagulation at pH 6 in jar tests.

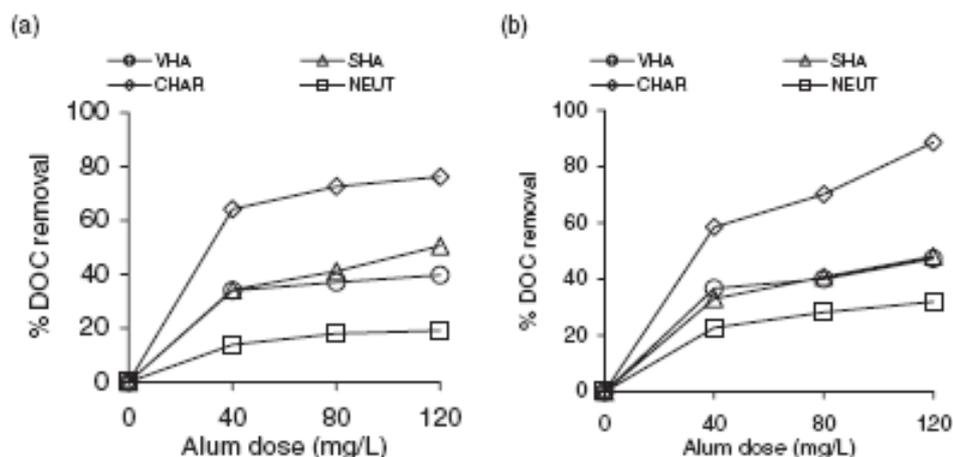


Figure 2.5 Percentage removals of DOC fractions by alum coagulation at pH 6 from a) Mt.Zero, and b) Moorabool raw waters (van Leeuwen et al 2002).

Sharp et al (2006, 2006b, 2006c) investigated how the NOM fractions influenced the coagulation performance. They found that the hydrophilic non-adsorbed fraction was least amenable to removal by conventional coagulation, attributed to a negligible charge density. The hydrophobic fulvic acid fraction most readily resembled the bulk raw water operational characteristics in relation to coagulation, such as floc size, strength, settling velocity, iso-electric point, generating small compact flocs and exerting the most influence on the charge balance of the system. The results demonstrate that simple fractionation (i.e. hydrophobic/hydrophilic balance of the water) could provide a fast and effective method for improved understanding of coagulation performance.

Jarvis et al (2006) studied NOM removal, i.e. the effects of ferric sulphate coagulation, NOM and polymer (polyDADMAC) on floc size, breakage, re-growth and settling. Iron precipitate (hydroxide) flocs formed in the absence of NOM were larger and had better settling characteristics than coagulant precipitate flocs formed in contact with NOM. Addition of polymer further improved floc size and settling rates. Polymer addition reduced the rate of floc degradation, but did not greatly improve floc re-growth after breakage, which was generally poor for all of the suspensions.

NOM composition and variability

NOM composition and properties, and thereby treatability, may vary with place (location) and time. Sharp et al 2004 fractionated raw water samples from Albert WTP by XAD resin adsorption into the hydrophobic (HPOA) and hydrophilic (HPIA) components. The HPOA fractions were then separated into the humic acid fraction (HAF) and the fulvic acid fraction (FAF). The non-adsorbed fraction was categorised as hydrophilic non acid (HPINA). Their results showed that the make up of the water varied throughout the year even during a period of relatively stable total DOC (Figure 2.6).

As the DOC increases the majority of the additional organic matter is considered likely to be hydrophobic (Malcolm, 1985). Sharp et al (2006, 2006b, 2006c) reported that during certain periods, particularly following initial periods of heavy rainfall, there is not only an increase in hydrophobic material as reported by Owen et al (1993); Ratnaweera et al (1999) but also an additional increase in the hydrophilic content of the water. This is in accordance with the findings by Scott et al (1998) who observed variations in the hydrophilic content of 20-80 % during a four-year study of water from a UK upland peat catchment system. The seasonal variations are often explained in relation to a microbially driven mechanism of DOC release (Scott et al 2001). The soil microbes are more active during the warmer and often drier periods in the summer/early autumn, but the result of this is not seen until the organic matter is transported from the watershed to the water source during the first rainfall.

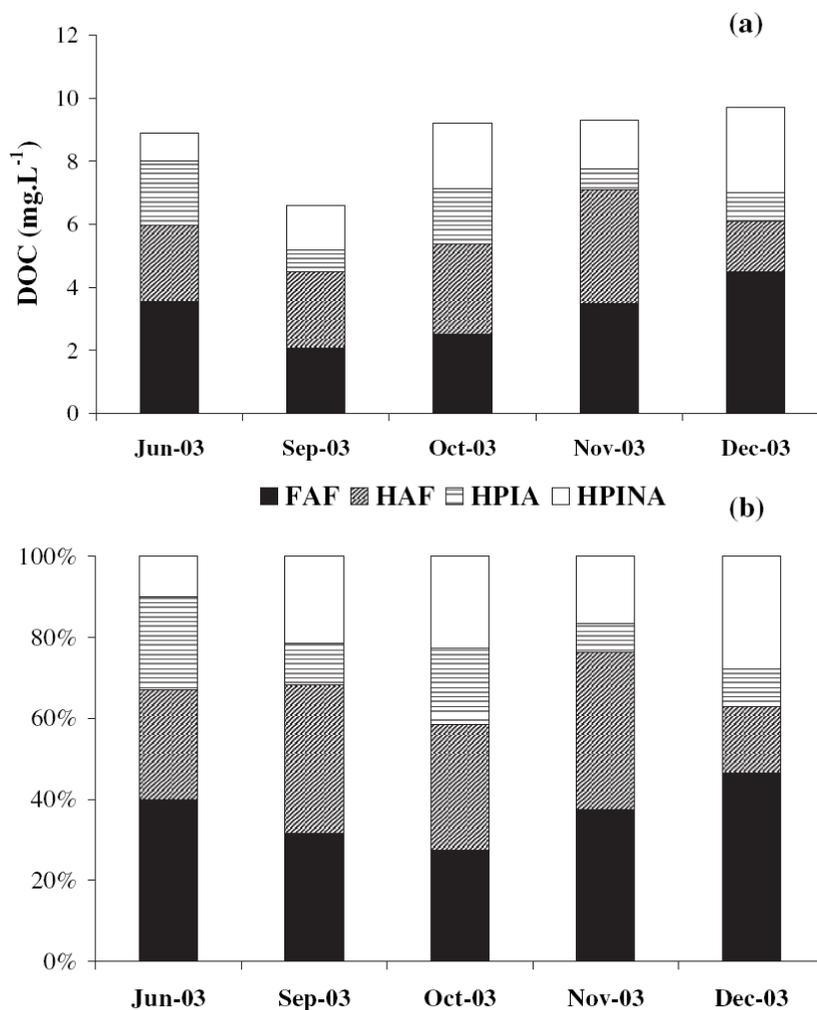


Figure 2.6 Seasonal variations in NOM fraction contribution to DOC in Albert WTP raw water, expressed as mg DOC/L (a) and percentage of raw water DOC (b) (Sharp et al 2004).

Liu et al (2006) studied the effects of pre-ozonation on organic matter removal by coagulation. With AlCl_3 , the coagulation efficiency was significantly deteriorated as a result of pre-ozonation (retardation of floc formation, decreased removal of turbidity, DOC, and UV_{254}). When PACl was used for coagulation, the adverse effects of pre-ozonation was mitigated, particularly when the specific ozone dosage was less than $0.69 \text{ mg O}_3/\text{mg TOC}$. From the differences between the UV_{254} and DOC removals, the results indicated that pre-ozonation changed the molecular structure of the organic matter. However, the mineralization capacity of ozone was not remarkable. Only 5 % or so of DOC was removed as a result of mineralization at $0.6\text{-}0.8 \text{ mg/L}$ of ozone alone. Fractionation results showed that the organic material was more low molecular weight and more hydrophilic after pre-ozonation, thus impairing the removal of DOC in the following coagulation.

Liu et al (2006 b, 2005) studied NOM removal by enhanced coagulation and polymer aided coagulation through jar tests, pilot and full-scale experiments in southern China. Both processes were able to remove UV-absorbing organic matter to 90 % or more. Enhanced coagulation was more effective with respect to removal of hydrophobic fractions, while the hydrophilic NOM fractions were better removed using polymer aided coagulation (polyacrylamide).

Colloidal charge and zeta potential

Sharp et al (2006c) reported that the majority of the total colloidal charge exists in the hydrophobic material, with values of 5.1 ± 1.3 , 3.6 ± 0.7 and 1.0 ± 0.6 meq/gDOC for the humic, the fulvic and the hydrophilic acid fractions, respectively. Humic and fulvic waters typically exhibit charge densities of 5-15 meq/gDOC (Edzwald, 1993; Tipping, 1993; Collins et al 1986). The charge densities of the hydrophilic fractions are generally very low (Edzwald, 1993).

Sharp et al (2006) studied seasonal variation in DOC. They calculated charge concentrations and showed that the majority of the charge load comes from the hydrophobic fractions (e.g. 0.0266 , 0.0263 and 0.0280 meq/L for April 2002, January 2004 and August 2004 samples, respectively). The charge density or carboxylic acidity of a fraction is likely to affect the coagulant demand. Higher charges are associated with the larger MW fractions. The charge load of each NOM-fraction shows seasonal variation, and it was concluded that coagulant demand cannot be calculated based solely on bulk parameters such as DOC in a specific raw water. The zeta potential values at a pH of greater than 4 were -13 ± 3.7 ; -17 ± 0.3 , and -25 ± 2.2 mV for the fulvic acid fraction, the raw water, and the humic acid fraction, respectively. In contrast, the zeta potential for the hydrophilic fractions was substantially lower and decreased with pH.

Ratnaweera et al (1999) performed jar tests to study the coagulation behaviour of eight natural Norwegian water samples containing NOM. They found that the optimum coagulant dose and the colloidal charge were both proportional to the initial colour, and that DOC and UV-absorbance was well

correlated with colour. Zeta potentials at optimum coagulation conditions deviated from zero, indicating that other coagulation mechanisms than charge neutralization are relevant. From this, they considered it difficult to use zeta potential as the only tool for online dosing control.

Sharp et al (2006c) performed bench scale and pilot scale coagulation experiments on three source waters: two from UK moorland catchments and one from a US snow melt source. From this they presented an overview and a discussion of the links between NOM character and treatability by coagulation. Their work demonstrated the importance of the polarity balance and the charge density of the NOM. NOM composition can vary both temporarily and spatially, with increased DOC concentrations associated with both an increase in hydrophobic content and charge density. The hydrophobic content controls the coagulant demand such that variation in the demand between sources or sampling periods can be accounted for by changes in the hydrophobic content and charge density of the NOM. The raw water hydrophilic content, and specifically the non-acid fraction, provides an indicator of the achievable residual. A clear relationship was revealed between zeta potential and residual DOC. For each source an optimum operational zeta potential exists, in the range between - 10 and +3 mV for the investigated raw waters, thus providing a useful guide range for operational control of enhanced coagulation processes.

Henderson et al (2006) investigated surface characteristics and floc properties of three common systems: NOM, algae and clay. They demonstrated that difficulties arise when coagulation is not optimized for the dominant substance/particle. Charge density and specific surface area were important parameters with respect to coagulant demand for charge neutralization of all three systems. Extra-cellular organic matter (EOM) affected the coagulant demand of algae to such an extent that it could dominate the coagulation process. Algal flocs were much weaker and required five times the flocculation period to reach steady-state floc size compared to NOM and clay flocs. Despite similarities between algae and NOM in terms of organic content and coagulant demand, the fact that algae is a dynamic, biological system creates numerous problems for the coagulation/flocculation processes.

Pommerenk and Schafran (2005) investigated the removal of sulphate, orthophosphate, fluoride, five organic acids (including humic acid) and NOM from two sources by adsorption on aluminium hydroxide precipitates. Phosphate was nearly completely removed from solution across a wide range of pH, and was observed to lower the surface charge and shift the isoelectric point of the hydroxide precipitate. Sulfate was removed to a lesser extent than phosphate, lowered the surface charge on the precipitate, but did not shift the isoelectric point. Fluoride was well-removed through adsorption to the precipitate, but did not influence the charge of the aluminium hydroxide. The organic acids displayed varying abilities to be removed through adsorption and different influences on surface charge. The results indicate the importance of the number and location of functional groups and their acid/base properties. The ability to strongly influence surface charge

illustrates the impact that adsorption of these anions can have on particle stability.

Coagulation, pH and iso- electric point (IEP)

Qin et al (2006) studied NOM removal by enhanced coagulation at a water works in Singapore. From jar tests using different coagulation pHs and alum dosages the optimum conditions were observed at a pH of 5.2 and a alum dose of 5 mg Al/L). Under optimal conditions, turbidity and DOC removals of 97% and 45% were obtained, respectively. The DOC removal obtained with conventional coagulation at pH 7.2 was only 35%. It was concluded that control of coagulation pH was critical for NOM removal in treatment of reservoir water.

Sharp et al (2006) reported that the IEP (the pH associated with zero charge) of the NOM fractions were 1.5, 1.6, 1.7, 4 and 4.7 for the humic acid (HAF), fulvic acid (FAF), raw water, hydrophilic acid (HPIA) and the hydrophilic non-adsorbed (HPNIA) fractions, respectively. The addition of coagulants shifted the IEP of the system, indicating interactions between the coagulant and the NOM fractions.

The interactions between the individual NOM fractions and coagulant (Me+) were monitored through changes in IEP of the sample, at various DOC to coagulant mass ratios (meq DOC : meq Al or Fe). The results show that the hydrophobic fractions produced the largest change in IEP (i.e. a small change in charge ratio imposed a large change in IEP). On this background, Sharp et al (2006) concluded that it is the hydrophobic fractions which appear to be critical in determining the resultant charge properties of the coagulant-NOM system and hence the subsequent coagulant dose requirements. At the coagulation pH of normal operation (pH 5-7), the hydrophilic fractions possess a negligible or slightly positive charge, leading to poor removal and values as low as 16 % were reported by Fearing et al (2004).

Jarvis et al (2005) studied how the NOM floc structure was affected by increased organic fraction in the floc. It was observed that when the organic floc fraction went significantly over a mass ratio of 1 mg DOC to 1 mg of Fe (coagulant), the floc size, settling velocity, fractal dimension, and strength were seen to decrease even when the NOM removal during coagulation remained high. These effects were proposed to result from adsorption of NOM on primary particle surfaces (i.e. coagulant precipitates). The operational significance of these results suggests that the correct coagulant dose must be applied to give good floc structure.

Sharp et al (2006b) stated that a number of UK and US water utilities have been experiencing operational difficulties associated with increased DOC levels during the autumn and winter periods, resulting in increased coagulant demand and increased production of DBPs. During a 3-year study period a seasonal change in NOM composition was observed, with the hydrophobic, fulvic acid fraction increasing from 36% in September to 61% in November. A

reduction in treatment performance was not simply due to an increased organic carbon concentration (from 4.3 to 14.5 mg DOC/L), but is also to a change in the charge density of the NOM. Hydrophilic NOM fractions were found to possess a negligible charge density (< 0.06 meq/g DOC), they were less amenable to removal by coagulation, and were therefore likely to indicate the DOC residual remaining after treatment. On the other hand, it was the hydrophobic NOM fractions, the fulvic acid fraction in particular, that exert the greater dominance on coagulation control. Understanding the seasonal changes in NOM composition and character and their reactivity with the coagulants should lead to a better optimisation of the coagulation process and a more consistent treated water quality.

Above we have described the pH effects on the interactions of NOM with surfaces of coagulant flocs, however, pH may also have indirect effects on removal of NOM, especially, on removal of humic substances. HS are macromolecules which behave like polyelectrolytes; thus their sorption is determined not only by availability of sites, but also by lateral interactions between themselves and the steric arrangements of the macromolecule. When the pH of water decreases HS acquire a coiled configuration (due to lower intermolecular electrostatic repulsion) in solution, thus they may be more effectively adsorbed on surfaces by hydrophobic interactions (Juhna et al 2003).

With the current trends forecasting an increase in DOC concentrations, an increased understanding of the treatment system and the involved mechanisms is vital, especially as the potential consequences of increasing NOM concentration levels are likely to impact dramatically on the water industry.

NOM characterization, e.g. fractionation, charge measurements, etc. during the coagulation process and comparison across different source waters, seasons and coagulants could be used in a proactive strategy in achieving a more robust and cost-effective water treatment plant operation in general, and during problem periods in specific.

3 Plant operation practices

3.1 Enhanced coagulation process design and major treatment objectives

Coagulation and filtration is widely used in water treatment because the process is very effective for controlling a wide range of contaminants. These includes particulate matter that causes turbidity; natural organic matter (NOM) that causes colour and UV-light absorption; algae; inorganic substances like iron, manganese and arsenic; bacteria, virus and protozoa (e.g. *Cryptosporidium* and *Giardia*). The USEPA indicates that enhanced coagulation is the best available technology to control DOC in drinking water treatment plants.

Jacangelo et al (1995) discussed the increased emphasis by the water industry on the removal of NOM as a result of new regulations. Among NOM removal options enhanced coagulation is the most widely used in the water industry. But when coagulation is unable to remove adequate concentrations of NOM, other treatment technologies such as GAC filtration and membrane filtration may need to be used.

The capability of a water treatment plant employing coagulation and filtration is of course dependent of the nature, design and operation of the coagulation and filtration steps themselves, but it is also highly influenced by the nature and extent of pre-treatment. Conventional filtration plants, i.e. plants with pre-sedimentation can cope with raw waters with high concentrations of turbidity (>1000-2000 NTU), NOM, and oxidized inorganic matter. Direct filtration plants, in which sedimentation is omitted, can effectively treat "ideal" raw waters with turbidity of 10-20 NTU, and colour up to 40 mg Pt/l (AWWA, 1980). Dissolved air flotation (DAF) is known to be very effective in removing algae. Algae generally do not settle well, and can clog filter beds.

The enhanced coagulation-filtration processes normally consists of a mixer (e.g. a flash mixer, or in-line static mixer), a flocculation step (omitted in contact filtration facilities), a separation step (sedimentation or flotation, omitted in direct and contact filtration facilities), and a filtration step. In addition, there are dosage facilities for coagulants, acid or base for pH control depending on raw water alkalinity, and polymeric flocculants and/or filtration aids. Corrosion control (i.e. increase in pH, alkalinity and calcium) is normally integrated in the process/the pH control system.

Pre-treatments may include microscreening, pre-ozonation, the addition of other types of oxidants, silica, powder activated carbon (PAC), etc. Post-treatments may include granular activated carbon (GAC) filtration for taste and odour removal, disinfection, post-corrosion control, etc. In addition, conventional deep bed filters can be replaced by membrane filters, e.g. NF, UF or MF with pre coagulation, etc).

Figure 3.1 shows an example of a treatment process including enhanced coagulation, pH- and corrosion control, dissolved air flotation (DAF), sand filtration, granular activated carbon (GAC) filters, and chlorine disinfection.

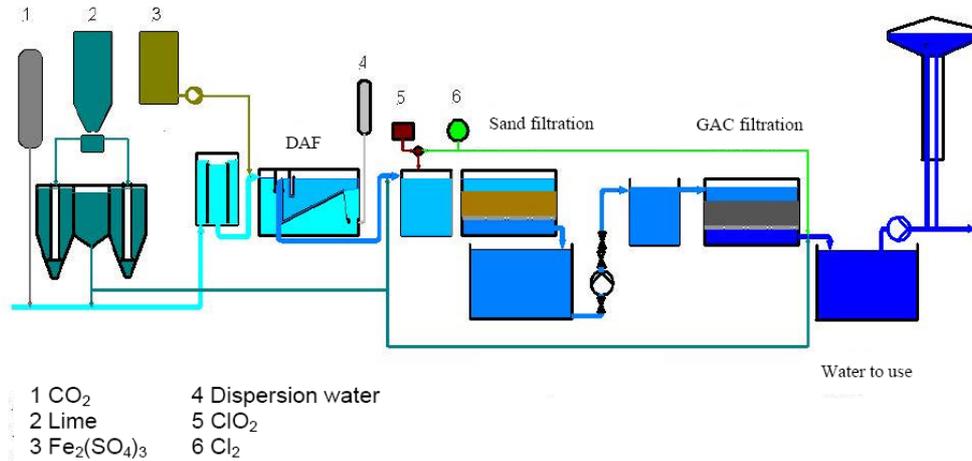


Figure 3.1. Treatment process scheme of the Rusko WPT (Jokela, et al 2006)

A less complex enhanced coagulation - contact filtration process scheme with integrated corrosion control (lime and CO₂), and post-disinfection by chlorination is presented in Figure 3.2. This process is widely used in Norway for treating soft and NOM-laden raw waters with low turbidity and low alkalinity.

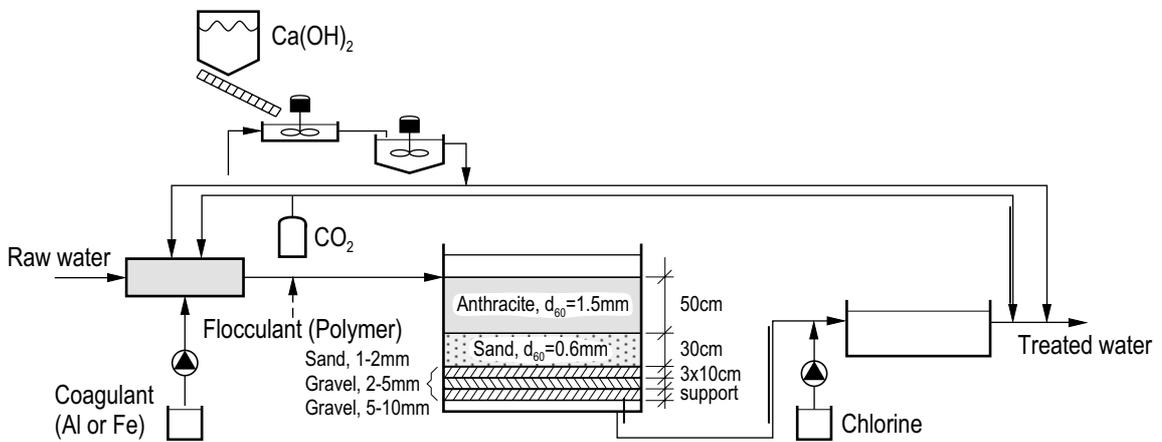


Figure 3.2. An enhanced coagulation-contact filtration process scheme, with dual media anthracite-sand filtration, lime/CO₂ for pH and corrosion control, and disinfection by chlorination.

Figure 3.3 illustrates an alternative filter bed design where an alkaline filter layer (CaCO₃) is applied as a third medium below the sand layer, a layer also capable of collecting possible metal coagulant residuals from the anthracite-sand layers, thus ideally allowing a reduction in coagulant dosage. This

design is made for integrated NOM and corrosion control purposes. The alkaline filter layer could also be designed a separate post filter unit.

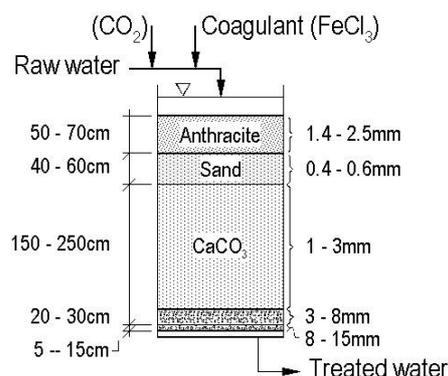


Figure 3.3 Illustration of a 3-M anthracite-sand-CaCO₃ filter design for integrated NOM and corrosion control.

Conventional filtration, direct filtration and contact (in-line) filtration concepts have all proven effective for controlling NOM as well as protozoa in water, provided optimum or near-optimum coagulation and filtration conditions and management.

In the last few years, membrane filters are also used in combination with coagulation, e.g. coagulation followed by ultra filtration. In this application, membrane fouling control is a key operation challenge, and it is very important also in this respect to find the optimum coagulation conditions. Optimum enhanced coagulation conditions seem the favourable conditions also in this respect.

3.2 Operation of enhanced coagulation-filtration facilities

Among the major operational challenges in enhanced coagulation facilities are:

- Selection of a proper type of coagulant, correct dosage and pH levels;
- Coagulation conditions that are well adapted to variations in raw water quality;
- Selection of filter bed that is well adapted to the water to be filtered;
- Establishing a monitoring system that controls filter effluent turbidity from each filter unit, including head loss development and head loss distribution within the filter bed;
- Establishing of a hydraulic control system that avoids peaks in filtrations rates as a result of backwashing of parallel filter units, return flows from sludge treatment, small buffer volumes/clean water tank, etc.;
- Proper filter backwash techniques, and filter bed inspection routines that are able to detect potential problems related to insufficient cleaning (mud ball formation, biofilms, inorganic precipitates, trends

towards shortened filter run times, increasing initial head loss and increasing rates of head loss development, etc.).

3.2.1 *Optimum coagulation conditions (coagulant type, coagulant dosage and pH)*

Deviations from optimum coagulation conditions (i.e. coagulant dose and pH) can seriously affect treatment performance with respect to residual coagulant concentrations, turbidity, particle counts, NOM, and micro organisms. A proper control of coagulant dosage and coagulation pH are the most important operation challenges. Rapid and large variation in raw water quality, with optimal adjustment of the coagulation conditions to the actual raw water quality (e.g. DOC, colour, etc.), represents major operational challenges.

Edzwald and Tobiason (1999) summarized principles for alum coagulation of NOM as follows: negatively charged NOM creates a coagulant demand for positively charged Al species resulting in a stoichiometric relationship between the alum dosage and the raw water DOC that is pH dependent. They addressed coagulation in a broader view than enhanced coagulation, termed multiple objectives coagulation. This includes maximization of particle, turbidity, TOC, and DBP precursor removal, and minimization of residual coagulant, sludge production and operating costs. Based on full-scale plant data they also demonstrated a dual coagulation strategy of alum and cationic polymer that reduces sludge production and overall operating costs compared to alum alone.

Edzwald (1993) presented a review on coagulation in water treatment, emphasizing the importance of raw water chemistry, NOM concentration and type, and the chemistry of coagulants. NOM rather than particles in water supplies can control coagulant dosage and selection. The removal of NOM with Al coagulants can involve hydrolysis, complexation, precipitation, and adsorption reactions. SUVA can be used to estimate whether the NOM is high or low in hydrophobic acids and to estimate removals of DOC by coagulation.

O'Melia et al (1999) stated that adsorption of NOM on oxides depends significantly on complex formation reactions between specific sites on oxide surfaces and functional groups on the NOM. Coagulation requirements can and often are set by the TOC concentration in a water source. Frequently there is a stoichiometric relationship between the required coagulant dosage and the TOC of the water to be treated. Other important factors include pH and the concentration of divalent cations.

Gray (1991) performed laboratory and pilot scale direct filtration experiments to study the influence of particles and NOM on filtration performance. He concluded that raw water NOM determined the coagulant dose and filtration performance to a far greater extent than turbidity. Below a certain particle concentration limit filter ripening may be extremely slow, and even low turbidity waters with moderate NOM concentrations require relatively high coagulant doses.

Eikebrokk (1996, 1999, 2001) demonstrated in contact filtration pilot experiments with aluminium and iron based coagulants that compliance with the total residual coagulant concentration standard of 0.1 mg Me/L was the determining factor with respect to identifying minimum coagulant dosage requirements when treating low turbidity surface waters with NOM-concentrations measured as colour and TOC in the range of 15-50 mg Pt/L and 2-6 mg TOC/L, respectively (Table 3.1). Compliance with the turbidity (<0.2 NTU) and colour (<5 mgPt/L) could be obtained with lower coagulant doses. He distinguished between absolute and practical minimum doses; the difference being that for the absolute minimum dose level the maximum residual metal concentration of 0.15 mg Me/L can be obtained only within a very narrow pH-window. When using the 25% higher practical minimum dosage, the pH window for optimum process performance and compliance with the water quality standards can be obtained within a wider range of pH.

Table 3.1. Minimum practical coagulant dose recommendations when using ALG, JKL or Chi in contact filtration of raw waters with colour levels of 15, 30 and 50 mg Pt/L. (After Eikebrokk, 2001).

Raw water colour	Raw water SUVA L/mgC-m	Minimum specific coagulant dosage mmol Me or Chi/gTOC; (µgMe or Chi/mgPt)			
		ALG pH 5.8-6.6	JKL pH 4.0-5.5	PAX pH 5.7-6.7	CHI pH 5.0-6.0
RW15	3.8	16 (78)	16 (162)	14 (67)	0.6 (110)
RW30	4.3	20 (63)	20 (128)	17 (54)	0.7 (80)
RW50	4.8	26 (61)	26 (100)	20 (49)	0.8 (70)

ALG-aluminium sulphate, JKL-ferric chloride; PAX-poly aluminium chloride; Chi-Chitosan
 For Me-coagulants: Dose levels needed to obtain < 0.1 mg residual Me/L, > 90% and 50-60% colour and TOC reduction, resp. Absolute minimum doses are 25% lower than the given practical minimum doses.
 For Chitosan: Dose levels needed to obtain > 60 % and 20-35% colour and TOC reduction, resp.

Given the atomic masses of 26.98 for Al and 55.85 for Fe, the dose levels of 16-26 mmol per gram of organic carbon shown in Table 1 represent 0.43-0.70 mg Al per mg C, and 0.89-1.45 mg Fe per mg C, respectively.

The optimum range of pH (measured on-line in filter effluent water) for the different coagulants were as follows:

- ALG pH 5.8-6.6
- JKL pH 4.0-5.5
- Chi pH 5.0-6.5

For chitosan it should be noted that an increase in colour and TOC removal efficiency was observed with decreasing pH in some cases. Coagulation at a pH below 4-5 may however interfere with turbidity removal and is not likely to be cost-effective.

The great importance of controlling the coagulation pH within strict limits was clearly demonstrated. The width of the optimum "pH-window" increased with increasing coagulant dose level, indicating the relationship between coagulant dosage and ease of operation from the operator's point of view.

As an illustrating example, raw water with a specific UV-absorption (SUVA) of 4.8 and a TOC of 5 mg/L would require a minimum dose of 3.5 mg Al/L and 7.25 mg Fe/L to cope with the residual metal standard of 0.1 mg Me/L. The correspondent reductions in colour and TOC would be in the range of 90 and 50-60 %, respectively. For the organic coagulant chitosan (derived from shrimp and crab shells) a dose of 4.5 mg/l would be required to obtain colour and TOC reductions in the range of 60 and 20-35 %, respectively.

The specific coagulant dose requirements presented above agree well with data presented by Pernitsky and Edzwald (2006) who tested coagulation requirements for raw waters with different NOM, turbidity and alkalinity levels. For all waters examined, they found that 0.5-0.8 mg Al per mg of TOC in the raw water was required, which is in accordance with stoichiometric estimates presented by Van Benschoten and Edzwald (1990) and Edzwald (1993).

The dosage requirements also agree well with data from the Information Collection Rule (ICR) database, the largest nationwide water quality survey ever conducted in the U.S.A. Archer and Singer (2006) presented data on enhanced coagulation performance for water treatment facilities. They examined the relationship between SUVA and the amenability of water to coagulation for NOM removal, and demonstrated that the removal of TOC, UV-absorbing organics, and DBP precursors was a function of the raw water SUVA value.

The applicability of SUVA as an indicator of NOM aromaticity and treatability by coagulation was discussed above. In a study by Archer and Singer (2006) the relationship between SUVA and the susceptibility of water to enhanced coagulation was evaluated using the information collection rule database. A total of 269 surface water treatment plants of the 500 ICR plants were included in the study. Records with TOC concentrations less than 2 mg/l were omitted because enhanced coagulation is not required for such waters. The resulting data set contained 2334 enhanced coagulation matrix entries. The raw water quality data on alkalinity, TOC, UV-absorption and hardness were divided in 9 elements (a 3x3 matrix with 3 alkalinity levels and 3 TOC levels). Influent and settled water records were used to observe the decrease in TOC, UV_{254} and TSUVA (i.e. UV_{254}/TOC) after coagulation and settling.

In a second analysis treatment plants that used any kind of oxidative pre-treatment (e.g. pre-chlorination) were omitted because of the potential of such pre-treatment to alter the characteristics of NOM and its response to

coagulation. The resulting data set consisted of 42 treatment plants with a total of 592 sample observations.

As expected, the results showed that the reduction in UV absorbance exceeded the corresponding reduction in TOC, in line with several other studies showing preferential removal of UV absorbance over TOC due to the hydrophobic nature of UV-absorbing substances and the efficient removal of such hydrophobic fractions obtained by coagulation. TOC removal and TSUVA increased with increasing raw water TOC concentration regardless of raw water alkalinity level (Figure 3.4).

These results indicate that the aromatic content of the organic carbon increases with increasing raw water TOC, and that natural waters with high TOC concentrations contain mostly aromatic humic substances with a high UV absorbance amenable to removal by coagulation. Conversely, raw waters that are low in TOC are likely to be dominated by hydrophilic NOM which is less easily removed by coagulation.

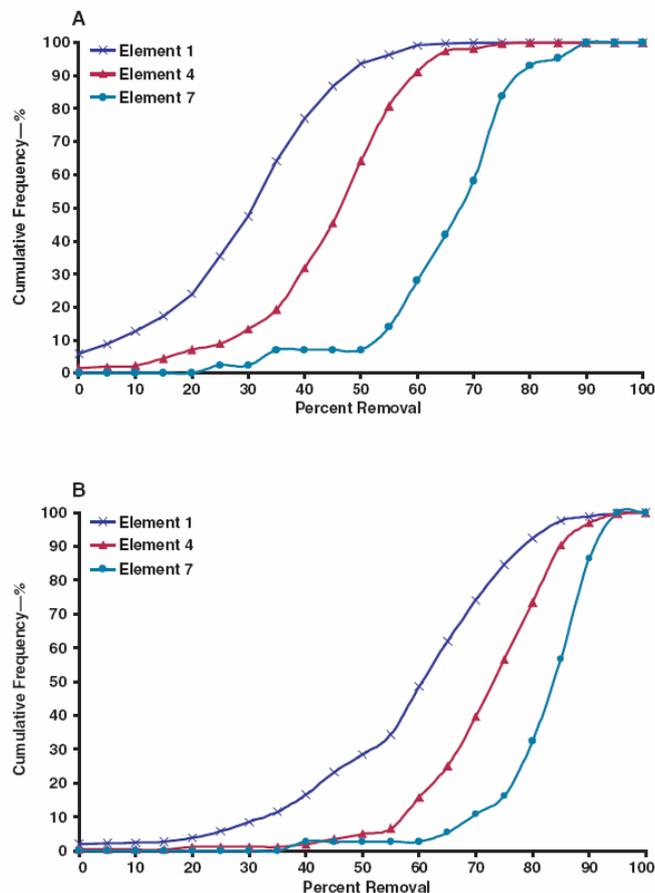


Figure 3.4 Percent removal of TOC (A) and UV-absorbance (B) as a result of conventional coagulation and clarification for raw waters with low alkalinity (0-60 mg CaCO_s/L), and different TOC levels, i.e. Elements 1, 4 and 7 represents raw water TOC levels of 2-4; 4-8; and > 8 mg TOC/L, respectively. The results are based on ICR data base analysis (Archer and Singer, 2006).

Furthermore, TOC removal increased with decreasing raw water alkalinity, regardless of raw water TOC concentration level (Figure 3.5). These results suggest that coagulation of NOM in high alkalinity waters is less effective because of the buffering capacity which prohibits many utilities from reaching the optimum pH of 5.5-6.5 for alum coagulation.

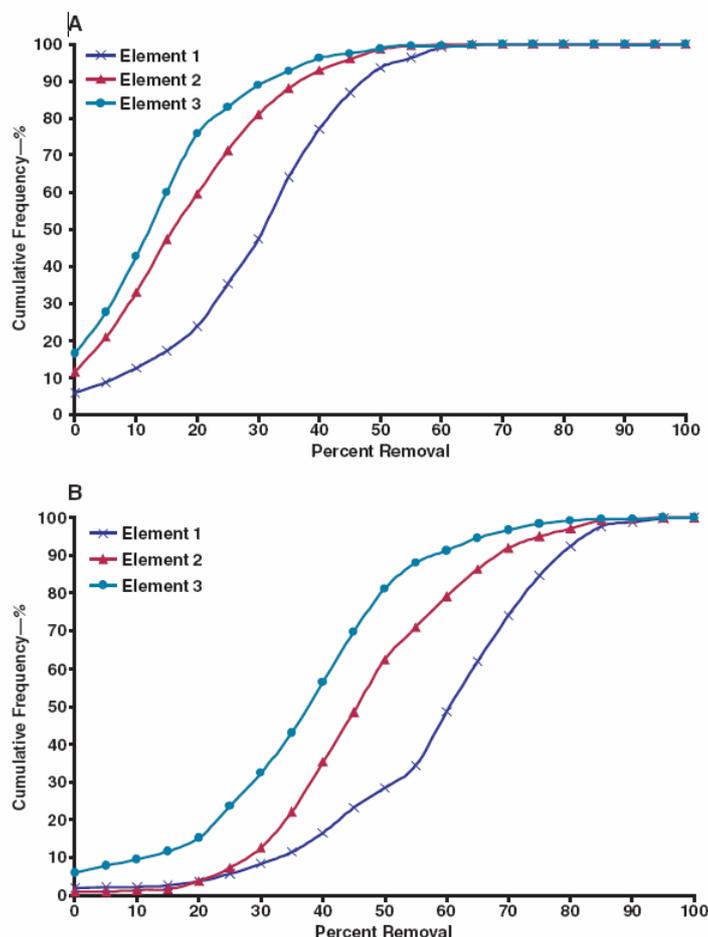


Figure 3.5 Percent removal of TOC (A) and UV-absorbance (B) as a result of conventional coagulation and clarification for raw waters with 2-4 mg TOC/L, and different alkalinity levels, i.e. Elements 1, 2 and 3 represents raw water alkalinities of 0-60; 60-120; and > 120 mg CaCO₃/L, respectively. The results are based on ICR data base analysis (Archer and Singer, 2006).

In addition, the results show that TSUVA tends to decrease with increasing raw water alkalinity, suggesting that hydrophobic NOM (i.e. humic substances) may be unstable in high alkalinity waters, and possibly tend to form complexes with calcium and magnesium and potentially precipitate due to the corresponding high hardness concentration. Andersen et al (2000) found that large molecular weight NOM was removed as a result of liming of water reservoirs. The above observations support the capability of SUVA to

serve as an indicator of aromaticity of organic carbon, and of treatability of NOM by coagulation.

TSUVA values decreased as a result of coagulation, and the low alkalinity raw waters experienced the largest reductions. These results indicate that coagulation preferentially removes UV-absorbing, aromatic organic carbon, and imply that raw waters with high alkalinity and hardness, and corresponding low TSUVA values, are comprised primarily of hydrophilic organic carbon that is less susceptible to coagulation.

Figure 3.6 shows a frequency distribution plot for the applied specific alum coagulant dosage, i.e. mg Al applied per mg TOC in raw water. The mean value for the 1571 records is 0.7 mg Al/mg TOC. Some of the variability of the specific coagulant dosage is related to high alkalinity raw waters. The results in Figure 3.6 agree well with the results presented above (e.g. Table 3.1).

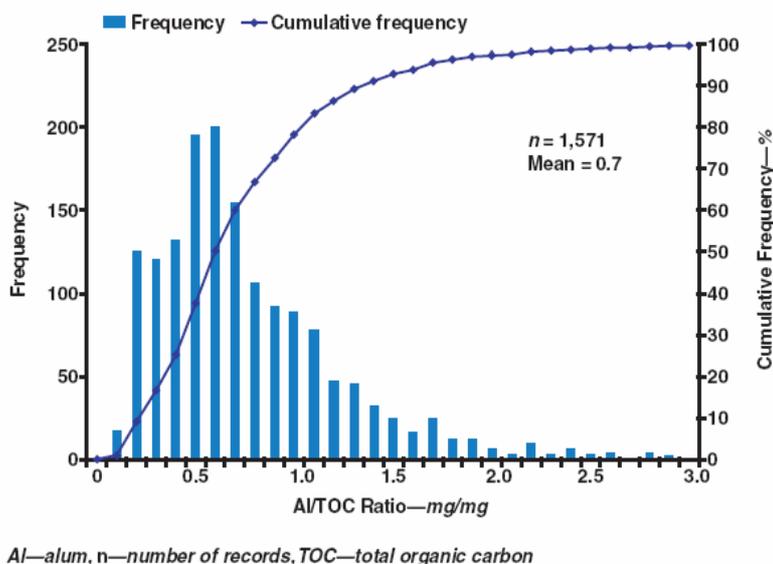


Figure 3.6. Frequency distribution plot for specific coagulant dosage (mg Al/mg TOC) based on ICR database (Archer and Singer, 2006).

From their ICR database analysis, Archer and Singer (2006) also presented the obtained reductions in TOC and UV absorption classified according to the TSUVA (i.e. TOC/UV-adsorption) level of the raw waters (Figure 3.7). The results illustrate well the relationship between TSUVA and the removal of TOC and UV absorbance.

Mesdaghinia et al (2006) studied enhanced coagulation effectiveness in removal of disinfection by-products. From jar test experiments on Iranian river water they concluded that NOM removal was a function of coagulant type, coagulant dose, and pH of coagulation. Ferric chloride was consistently more effective than alum in removing NOM. Coagulation pH appeared to be

a determining factor for maximum NOM removal, and the removal of DBP precursors was significantly enhanced at pH 5.5 in comparison with the initial pH of the water. Pre-adjustment of pH with sulphuric acid reduced the required coagulant dosage and thus the production of sludge.

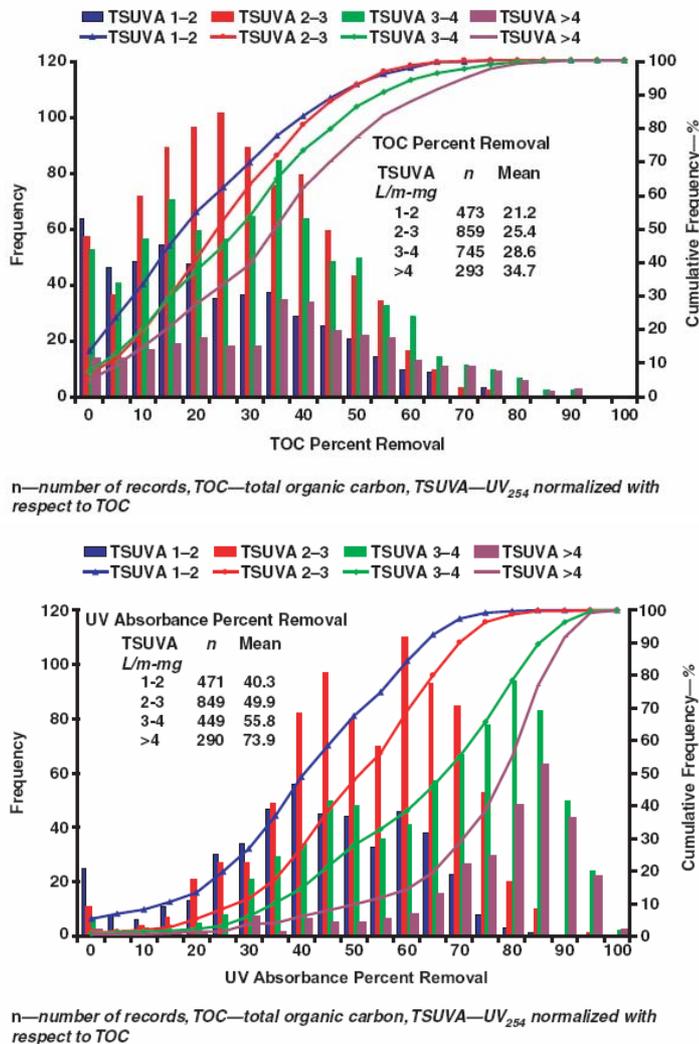


Figure 3.7 Frequency distribution plots of TOC removal (upper) and UV absorbance removal (lower) categorized by raw water TSUVA from ICR database records (Archer and Singer, 2006).

Pernitsky and Edzwald (2006) presented guidelines for the selection and use of poly aluminium chloride (PACl) and alum in terms of raw water quality and treatment method. NOM was identified as the most important parameter affecting coagulant dose. The nature of NOM, as measured by SUVA, was useful for predicting the degree of NOM removal expected. Raw water turbidity and NOM did not influence the type of coagulant that was most effective. As it relates to pH of coagulation, raw water alkalinity is important for choosing one coagulant type over another. The basicity of the PACl should be matched to raw water alkalinity, so that coagulation pH is as close as possible to the pH of minimum solubility of the coagulant. PACls

containing sulphate were found to have the best settling characteristics, but showed the highest head loss rates in direct filtration applications. Dissolved air flotation performance was relatively insensitive to coagulant type.

Jiang and Graham (1996) studied the consequences of enhanced coagulation, i.e. the use of excess coagulant and lowering the coagulation pH in order to improve NOM removal, including increased sludge production and increased treatment costs, and the need for chemical storage and feed facilities to be changed. They stated that the use of various types of pre-polymerized coagulants that have been developed in recent years can improve NOM and trihalomethane formation potential (THMFP) removals at relatively lower dosages, and thereby reduce the operational and economic consequences of enhanced coagulation.

Gao and Yue (2005) studied turbidity and NOM removal by poly-aluminium-chloride sulphate (PACS) coagulation. The coagulant was synthesized using $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ and Na_2CO_3 as raw materials. They found that the coagulation performance of PACS in water treatment was affected by the PACS particle size distribution and zeta potential which were both affected by the basicity ($[\text{OH}^-]/[\text{Al}^{3+}]$) and the sulphate to aluminium molar ratio ($\text{SO}_4^{2-}/\text{Al}^{3+}$) of the coagulant. They concluded that PACS achieved an optimum NOM and turbidity removal when the coagulant was prepared with a basicity of 2.0 and a sulphate to aluminium molar ratio of 0.0664. Gao and Yue (2005b) studied NOM coagulation performance of PACS with this composition and compared it with PACl, FeCl_3 and alum. They found optimum coagulant dose and pH values of 5.0 mg Al/L and pH 5.0-8.2 for PACS and PACl, 7.0 mg Fe/L and pH 5.0-6.0 for ferric, and 7.0 mg Al/L and pH 5.0-7.0 for alum. At optimum conditions, the selected PACS achieved the best NOM removal, followed by PACl, ferric, and then alum. Under the optimum coagulant conditions with PACS and PACl, the residual aluminium concentration in treated water was 115 $\mu\text{g/L}$.

Matilainen et al (2005) compared the efficiency of aluminium and ferric sulphate coagulants for NOM removal during coagulation/flotation of drinking water in Finland. Approximately 95% of high molar mass organic substances (HPSEC) were removed with both coagulants. The greatest difference between the coagulants occurred in the removal of organic compounds having molar masses of 1000-4000 g/mol, which were removed 25% more efficiently with the iron-based than the aluminium-based coagulant. Low molar mass material was poorly removed regardless of the coagulant (10%). In terms of overall NOM removal, iron was 10% more efficient than aluminium. However, turbidity removal during coagulation-flotation was more efficient with aluminium, especially during the winter period. Turbidity was effectively removed during filtration.

Matilainen et al (2002) studied the removal of NOM in the different stages of the water treatment process at Rusko WTP in Tampere, Finland. The treatment process steps include coagulation, flocculation, clarification by sedimentation or flotation, activated carbon (GAC) filtration and disinfection.

They used high-performance size exclusion chromatography (HPSEC) to determine changes in NOM character (molecular size) during the treatment steps, and measured water quality in terms of TOC, KMnO_4 -number and UV-absorbance. Significant correlation was established among these parameters. The results showed that high MW organic matter was clearly easier to remove in coagulation and clarification than low MW matter. Depending on the regeneration status, the GAC filters were able to reduce the amount of low MW organics to some degree, but were not able to remove most of these substances.

Fearing et al (2004) reported that over 70% removal of hydrophobic and hydrophilic acid fractions, while only 16% of the hydrophilic non-acid fraction was obtained using conventional coagulation treatment during elevated NOM loadings at a treatment facility. From jar tests on isolated NOM fractions, it was concluded that increased removal of the hydrophilic fractions could be obtained when conditions were optimized. From this, an optimized two-stage coagulation process was proposed to increase the removal of recalcitrant fractions of NOM.

To gain more insight into the types of NOM that are recalcitrant to removal by coagulation, van Leeuwen et al (2002) studied alum coagulation of two Australian water sources. They used a simplified fractionation technique (Chapter 2) to isolate four DOC fractions: VHA, SHA, CHA, and NEUT. The isolated fractions were then treated by alum coagulation. The fractions were characterized by a number of different methods: high performance size exclusion chromatography (HPSEC), bacterial regrowth potential (BRP), trihalomethane formation potential (THMFP), pyrolysis gas-chromatography mass spectrometry (Py-GC-MS), and thermochemolysis. The highest removal of DOC by alum coagulation was obtained in samples spiked with the CHA fractions while the NEUT fractions were the most recalcitrant. The CHA fraction contained the highest MW organic matter, and the NEUT fraction contained the lowest MW material. The NEUT fractions had the highest relative proportion of saccharide derived organic material, and showed considerable variety in BRP, and thereby in the ability to support microbial growth.

Wang et al (2002) studied the effectiveness of enhanced alum coagulation for removal of NOM at various alum dosages and pH conditions for three source waters in Taiwan. Jar tests were performed with alum dosages ranging from 60-120 mg/L of alum, and pH values from 5.0 to 8.0. DOC removals of up to 50 % were achieved, depending on raw water DOC and alkalinity levels.

Freese et al (2001) performed laboratory and pilot scale tests to compare the effectiveness of enhanced coagulation with ozonation and granular activated carbon adsorption in treating various types of raw water in South Africa. Reductions of up to 50% in trihalomethane formation potential and 50-70% in organic carbon and colour were obtained using enhanced coagulation, which compared favourably with ozonation and GAC filtration. The latter process was especially effective in the removal of micro pollutants, generally being in

excess of 70%. Inorganic coagulants were found to be more effective than polymeric coagulants for organic matter removal, and the addition of inorganic acids to depress pH allowed for increased organics removal at lower coagulant doses.

Bell-Ajy et al (2000) studied conventional and optimized (enhanced) coagulation for NOM removal in jar tests of raw water samples from 16 US water utilities and two water treatment plants in Pennsylvania. Results showed that enhanced coagulation can enhance the removal of TOC, DBP precursors, particles, and turbidity compared with conventional coagulation. The effectiveness of the optimized treatments depended on the pH of coagulation and the type and dosage of the coagulant.

As stated above, it is important to apply a minimum coagulant dosage or higher to comply with the water quality standard. On the other hand, coagulant overdosing resulted in excessive sludge production, need for more chemicals for pH and corrosion control, early filter breakthrough and increased rate of head loss development.

The negative effects of increased filtration rates on filtered water turbidity due to increased hydraulic loading when parallel filter units were taken out of operation to be backwashed were also clearly demonstrated in the experiments.

Based on numerous runs in the contact filtration pilot plant, operational models were developed as presented in Chapter 7.

3.2.2 *Optimum filtration conditions.*

Filters should be backwashed at the right time and with sufficient power to avoid solids accumulation over time. Continuous monitoring of turbidity or particle content from every filter unit is recommended in order to control the filtration efficiency including filter ripening and breakthrough, to identify malfunctioning filters, and to start backwash at the correct time. Filter head loss should also be measured, as well as head loss distribution within the filter bed that can give valuable information of the interactions between the filter bed and the water that is filtered.

Figure 3.8 illustrates well the typical phases of a filter run, with initial ripening, a period with good and stable filtrate quality, and finally a breakthrough of particles/turbidity. Compared to conventional turbidity removal, enhanced coagulation systems with elevated NOM and coagulant dosages typically yield short filter runs, i.e. more frequent backwash is required due to loose flocs and increased solids loads creating early breakthroughs and more rapid head loss development.

The importance of monitoring head loss and turbidity from each filter unit is well demonstrated in Figure 3.8. These are the parameters that terminate the filter run length and decide when backwash should be initiated.

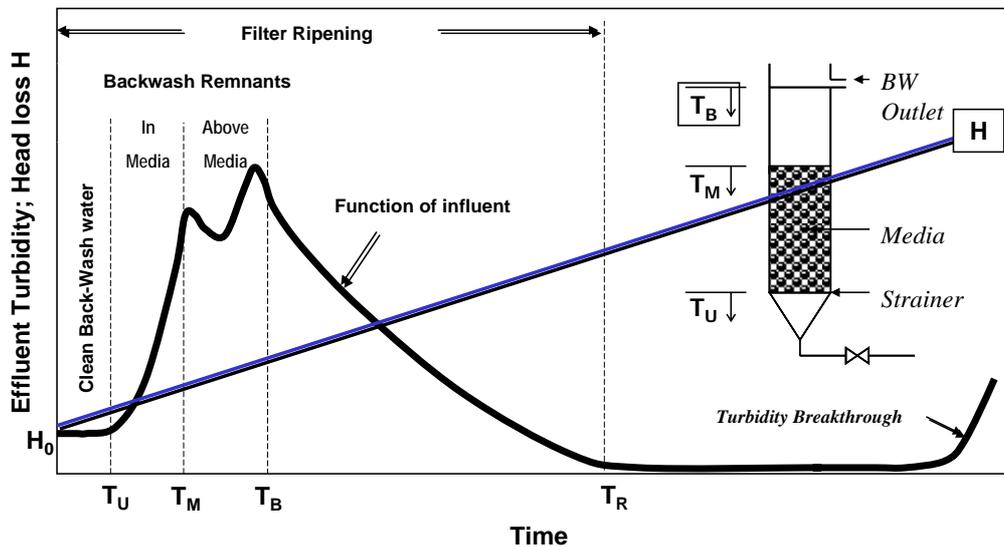


Figure 3.8. Typical filter ripening, and development of filter effluent turbidity and head loss during filtration. The detected turbidity pattern is explained from water initially present in the underdrains (T_U), within the pores of the filter media (T_M), and from the backwash remnant water above the media (T_B). Total time of ripening is T_R . Adapted from Amirtharajah, 2002.

Filter ripening

During filter ripening the filter effluent quality is deteriorated. As an example, removal of *Cryptosporidium* and *Giardia* is normally reduced by 0.5-1-log during ripening compared to a removal efficiency of 2-5-logs during the stable operation. Because the relatively large protozoan particles ripen faster than smaller particles, turbidity is considered a conservative measure for control of filter ripening. A filter-to-waste procedure is often used during ripening (normally 10-60 minutes), but ripening time can also be reduced by several other means, e.g.:

- More effective backwash techniques (Ch. 3.2.3)
- Coagulant overdosing in a short time period following backwash
- Treatment (coagulation) of backwash water

Filter rate control

The rate of filtration should be kept stable or declining during a filter run. It is well documented that even moderate, short-term increases in filtration rates can produce concentration peaks in turbidity, particle counts and micro organisms including protozoa.

Filter breakthrough

Enhanced coagulation using metal coagulants normally produce relatively short filter runs due to loose flocs and early breakthroughs. Because larger particles like protozoa breaks through earlier than the bulk of particles (turbidity), there is reason to be careful in the pre-breakthrough phase. Some facilities therefore initiate backwash before turbidity or particle breakthrough can be detected, i.e. after a certain filter run time or volume of water treated. In this way a safety factor can be included to avoid breakthrough of protozoa.

The application of filter aid polymers in small quantities (0.05-0.2 mg/L) have proven very effective in order to increase floc strength, increase attachment and/or decrease particle detachment within the filter bed and thereby prolong the filter run times. This allows for the use of higher filtration rates and thereby an increased treatment capacity. However, there is some variability in the level of success of polymer use, and there seem to be a need for more research on this issue.

3.2.3 Removal of protozoa during enhanced coagulation

States et al (2002) studied the influence of enhanced coagulation and decreased coagulation pH levels on the removal of *Cryptosporidium* oocysts, TOC, turbidity and particle counts. A series of pilot-plant trials were performed with commonly used coagulants (ferric chloride, alum, and poly aluminium chloride) at various pH levels to treat river water spiked with large numbers of *Cryptosporidium* oocysts. The results show that TOC removal is significantly enhanced by coagulation at lower pH levels and that all three coagulants are effective in removing *Cryptosporidium* oocysts (mean removal = 4.3 log units). However, turbidity and particle counts appeared to be unreliable indicators of oocyst removal. The investigation suggested that lowering coagulation pH does not interfere with removal of *Cryptosporidium*.

Dai and Hozalski (2002) performed experiments in bench-scale 0.25 m deep rapid filters with 0.55 mm glass beads to study how the removal of *Cryptosporidium* oocysts by filtration was effected by NOM and biofilms. They found that the oocyst removal efficiency was decreased as a result of presence of NOM and biofilm-coatings in the filter bed, indicating that water treatment facilities employing biologically active filters have a greater potential for oocyst breakthrough and proper coagulation is critical for effective removal of oocysts in the filters. Oocysts pre-equilibrated with NOM were more hydrophobic and significantly more negative than those obtained for untreated oocysts. Fortunately, the use of alum for coagulation was able to neutralize the surface charge of the NOM-coated oocysts and provide high removal efficiency.

Xagorarakis and Harrington (2004) evaluated the removal of viable *Cryptosporidium parvum* oocysts and changes in zeta potential during alum coagulation and sedimentation. The removal of oocysts and their zeta

potential was evaluated at three raw water DOC concentration levels and a wide range of alum doses and coagulation pH values. The study showed that the NOM content of the raw water, i.e. the initial DOC concentration affected the removal and zeta potential of the oocysts. Charge neutralization was not considered a relevant removal mechanism for oocysts under the conditions used in this study. Sweep coagulation appeared to be the primary removal mechanism at the lowest DOC concentration tested. For the highest DOC concentration used in this study, optimal coagulation for oocyst removal coincided with optimal coagulation conditions for NOM removal, suggesting that NOM plays a key role in the interaction between oocysts and the aluminium hydroxide precipitate.

Logsdon (2000) stated that deviations from optimum or near-optimum coagulation and improper management of filtration rate increases can severely deteriorate filtration performance for removal of protozoa. Likewise, improper backwash water recycling can disrupt coagulation.

Continuous monitoring of coagulation and filtration is an aid to effective management of the treatment process. Production of filtered water having a turbidity of 0.1 NTU or lower should be the goal if effective control of cysts and oocysts is to be attained. However, no concentration of cysts could be associated with a specific value of filtered water turbidity.

Hamilton et al (2002) concluded that if oocysts are present in a work's raw water, there is strong evidence to suggest that minimising treated water turbidity/particle count will reduce *Cryptosporidium* risk. However, neither particle counters nor turbidimeters are able to detect *Cryptosporidium* oocyst or reliably predict their occurrence in treated waters. Particle counters have demonstrated some benefits in three areas, namely:

- a) higher sensitivity to changes in water quality at low turbidities (<0.1 NTU)
- b) higher sensitivity changes associated with larger particle sizes (e.g. filter breakthrough events)
- c) the ability to monitor changes in particle size distribution

Logsdon (2000) reviewed five pilot plant investigations, treating raw waters with turbidity generally below 10 NTU:

- Logsdon et al (1981) investigated *Giardia* removal (seeded cysts) in direct filtration pilot trials with a dual media anthracite and sand filter, a filtration rate of 10 m/hr, and a coagulant dose of 10 mg/l alum (i.e. approx. 0.9 mg Al/l). Operational variations tested included sub-optimal and very inadequate coagulation, filtration rate increases, and turbidity breakthrough with high head loss.
- DeWalle et al (1984) investigated *Giardia* removal (seeded cysts) in a dual media anthracite and sand filter. Filtration rates were typically 10 m/hr, with variations within 4.3-10 m/hr. Alum at a dosage of 12

mg/l (i.e. approx. 1.1 mg Al/l) was used in most runs, with some testing of sub-optimal coagulation.

- Hendricks et al (1999) investigated *Giardia* and *Cryptosporidium* removal in two types of contact (in-line) filters: 1) a dual media anthracite-sand filter, and 2) a monomedium anthracite filter. An alum dosage of 26 mg/l (i.e. 2.4 mg Al/l) was generally used, with some runs at a sub-optimum dosage of 13 mg/l (i.e. approx. 1.2 mg Al/l).
- Ongerth and Pecoraro (1995) investigated *Giardia* and *Cryptosporidium* removal in direct mixed media filtration (anthracite over sand and garnet). The filtration rate was 12 m/h, and the optimum alum dosage was about 10 mg/l (i.e. approx. 0.9 mg Al/l) at a pH of 6.4-6.6. In a sub-optimal coagulation run, an alum dosage of 5 mg/l (i.e. approx. 0.4-0.5 mg Al/L) was tested.
- Patania et al (1995) investigated *Giardia* and *Cryptosporidium* removal in conventional filtration, and a filtration rate of 15 m/h. Ferric chloride was used as the coagulant, typically at a dosage of 15 mg/l with or without Cat-Floc T at 1 mg/l. They evaluated filtration at optimum coagulation conditions during filter ripening and later in the filter runs when stable filtered water quality had been attained.

Logsdon (2000) concluded from the review that at optimum coagulation conditions when filtered water turbidity was 0.1 NTU or lower, removal of cysts and oocysts was more effective than when turbidity was above 0.1 NTU. Sub-optimal coagulation resulted in filtered water turbidity in the range of 0.1-1 NTU. Higher concentrations of cysts and increased filtered water turbidity were observed during filter ripening. Filtration rate increases ranging from 50% to 150% in 10 seconds did not cause filtered water turbidity to increase when flocs were strengthened with a non-ionic polymer (Logsdon et al 1981). When alum was used with no polymer, a filtration rate increase from 10 m/hr to 27 m/hr for a period of two minutes caused turbidity to increase from 0.3 to 1.0 NTU, and the *Giardia* cysts concentration increased 25-fold. When the filtration rate was decreased to 10 m/hr, both turbidity and *Giardia* cyst concentration returned to levels observed before the rate increase.

The results also showed that turbidity breakthrough at the end of a filter run can be accompanied by a massive discharge of micro organisms. Recycling of filter backwash water by returning it to the influent raw water will normally increase the concentration of suspended solids and micro organisms in the influent water, and it may upset the coagulation chemistry.

Emelko (2003) used bench-scale glass filter columns to investigate dual- and tri-media filter removals of both viable and formalin-inactivated *Cryptosporidium parvum* oocysts. She found that formalin-inactivated *C. parvum* oocysts were good surrogates for viable oocysts in filtration studies conducted during stable operation, ripening and coagulation failure, and that

dual and tri-media filters performed equally well. *C. parvum* removal was high (4.6-5.8-log) during stable operation, but was moderately lower (4.0-5.7-log) during filter ripening. During the coagulant failure conditions, the *C. parvum* removal was severely compromised relative to both filter ripening and stable operation. Removals of non-coagulated oocysts ranged from 0.5-2.5-log.

3.2.4 Filter backwashing practices

A successful filtration process requires that the solids captured within the filter bed is carefully controlled and periodically removed by backwashing normally once to twice a day. In continuous filtration systems however (e.g. Dynasand), also the backwash is performed continuously.

Solids capture and solids accumulation capacity in a filter bed is normally controlled by measuring filter effluent turbidity (breakthrough) and head loss development. In some plants, the head loss distribution over the filter bed depth is measured as well, in order to control that the filter and solids/flocs to be filtered are well adapted to each other.

Filter backwash efficiency is affected by a number of factors, including:

- Filter configuration (1M, 2M, 3M, grain size and density, layer depth, etc.);
- Water temperature;
- Backwash water flow rate and duration;
- Air scour rate and duration;
- Filter bed (layer) expansion;
- Backwash technique, e.g. expansion type of water backwash; collapse pulse type of backwash using air and water simultaneously; extended terminal subfluidization wash (ETSW);
- Cleaning chemicals (chlorine, acids, bases, etc. used periodically in order to remove biofilms, inorganic precipitates, etc.).

Among the operational problems occurring in filtration processes, mudball formation and poor initial filtrate quality are associated with the effectiveness of backwashing. Backwashing with water alone to fluidize the media is a weak cleaning process, and the use of air scour simultaneously with subfluidization water wash (i.e. "collapse-pulsing" with air pockets forming and collapsing within the bed) can significantly improve the effectiveness of backwashing (Amirtharajah 1993). For dual or multi-media filters, the collapse-pulsing backwash should be followed by an expanding water backwash to re-stratify the filter bed/layer configuration.

Fluidizing backwash

A fluidizing type of filter backwash applies backwash water rates high enough to expand and fluidize the filter bed, i.e. rates above the minimum fluidization velocity (V_{mf}) are needed. Typically 5-10 minutes of water backwash is used. A short air backwash (1-5 minutes) is also often used initially. Water is needed as a final step in order to secure re-stratification of dual- or multi media filter beds.

Collapse-Pulsing backwash

Amirtharajah 2002 stated that collapse-pulse air scour (CPBW) gives best cleaning due to abrasion between filter grains. Conventional fluidization-type of water backwash is less effective than collapse-pulse backwash (CPBW), removing significantly less turbidity/particles and biofilm from the bed during backwash. CPBW can be obtained from the following equation:

$$aQ_a^2 + \% (V/V_{mf}) = b \quad (3.1)$$

where a, b = coefficients for a given filter media
 Q_a = air flow rate
 $\% (V/V_{mf})$ = per cent of minimum fluidization water flow



Fitzpatrick (2004) conducted experiments in a laboratory filter column to establish the best wash regime for filter sand taken from a water treatment plant in London, UK. A range of different air-water combinations were investigated, spanning a range of collapse-pulsing combinations. Chlorinated backwash water was also used to assess whether this could be effective in removing fouling layers/biofilm on the filter grains. Best cleaning in the laboratory was obtained by collapse-pulsing, with a water velocity at 60% of V_{mf} and an air velocity of 48 m/h. This resulted in the cleanest grain surfaces, particularly after 20 ppm chlorine was employed in combination with the air-water wash.

The extended terminal subfluidization wash (ETSW)

The extended terminal subfluidization wash (ETSW) is an advanced backwashing strategy that involves extending the normal backwash duration at a subfluidization flow rate (with little or no bed expansion) for an amount of time sufficient to displace the entire volume of water contained within the filter box, thereby removing the backwash remnant particles that are normally left within and above the media (Figure 3.8), and without generating further remnants that can pass through the filter following restart (Amburgey and Brouckaert, 2005).

The minimum fluidization velocity can be calculated as follows:

$$V_{mf} = \mu / \rho d_{eq} (33.7^2 + 0.0408Ga)^{0.5} - 33.7 \mu / \rho d_{eq} \quad (3.2)$$

where Ga is the Galileo number defined as:

$$Ga = d_{eq}^3 \rho (\rho_s - \rho) g / \mu^2 \quad (3.2)$$

where g is the acceleration of gravity, d_{eq} is the grain diameter, μ is the dynamic viscosity of the water, and ρ_s and ρ are the mass density of the filter media and the water, respectively.

The minimum fluidization velocity for dual media filters can be calculated as follows (Amirtharajah et al 1991):

$$V_{mf \text{ dual}} = V_{mf \text{ anthracite}} [V_{mf \text{ sand}} / V_{mf \text{ anthracite}}]^{x_s^{1.69}} \quad (3.4)$$

where x_s is the mass fraction of sand in the filter.

Equation (3.4) is valid when the anthracite has a higher V_{mf} than the sand, otherwise the sand and anthracite terms must be interchanged.

Amburgey and Brouckaert, 2005 performed illustrating calculations on minimum fluidization velocities and ETSW backwash requirements for relevant filter media configurations. Table 3.2 shows some typical properties of sand and anthracite filter media. These values can be used to determine the minimum fluidization velocities shown in Table 3.3 of the d_{10} -sized media grains of hypothetical filter designs at a range of water temperatures, using Equations (3.2) to (3.4). Table 3.4 contains the calculated minimum fluidization velocities for the d_{90} -, d_{60} -, and d_{10} -sizes of the selected filter media at a constant water temperature of 20 °C. Some experimentation is required, however, to select the final ETSW rates and durations at specific sites.

Table 3.2 Specification and properties of some typical graded beds of filter media (Amburgey and Brouckaert, 2005).

	Sand	Anthracite	450mm anthracite and 300mm sand	Deep bed anthracite
Effective size (ES) d_{10} (mm)	0.55	1.1	1.1 and 0.55	1.6
Uniformity coefficient (UC = d_{60}/d_{10})	1.4	1.3	1.3 and 1.4	1.3
d_{60} (mm)	0.77	1.43	1.43 and 0.77	2.08
d_{90} (mm)	0.96	1.70	1.70 and 0.96	2.48
Media density (g cm^{-3})	2.6	1.55	1.55 and 2.6	1.55

Table 3.3 Calculated minimum fluidization velocities for typical d_{10} -sized media (Amburgey and Brouckaert, 2005).

	Sand	Anthracite	450mm anthracite and 300mm sand	Deep bed anthracite
V_{mf} (0°C) (m h^{-1})	5.8	7.8	6.9	16.0
V_{mf} (5°C) (m h^{-1})	6.7	9.1	8.1	18.4
V_{mf} (10°C) (m h^{-1})	7.8	10.5	9.3	20.9
V_{mf} (15°C) (m h^{-1})	8.9	11.9	10.6	23.3
V_{mf} (20°C) (m h^{-1})	10.1	13.4	12.0	25.8
V_{mf} (25°C) (m h^{-1})	11.3	14.9	13.4	28.2
V_{mf} (30°C) (m h^{-1})	12.6	16.4	14.8	30.4

Table 3.4 Calculated minimum fluidization velocities for various media sizes at 20 °C (Amburgey and Brouckaert, 2005).

	Sand	Anthracite	450 mm anthracite and 300mm sand	Deep bed anthracite
V_{mf} d_{10} -sized media (m h^{-1})	10.1	13.4	12.0	25.8
V_{mf} d_{60} -sized media (m h^{-1})	19.1	21.4	20.5	38.4
V_{mf} d_{90} -sized media (m h^{-1})	28.6	28.5	28.5	48.5

3.2.5 *Filter bed evaluation methodology and inspection*

Ceronio et al (2002) investigated three well-run water treatment facilities in South Africa to measure their operational efficiency. The investigation methodology included a detailed evaluation of filter media, backwashing procedures and backwashing efficiency.

The evaluation of the filter media was based on the guidelines for filter evaluation suggested by AWWA (2000) and consisted of the following procedures:

- a) Visual inspection of the filter for cracks, holes, depressions or mounds
- b) Determination of washwater through and walkway levels
- c) Determination of media levels to determine media loss
- d) Gravel layer assessments - to determine underfloor system integrity and washwater distribution
- e) Mudball and media intermixing inspections
- f) Filter coring for sieve analysis
- g) Floc retention analysis to determine backwash efficiency
- h) Bed-expansion and rate-of-rise measurements
- i) Backwash turbidity analysis, to determine the efficiency of backwash water usage.

It was found that the three plants had the following problems in common: excessive media loss; inconsistent media gradings; modified backwash velocities; inefficient backwash water utilization and the presence of mudballs. Of these, the low backwash velocities were found to be one of the most significant problems.

4 Operational challenges and bottlenecks

4.1 Coagulant dosage and coagulation pH

Proper coagulation is essential for good clarification and filtration performance and for the control of pathogens and disinfection by-products. Improper coagulation can cause high residual coagulant residuals in treated water and post-treatment precipitation of particles causing turbidity, deposition and coatings of pipes in the distribution system. Minimizing of solids residuals from coagulation has also become a more important part of utility operations due to increased disposal costs and landfilling restrictions Pernitsky and Edzwald (2006).

These issues have put additional pressure on utilities to optimize coagulation to meet the multiple treatment objectives (modified after Edzwald and Tobiason, 1999): 1) to maximize the removal of particles, turbidity and microorganisms/pathogens by downstream solid-liquid separation, 2) to maximize TOC and DBP removals, 3) to minimize residual coagulant concentrations, 4) to minimize solid residuals (sludge) production, and 5) to minimize operating costs.

Budd et al (2004) stated that optimization of coagulation is central to the drinking water industry's ability to meet goals for particulate (turbidity) and NOM removal. They stressed the importance of adopting a holistic view of treatment objectives when considering possible changes to the coagulation process, and highlight the necessity of evaluating coagulation as a multiple-input process that can be fine-tuned through adjustment of two fundamental parameters - pH and coagulant dose. Changes that might be undertaken include trying a different coagulant dose and pH, using alternative coagulants, and adding coagulants in a different sequence. Their recommendations were based on coagulation evaluations performed at a number of US water treatment facilities over the past 15 years.

Gregory and Carlson (2003) studied the impact of coagulation pH, zeta potential and floc formation kinetics on particle removal during settling and filtration. They concluded that higher coagulation pH - and thus higher alum coagulant doses - could be advantageous during periods of rapidly changing water quality conditions, such as high-NOM runoff events. The rate of floc formation measured immediately following coagulant addition could be indicative of overall process performance.

Pernitsky and Edzwald (2006) presented guidelines for the selection and use of polyaluminium chloride (PACl) and alum coagulants in terms of raw water quality and treatment method. The concentration of NOM was found to be the most important parameter affecting coagulant dose. SUVA was useful for characterizing the nature of the NOM, and thus for predicting the degree of NOM removal expected. Raw water turbidity and NOM did not influence the

type of coagulant that was most effective. Raw water alkalinity, as it related to the pH of coagulation in cases where independent pH control is not used, was found to be very important for choosing one coagulant type over another. PACl basicity should be matched to raw water alkalinity, so that coagulation pH is as close as possible to the pH of minimum solubility of the coagulant. The solids separation process used was also found to be important for coagulant selection. Raw waters coagulated with PACls containing sulphate was found to have the best settling characteristics, but showed the highest headloss rates in direct filtration applications. Dissolved air flotation performance was relatively insensitive to coagulant type.

Coagulant dosage and coagulation pH is traditionally determined according to results from jar-tests or operator's experience, which often lead to coagulant overdosing or insufficient dosing, reduced water treatment performance and increased treatment costs.

From extensive pilot experiments, Eikebrokk (1996, 1999) showed how residual metal (Al, Fe) determined the coagulant dosage requirement in order to meet the relevant Norwegian drinking water quality standards for treatment plants applying coagulation (i.e. Residual Al or Fe < 0.15 mg/L; Turbidity < 0.2 NTU, Colour < 5 mg Pt/L, and TOC < 3 mg/L).

The example of pilot investigation results using ferric coagulation-contact filtration presented in Figure 4.2 illustrates that although TOC removal efficiency in excess of 60% was obtained with a coagulant dosage of 5 mg Fe/L, a dosage of 6.5 mg/L was required to comply with the residual Fe standard of 0.15 mg/L. With 5 mg/L of Fe, a residual Fe concentration in excess of 0.4 mg/L was detected. In addition, the figure demonstrates well how an increase in coagulant dosage level is able to widen the pH-range of optimal treatment performance. With the 5 mg Fe/L dosage, the optimum pH-range is very narrow (4.9-5.2). With 6.5 mg Fe/L however, more than 60% TOC removal was achieved within a wide range of pH (3-6). However, even 6.5 mg Fe/L was close to the minimum dosage required in order to comply with the 0.15 mg residual Fe/L standard within a reasonably wide pH-range (5.0-5.5).

Similar curves and relationships were obtained with alum and poly aluminium chloride, but the optimal pH-values were higher with these coagulants (pH 5.5-6.5).

Emelko (2003) investigated the removal by alum coagulation of viable as well as formalin-inactivated *Cryptosporidium parvum* oocyst in bench-scale dual- and tri-media filters. Her results indicated that formalin-inactivated oocysts were suitable surrogates for viable oocysts. She also found that poor coagulation conditions severely compromised removal of viable and inactivated oocyst by dual- and tri-media filters compared to stable operating conditions and filter ripening, emphasizing the importance of optimized coagulation for the successful removal of oocysts during filtration. During filter ripening, the *C. parvum* removals were moderately lower

(approximately 0.5-1-log) than during stable operation, while during coagulation failure conditions the *C. parvum* removal capacity of both dual- and tri-media filters was severely decreased (by more than 3-log) relative to both ripening and stable (optimised) conditions (Figure 4.3). Tri-media (3-M) filters offered only marginally higher oocyst removals than dual-media (2-M) filters.

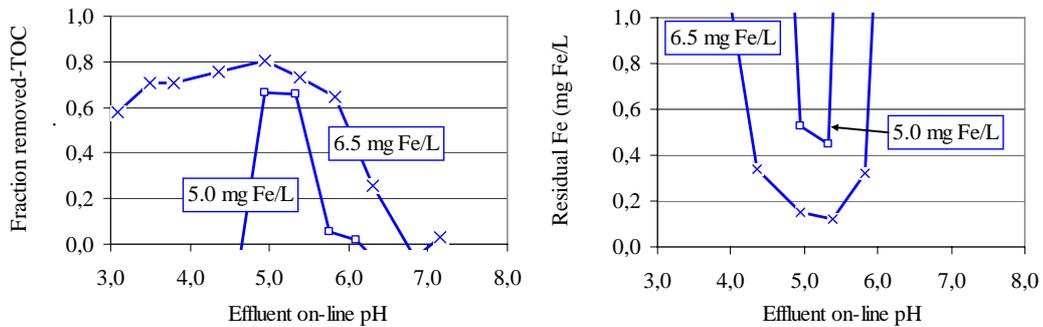


Figure 4.2 Effects of ferric chloride dosage level and coagulation pH on the removal of DOC (left) and residual Fe (right) in enhanced coagulation- dual media contact filtration pilot-scale experiments. (Raw water: 50 mg Pt/L; 5 mg TOC/L; SUVA 4.8 L mg/m.) (Adapted from Eikebrokk, 1999).

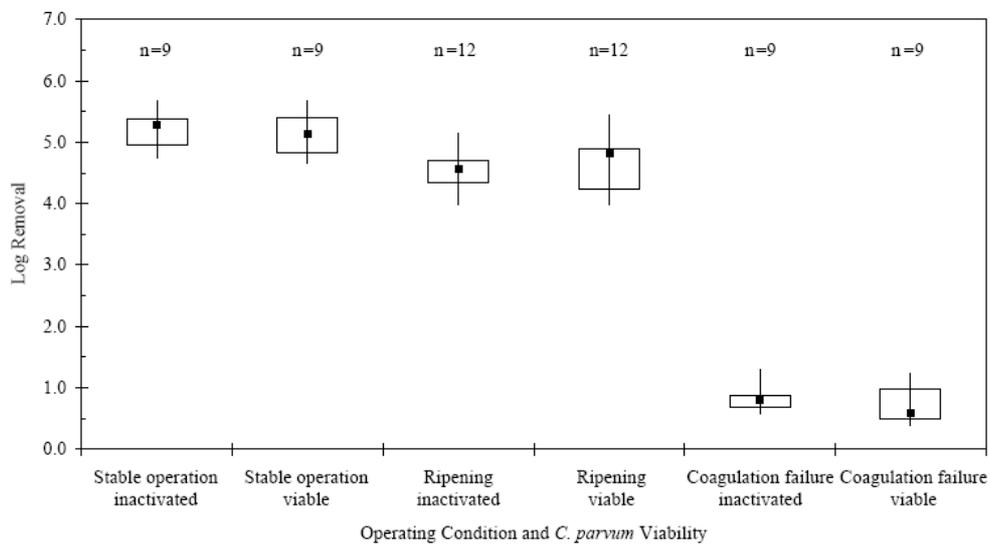


Figure 4.3 Dual media removals of viable and inactivated *C. parvum* oocysts by alum coagulation and filtration during stable operation, filter ripening, and coagulation failure (n = number of sample pairs). Emelko, 2003.

Although a sub-optimal coagulant dose may result in excessive residual metal concentrations, reduced removal of pathogens like *Giardia* and *Cryptosporidium*, and non-compliance with the water quality standards, coagulant overdosing should be avoided. In conventional filtration applying pre sedimentation the negative effects of overdosing is normally limited to

increased costs, excessive sludge production, etc. However, restabilization may occur, resulting in poor treatment performance.

The negative consequences of coagulant overdosing are however significantly larger in direct and contact filtration processes where the filter has to cope with the excessive amount of sludge formed. The example shown in Figure 4.3 illustrates the negative effects of alum coagulant overdosing in contact filtration processes. The figure demonstrates the negative effects of coagulant overdosing on filtration performance in terms of breakthrough and head loss development, and thereby also on dual media filter run time and treatment capacity/net water production. From an optimum coagulant dosage of 1.0 mg Al/L, a dose increase to 1.8 and further to 3.1 mg Al/L, increases the solids load to the filter and reduces the filter run time from about 15 hours to about 7.5 and 6 hours, respectively. The head loss increase as a result of coagulant overdosing is also significant, from 6.3 cm/hr at the optimum dosage of 1.80 mg Al/L to 9.2 and 12.8 cm/hr at alum doses of 1.8 and 3.1 mg Al/L, respectively. The applied filtration rate was 7.5 m/hr.

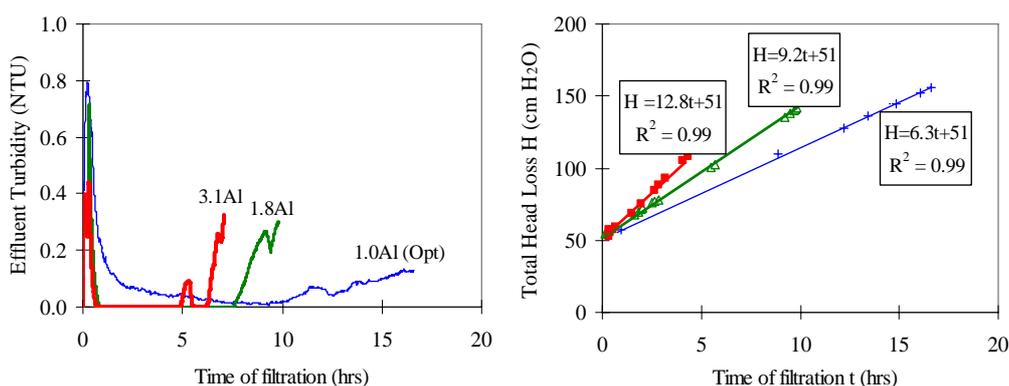


Figure 4.3. Effects of coagulant dosage level on filter run time, i.e. terminated by breakthrough and/or head loss. (Contact filtration with alum; 2-M filter; 7.5 m/hr). Eikebrokk, 1999.

4.2 Effects of temperature

Temperature affects the solubility of the metal hydroxide precipitate and the rate of formation of the metal hydrolysis products. Cold water temperatures often cause decreased performance for sedimentation and make proper coagulant selection more important. High-basicity PACl are reported to be less affected by cold temperatures than alum is, and the presence of sulphate in a PACl resulted in a better settling floc. The negative effect of temperature tends to be greatest with dilute suspensions.

Direct filtration and DAF processes are much less affected by cold water temperatures than sedimentation is (Pernitsky and Edzwald, 2006).

4.3 Effect of pH adjustment on overall performance of the plant

Enhanced coagulation involves adjustment of pH, alkalinity and increase coagulant doses. This can result in number of secondary impacts on water treatment (AWWA, 2005):

- *Increases solids.* The higher coagulant dosages directly result in increased sludge volumes.
- *Poorer dewatering characteristics.* The increased metal (Al^{3+} , Fe^{2+} , Fe^{3+}) concentrations typically result in poorer dewatering characteristics. As a result, a change to enhanced coagulation may result in lower ultimate, dewatered solid concentrations.
- *Increased concrete/metal corrosion.* The lower pH of the coagulated water for TOC removal will be significantly more aggressive on concrete and metals as compared to a more neutral coagulation pH used for turbidity removal.

If pH is lowered to improve coagulation and organic removal, it is typically necessary to raise pH in the final effluent from the plant to provide a less corrosive finished water. The pH may be adjusted at one or more points in the treatment, including rapid mixing, prefiltration and postfiltration. In case of enhanced coagulation it is recommended to readjust the pH after the filtration process as compare to prefiltration. This is due to the fact that some organic matter may be adsorbed onto the floc that may carry over from the clarification process, and any prefiltration pH adjustment may then result in “release” of that organic matter, which could pass through the filters and contribute to subsequent DPB formation.

One of the greatest practical problems faced in removing soluble NOM from low turbidity waters is inability to produce an acceptable floc (Gregor et al 1997). Natural turbidity provides a ready source of nucleating sites for floc development, and once present, these flocs act as adsorption sites for soluble NOM. For low turbidity waters, these essential floc nuclear sites can be provided by lime that is used for pH and alkalinity correction, provided the lime is added in sufficient quantity and at the point where it retains some particulate nature. If pH adjustment with lime is not needed, bentonite clay or activated silica can be used as a coagulant aid.

4.4 NOM removal and filter run times

It is well known that metal-based coagulation of NOM forms loose flocs and results in short filter run times compared to conventional turbidity removal. Polymers can be effective in order to increase floc strength and/or attachment forces within a filter bed, and thus maintain reasonable filter run times even at relatively high filtration rates.

Conventional filtration

In conventional filtration systems applying flocculation and sedimentation prior to filtration, a large proportion of the solids (sludge) is removed during sedimentation. The solids loads to the filter is reduced as a result of the pre-settling, and relatively long filtration times can be obtained before breakthrough or head loss terminates the filter run.

However, sub-optimal coagulant dosage will result in inadequate treatment performance with respect to NOM and pathogens, filtered water turbidity, and residual coagulant concentrations.

On the other hand, coagulant overdosing may reduce treatment performance due to restabilization of colloids in some cases. In addition, coagulant overdosing will inevitably result in excessive sludge formation and increased chemicals and residuals management costs. However, it can also increase the floc volume fraction, and thus increase flocculation and separation efficiency. For high alkalinity raw waters, coagulant overdosing may be a practical alternative to supplemental acid addition to depress coagulation pH to optimum values.

For low alkalinity raw waters however, coagulant overdosing can create an additional need for base addition to prevent pH from being reduced to sub-optimal values. Polymeric coagulants with high basicity may be suitable in such situations.

Dissolved air flotation (DAF)

Enhanced coagulation followed by dissolved air flotation (DAF) and filtration processes may be a good treatment alternative for NOM removal, especially when NOM levels are too high for direct and contact filtration applications, and where algae removal is required. DAF requires good coagulation so that the air bubbles can attach to the floc particles. Pernitsky and Edwald (2006) found that DAF was relatively insensitive to coagulant selection. However, high-basicity PACls were more effective at lower doses and/or over a broader range than alum or other PACls for some waters.

Direct and contact (in-line) filtration

Enhanced coagulation and direct or contact filtration has proved a cost-efficient treatment alternative for raw waters with a low to moderate coagulant demand, thus minimizing sludge production and solids load to the filters. These processes are considered feasible for raw waters with turbidity less than 10 NTU, colour less than 40 mg Pt/L and algae biomass less than 10 µg/L as chlorophyll-a (Janssens and Buekens 1993), i.e. for raw waters with a relatively low coagulant demand.

Coagulant overdosing is normally not a good strategy in direct or contact filtration applications, because filter run times are sensitive to solids loading. Thus, coagulant overdosing will normally result in early filter breakthrough, increase in the rate of head loss development and thus reduced filter run

times and reduced treatment capacity. Treatment performance may also be compromised because of the increased frequencies of filter backwash and filter ripening.

Due to the relatively short filter run times normally obtained in enhanced coagulation followed by direct- or contact filtration processes, it is particularly important to avoid coagulant overdosing in such processes. The use of a polymer as filter aid in order to increase floc strength and/or attachment forces within the filter bed has proven to be an efficient tool to increase filter run times, and/or to allow for the use of higher filtration rates while still maintaining reasonable filter run times. However, the results from filter aid polymer applications appear inconsistent; In some cases the filter run times can be increased 2-3 times compared to the use of a coagulant alone (Eikebrokk, 1982), while in other cases the effects are rather small. The reasons for this do not seem well known or described in literature.

Due to the high coagulant dosage requirements, and the increased sludge production and solids loading to the filters, filter run time and treatment capacity considerations are of specific interest for enhanced coagulation applied in direct- and contact filtration processes.

Treatment capacity considerations - Net water production from a filter unit

Net water production, i.e. net water produced per unit time and area, can be applied as an optimization criterion for the filter units. When filtered water is used for backwash, the net treatment capacity, i.e. the net daily water production, per unit filter area can be calculated as the number of filter runs per day times the net water production per filter run (Eikebrokk 1982):

$$Q_d = 24/t_p (v_f \cdot t_f - v_s \cdot t_s) \quad (4.1)$$

- where
- Q_d = Net daily water production ($m^3/m^2 \cdot \text{day} = m/d$)
 - v_f = Filtration rate ($m^3/m^2 \cdot \text{hr} = m/\text{hr}$)
 - t_f = Filter run time (hrs)
 - v_s = Water backwash rate ($m^3/m^2 \cdot \text{hr} = m/\text{hr}$)
 - t_s = Duration of water backwash (hrs)
 - t_p = Total time between each filter run = $t_f + t_s + t_a$
 - t_a = Additional time elapsed during backwash (air backwash, valve operation time, etc.)

This relationship is illustrated in Figure 4.4, where net daily water production Q_d is calculated as a function of filter run time for a number of filtration rates in the range of 5 to 20 m/hr. The capacity is calculated for two situations: A) No loss of filtrate during filter ripening, and B) Filtrate to waste during filter ripening. The figure illustrates well the huge impact of the filtration rate on net water production, and the relative insignificance of filter run time provided this is 6-8 hours or above.

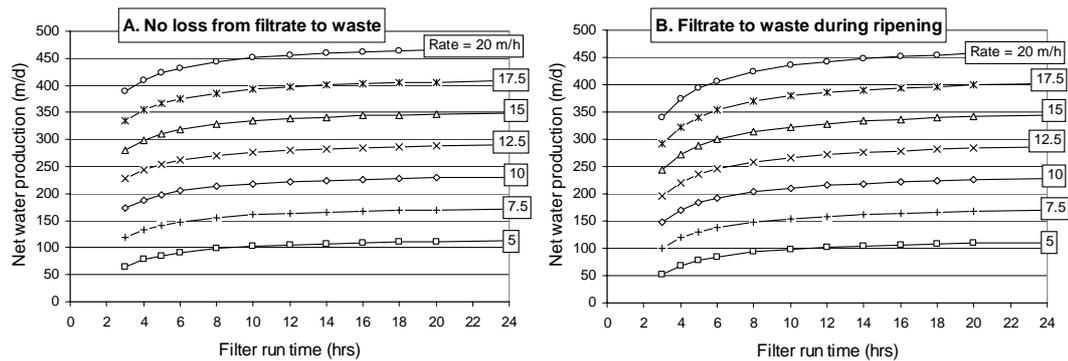


Figure 4.4. Effects of filter run time and filtration rate on the net daily water production per unit filter area. (Calculated with filter ripening time: 20 min; v_s : 45 m/hr; t_s : 8 min; t_a : 12 min).

With the net water production as a basis for considerations on treatment capacity and optimum operating conditions, the relationship between filtration rate and the obtained filter run time (i.e. until breakthrough or head loss termination) is an key operational parameter that is affected by the coagulant dosage level, the use of a polymer as filter aid, etc. Compared to conventional coagulation and turbidity removal, NOM removal by enhanced coagulation and metal coagulants yield loose flocs and relatively short filter run times.

For enhanced coagulation and contact filtration processes, Figure 4.5 shows an illustrating example on how filtration rate affects filter breakthrough and filter run time. All filter runs were terminated by breakthrough, and the terminal head loss with alum alone was about 0.5 m of water gauge. With polymer, however, terminal head loss was close to 2 m. Thus polymer is a very valuable tool with respect to optimization of treatment capacity. Care should be taken, however, regarding polymer dose. In the experiments described above, polymer doses in excess of 0.2 mg/L tended to create backwash problems and increased risk of mud ball formation.

Such informative relationships between filtration rate and filter run time can be established from a limited set of experiments with varying filtration rates in pilot or full-scale facilities using actual coagulation conditions. Figure 4.5 illustrates well the great improvement in filter run time that can be obtained by the application of a polymer as filter aid. In other words, the use of polymer allows the application of higher rates of filtration while still maintaining reasonably long filter run times. As shown in Figure 4.4, this will increase treatment capacity.

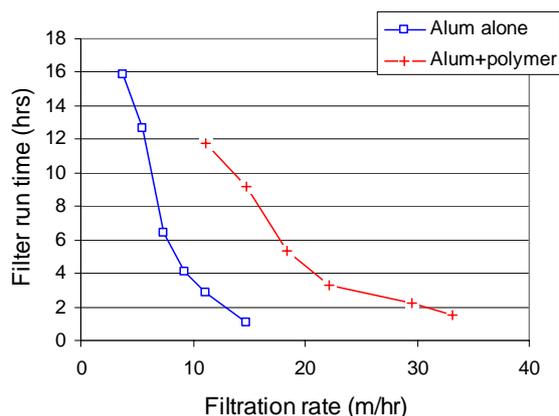


Figure 4.5. Effects of filtration rate on obtained filter run time in contact filtration experiments treating low turbidity (< 0.5 NTU), soft raw waters with colour of 20-45 mg Pt/L with alum alone, and with alum in combination with a non-ionic poly acryl amide as filter aid (0.1 mg/L). Dual media filter: 0.53 m 0.8-1.6 mm anthracite and 0.27 m 0.4-1.0 mm sand. (Eikebrokk, 1982).

Regardless of the type and degree of separation prior to the filter unit, optimal enhanced coagulation conditions are required to optimize the overall treatment performance. Major challenges seem to exist regarding optimum treatment process performance and operation, related in specific to residual coagulant concentrations, turbidity, NOM, pathogens removal (e.g. *Giardia* and *Cryptosporidium*), filter run times, and treatment capacity.

4.5 Metal coagulant residuals

As discussed above, total residual metal coagulant concentrations often determine the coagulant dosage needed in order to comply with the water quality standards.

Srinivasan et al (1999) analysed data from the Buffalo Pond WTP in Saskatchewan, Canada, in order to examine seasonal variations and factors influencing residual aluminium concentrations. Analysis of eight-year data showed that the DOC present in the raw water played a major role in controlling efficacy of alum coagulation. They found that when alum/DOC ratio was less than 7, insufficient alum addition led to incomplete coagulation resulting in colloidal material mostly consisting of organic aluminium in particulate form. Hence particulate aluminium increased in treated water. But this increase in residual aluminium did not increase the turbidity of treated water. This indicated that an adequate alum dose in response to dissolved organic carbon is important in minimising residual aluminium in treated water. The plant data also showed that when freshly regenerated GAC contactors were used, peaks of dissolved aluminium occurred as a result of alkaline metal (calcium and magnesium) oxides present in the regenerated GAC that shifted the pH of filtered water to alkaline range with consequent formation of soluble aluminium species like $Al(OH)_4^-$.

Srinivasan and Viraraghavan (2004) conducted aluminium speciation experiments in a pilot scale water treatment plant using different alum doses. In addition, they conducted jar tests at various alum/DOC ratios. They concluded that an alum/DOC ratio of at least 7.3 should be maintained in order to meet the operating guidelines of 100 µg/L of total aluminium proposed by Health Canada given that finished water soluble aluminium levels may be in the range of 35-40 µg/L. For lower alum/DOC ratios (1.37 and 5.3) most of the total aluminium in filtered water was in the form of particulate aluminium, and soluble organic aluminium increased compared to the level in raw water.

Jekel (1986) investigated the interactions of humic acids and aluminium coagulants and reported that at low coagulant dosages, i.e. less than 10 mmol of Al^{3+} /g DOC in raw water (i.e. 0.27 mg Al/mg DOC), high residual aluminium levels were found and low amounts of DOC were removed. He concluded that the minimum dosage of aluminium should be in the order of 20-40 mmol Al^{3+} per gram of DOC (i.e. 0.54-0.108 mg Al/mg DOC) to overcome the complexing and coagulation problem and to achieve low aluminium residuals.

Frommell et al (2004) discussed how minimization of aluminium residuals in plants using aluminium-based coagulants can help in minimizing the post-precipitation potential in the distribution system, thereby minimizing also the financial and public health risk associated with the formation of precipitates in water distribution systems. They examined the ability of orthophosphate to react with aluminium and thereby reduce residual aluminium coagulant concentrations. Results from bench-scale tests where orthophosphate was added during rapid mix showed a consistent ability to reduce residual aluminium concentrations when alum was used as a coagulant. The effect of orthophosphate addition varied with dose, pH, temperature, and chemical addition sequence. From these results, the use of orthophosphate as a residual aluminium control method was proposed.

Eikebrokk (2005) tested how orthophosphate addition affected residual aluminium concentrations in a dual media contact filtration pilot plant treating low- turbidity, humic raw water (0.12 ± 0.03 NTU; 4.30 ± 0.47 mg TOC/L). Alum was used for coagulation (3.0 mgAl/L) at a coagulation pH of 6.0. Orthophosphate added just after the coagulant was tested in three dose levels (0.18; 0.8; and 1.8 mg P/L). The results indicated that orthophosphate was effective when added in low and stoichiometric amounts relative to the actual residual aluminium concentration, i.e. the addition of 0.18 mg P/L for a residual Al-concentration of 0.15 mg Al/L reduced the residual Al-concentrations significantly. Under these conditions turbidity, sludge production, and filter run times was not affected to any great extent.

Overdosing of orthophosphate however, resulted in significantly increased residual aluminium and phosphate concentrations, increased sludge production, more rapid head loss development, reduced filter run times and reduced NOM removal efficiency. Therefore, a successful application of

orthophosphate as a tool for residual coagulant control in contact filtration systems is supposed to be rather challenging because of the dependency of a strict process control system where the amount of orthophosphate added must be dynamically controlled by – and adapted to – the actual residual coagulant concentration.

Alkaline postfiltration

The use of enhanced coagulation with integrated corrosion control using dual media filtration and alkaline post filtration with CO₂ has gained a lot of interest in recent years. In Norway, several plants of this kind have been built in the past 10-15 years. The alkaline filter material (crushed CaCO₃) is used as an additional bottom layer (2-3 m deep) in anthracite-sand filters, or as a separate filter unit.

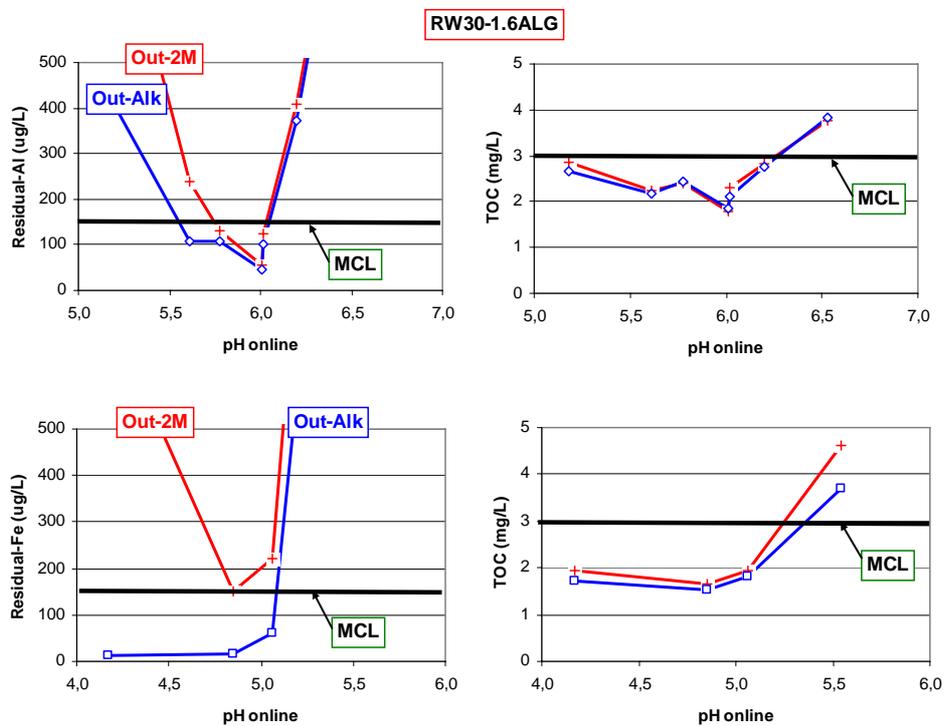


Figure 4.6. Residual Al and TOC concentrations (upper figure), and residual Fe and TOC (lower), in dual media filter effluents (Out-2M), and in dual media and post alkaline filtered water (Out-Alk). Pilot-scale enhanced coagulation-contact filtration; Raw water: 30 mg Pt/L and 4.3 mg TOC/L; Alum and ferric doses of 1.6 and 3.8 mg Me/L, respectively. Eikebrokk (2005).

As discussed before, residual coagulant concentrations normally determine coagulant dosage requirements in enhanced coagulation processes. In addition to the ability of increasing pH, calcium and alkalinity levels, a main benefit of the alkaline filter layer is the ability of reducing the residual metal coagulant concentrations. Figure 4.6 illustrates well how a post alkaline filter is able to reduce significantly the residual Fe-concentration from an enhanced coagulation-dual media contact filtration process, thereby allowing a reduction in coagulant dosage. In this way, the combination of enhanced coagulation and corrosion control by alkaline post filtration may reduce the

coagulant demand, the sludge production, and the solids load to the filters. This in turn will increase filter run times and increase the applicability of direct and contact filtration processes in the direction of raw waters with higher than recommended NOM levels.

4.6 Operational problems due to seasonal variation in NOM

Chow et al (2005) collected raw and treated water samples (prior to chlorination) on a monthly basis from two Australian WTPs for an 18-month period in 2001 and 2002. They calculated the 'specific alum demand' to evaluate DOC removal performance, defined as the required alum dose divided by the concentration of DOC removed by the treatment process. The required alum dose was the plant alum dose subtracted by the dose required to remove turbidity (i.e. 1 mg/L of alum per 0.48 NTU). From this approach, the great seasonal variation in specific alum coagulant demand is well illustrated (Figure 4.7).

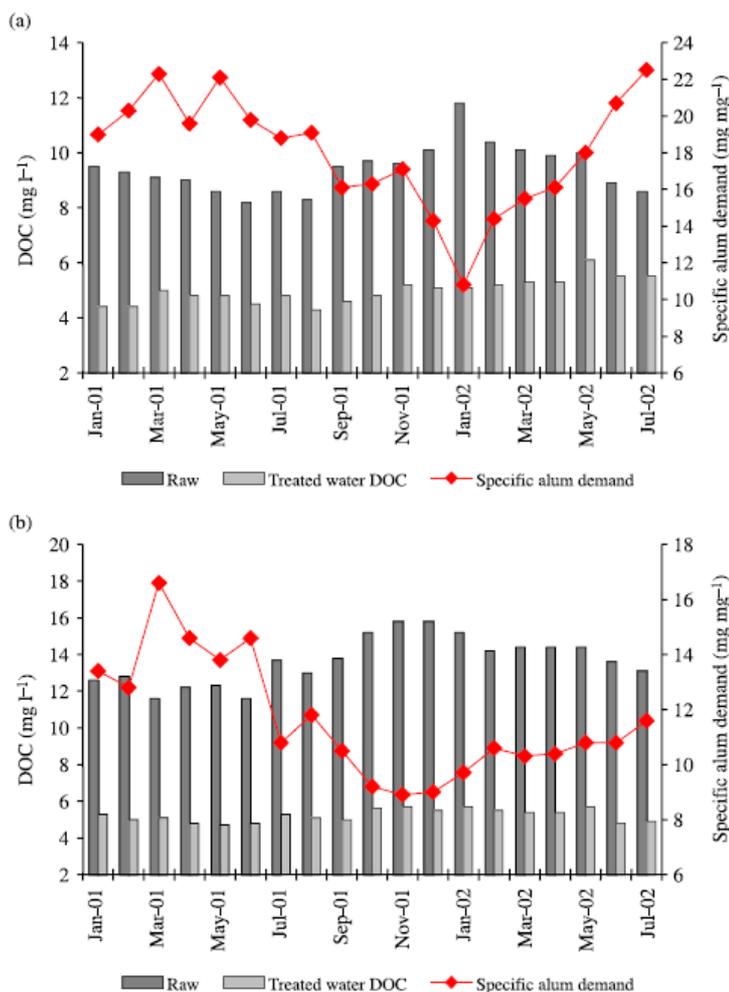


Figure 4.7 Seasonal variation in raw and treated water quality and specific alum demand: a) Happy Valley and b) Myponga WTP. Chow et al 2005.

In addition, Figure 4.7 shows that despite of the increase in raw water DOC levels near the end of 2001, the specific alum demand was significantly

reduced. Fractionation studies revealed that the increase in DOC was mainly an increase of the VHA fraction, a fraction that is typically considered easily removable by alum coagulation.

The results presented in Figure 4.7 illustrate well the challenges in coagulant dosage control imposed by the seasonal variation in NOM concentration and character.

Fearing et al (2004) investigated NOM control options related to seasonal periods of high rainfall that was shown to cause elevated NOM loadings at treatment plants and correspondent difficulties in removing sufficient NOM to meet water quality standards (i.e. THM) in the UK. Three treatment options for improved NOM removal were studied, either by: 1) optimising current coagulation processes, 2) pre-coagulation, and 3) post-coagulation. From NOM fractionation into hydrophobic and hydrophilic fractions it was shown that certain fractions were more recalcitrant to treatment during the times of high rainfall and high NOM loadings. By optimising coagulation for these fractions a staged coagulation was proposed. Although no significant increase in the removal of DOC or UV-adsorption could be observed as a result of the optimisation, filter run times were significantly increased.

Furthermore, the combination of ferric coagulation and magnetic ion exchange (MIEX®) showed that although the DOC in treated water was not significantly reduced, the THM formation potential was reduced by more than 50% for the lower MW compounds that are known to be untreated by conventional coagulation. Finally the addition of a range of adsorbents including carbons, hydroxides and clays to both raw water and the isolated low MW fractions of NOM showed that an increase in DOC and UV254 removal was achievable.

Bose and Reckhow (2006) fractionated NOM from a surface water source and identified fulvic acids and hydrophilic neutrals as the two most abundant fractions. Adsorption affinity of these fractions on preformed aluminium hydroxide flocs was found to increase with increasing charge of the fractions, except for the two most highly charged fractions, fulvic acids and hydrophilic acids, which showed less adsorption affinity than expected from their specific organic charge. Preozonation of the raw water resulted in a decline in DOC removal by alum coagulation with increasing ozone dosages. It appeared that ozone applied to raw water reacted preferentially with the humic fractions of NOM, resulting in the detrimental effects of ozonation on the subsequent NOM removal by alum coagulation being magnified. Ozonation of the isolated NOM fractions showed that prior ozonation of the fulvic acid fraction resulted in a decline in adsorption affinity on aluminium hydroxide surface. For the hydrophilic neutral fraction however, adsorption affinity increased as a result of preozonation. Ozonation of pre-coagulated water demonstrated beneficial effects of ozonation on the removal of non-humic fractions of NOM through alum coagulation. In order to maximize DOC and UV254 removal for raw waters containing both humic and non-humic NOM, a staged coagulation treatment with intermediate ozonation was proposed.

4.7 Increasing NOM concentrations due to climate change

Eikebrokk et al (2004) discussed possible causes of the observed NOM increase in Northern European and Northern American water sources and the correspondent impacts on coagulation/contact filtration processes (Figure 4.8). From operation models derived from extensive pilot plant investigations the significant impacts on operational parameters like coagulant demand, sludge production, filter run length, etc. from increasing raw water NOM levels are quantified and discussed. To illustrate the consequences, an increase in raw water colour level from 20 to 35 mg Pt/L, increased the predicted coagulant demand, the sludge production, the number of backwashes per day, and the residual DOC by 64%, 64%, 87%, and 26%, respectively. In addition, treatment capacity and filter run length was decreased by 10% and 47%, respectively (Figure 4.9).

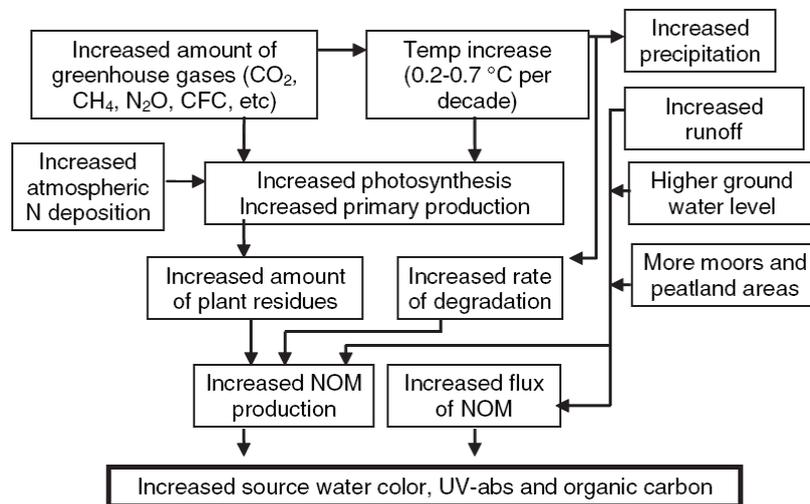


Figure 4.8. Possible reasons for increasing NOM in surface waters. Modified from Forsberg (1992) and Liltved (2002).

Korth et al (2004) reported an increase in NOM in some German drinking water reservoirs during the past 10-15 years, and that the impacts of such a change on the drinking water quality were almost unknown although this represents a serious concern for the water industry. A research project that was carried out at several drinking water reservoirs and water works indicate that the NOM increase is predominately caused by an increasing input to the reservoir of high and intermediate MW humic substances from the catchment area. During water treatment, the fractions of the high MW humic substances and polysaccharides were significantly reduced. In contrast, low removal efficiencies were detected for the low and intermediate MW fractions. In summary, the increased raw water NOM concentration resulted in a correspondent increase in the NOM concentration of the treated water. In addition, the concentration of biodegradable substances (BDOC) in treated water also increased with the raw water NOM content. No relationship was

found between treated water AOC and raw water NOM. However, AOC concentrations increased significantly during snowmelt and the circulation periods in the reservoir.

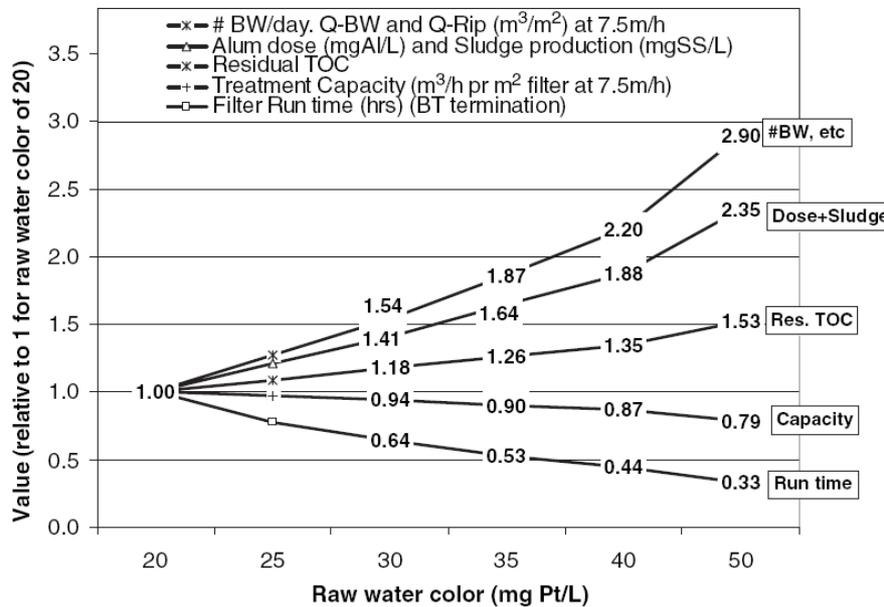


Figure 4.9. Impacts of increasing raw water NOM content (i.e. colour) on major coagulation-contact filtration operational characteristics. Alum coagulation, 7.5 m/hr filtration rate (Eikebrokk et al 2004).

4.8 The importance of mixing

Rapid mixing after coagulation is an important design parameter. The coagulant must be uniformly mixed with the raw water. In case mixing is poor local under- and overdosing occurs, resulting in poor performance of the process. The parameter expressing mixing intensity is called the velocity gradient or G-value (s^{-1}). Recommended G-value for rapid mixing is minimally $1500 s^{-1}$.

Mixing intensity and time has significant effect on the mechanisms (e.g. sweep coagulation, sedimentation) involved in the following process of coagulation (Chichuan et al 2002). Between the hydrolysis of the coagulant in water and the development of large flocs, short-lived water soluble aluminium and iron(III) hydroxide complexes, metal hydroxide sols are formed, which also carry a positive charge (Licisko et al 1996). These latter two metal hydroxide types (which also exist for a brief period - a few seconds - only) are the ones capable of destabilizing the dispersion and adsorbed NOM on their surfaces (see description of coagulation mechanisms in Chapter 7). The bond between the suspended solids to be removed and the metal hydroxide sols and water soluble metal hydroxide complexes must be established within this short period. Rapid mixing of the coagulant will ensure rapid hydrolysis of the coagulant, contact between the sols and the

suspended solids and will retard the development of large flocs which are inactive in destabilizing the dispersion and removing of NOM.

For enhanced coagulation - contact filtration, mixing may not be that important. Eikebrokk (2000) presented results obtained from enhanced coagulation-contact filtration pilot investigations (Figure 4.9). Three different mixers were used: 1) An on-line mixer (OLM) with very high intensity and turbulence level created in a narrow centre hole with a peripheral coagulant addition in the turbulent zone downstream, 2) An empty-pipe mixer (EPM) with very low mixing intensity, and 3) A more practically applicable design of mixer No. 1, with a larger diameter centre hole (OLM+). The mixers can be seen as high and low extremes with respect to mixing intensity and head-loss (i.e. the mixers OLM with 6 m head loss, and EPM with almost no head loss, respectively), and an intermediate, more practically applicable mixer with 0.1 m head loss (OLM+).

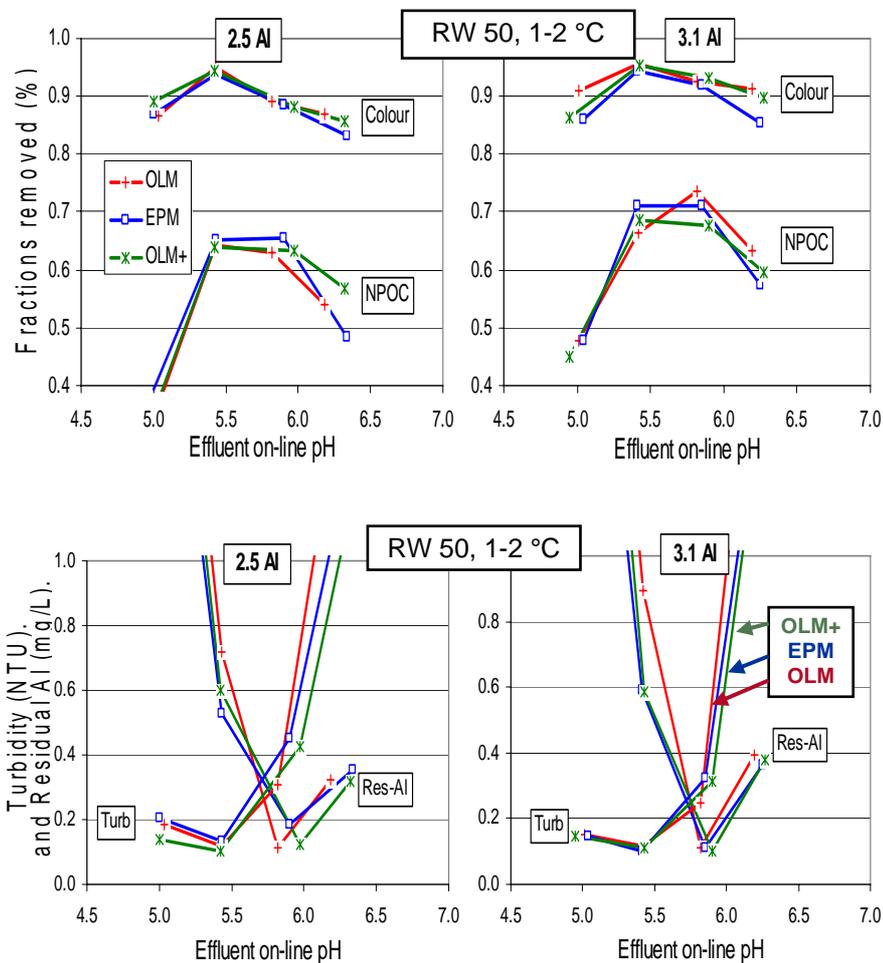


Figure 4.10. Enhanced coagulation-contact filtration performance in terms of colour and TOC (NPOC) removal, and residual turbidity and Al (lower) obtained with alum coagulation in two dosage levels (2.5 and 3.1 mg Al/L) at 1-2 °C, using three different mixers - OLM (extreme intensity), EPM (empty pipe), and OLM+ (intermediate intensity). Raw water quality: Turbidity < 0.2 NTU; 50 mg Pt/L; 5 mg DOC/L. (Eikebrokk, 2000).

From Figure 4.9 it appears that NOM removal efficiency in terms of colour and TOC (NPOC) is not influenced to any great extent by the mixing intensity, regardless of the applied coagulant dosage (2.5 or 3.1 mg Al/L, i.e. 0.50 or 0.62 mg Al/mg TOC). Only the residual coagulant concentration seems to be negatively affected at the lowest coagulant dosage. Because mixing is considered to be more critical the colder the water is, the results presented in Figure 4.10 were obtained with water that was chilled down to about 1 °C by means of a reverse heat pump. The differences in performance between the three mixers were surprisingly small, and possible explanations can be: 1) in contact filtration, the adsorption of NOM (colour and TOC) to aluminium hydroxide deposits in the filter bed is a more important coagulation mechanism than the more mixing-dependent pre-formation of filterable flocs, 2) unless the coagulant dosage is above a critical level, Al-containing colloids formed during coagulation are not efficiently removed during filtration.

4.9 Optimization procedures – Coagulation and filtration profiles

Based on the above results, Eikebrokk (2004b) suggested a systematic on-site approach to identify the “coagulation profile” for a specific raw water/water treatment plant (Figure 4.11). For a reasonable number of different coagulant dosage levels (normally 3-4), coagulation pH is varied in a step-wise manner within each dosage level, and filter effluent samples are analyzed after steady-state performance is established after every change in coagulation conditions. In this way not only the optimum pH and coagulant dosage can be identified, but also the interrelationship between dose and pH, e.g. a higher coagulant dosage normally widens the “pH-window” for optimum treatment performance.

Similarly, a “filtration profile” can be identified by running tests with different filtration rates, thereby identifying the effects of filtration rate - and thereby solids load rates - on filter run times determined by breakthrough or head loss (Figure 4.11). These tests should be performed under optimal coagulation conditions found during the coagulation profile identification tests described above. Coagulation and filtration profile identification tests should be performed during selected periods during the year, covering the range of seasonal variations in raw water quality. This will give operators very valuable information and insight into the optimum operation conditions of their water treatment facility, and how these optimal conditions are affected by variations in the raw water quality.

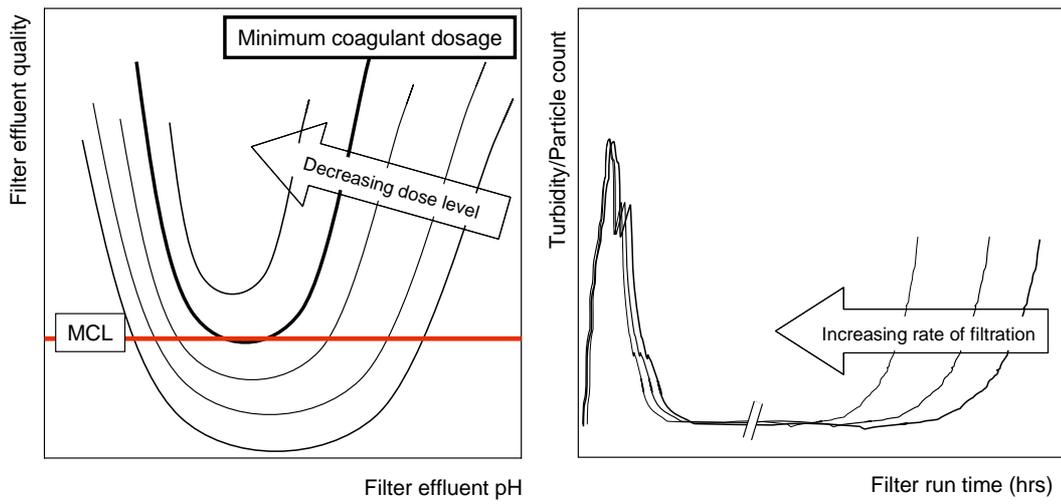


Figure 4.11 Proposed tests to identify enhanced coagulation and filtration profiles (Eikebrokk, Adelaide 2004b).

5 Pre- and post treatments

5.1 Ion exchange

In order to remove NOM over a broader range of molecular weight compounds, enhanced coagulation can be combined with other unit processes. The magnetic ion exchange (MIEX® DOC Process) was developed specifically for the removal of problematic NOM in drinking water production. This process can be used in various operating scenarios: Retrofit into conventional water treatment plants, and installation into new plants combined with microfiltration. MIEX and coagulation have been a very effective combination with respect to turbidity and NOM removal (Morran et al 2004).

Son et al (2005) studied NOM removal by enhanced coagulation and magnetic ion exchange (MIEX®) in bench scale experiments. For enhanced coagulation, the removal efficiencies of high molecular weight organic matter (>10 kDa) were much higher than those of low MW organic matter. With MIEX, however, the low MW fractions were removed more efficiently than the high MW fractions. High MW compounds were found to be responsible for UF membrane fouling.

5.2 Activated carbon adsorption

From a case study, Najm et al (1998) stated that the combination of enhanced coagulation and powdered activated carbon (PAC) could increase the removal of NOM and DBP precursors, and concluded that this process combination can be a cost-effective alternative to granular activated carbon (GAC) filtration and to enhanced coagulation alone.

Ho and Newcombe (2005) studied the effect of NOM, turbidity and floc size on powdered activated carbon (PAC) adsorption of the odour compound 2-methylisoborneol (MIB) during alum coagulation. They found that as the alum dose increased, the adsorption of MIB decreased. This was due to the size of the flocs, with larger flocs incorporating PAC into their structure, thus reducing the efficiency of mixing and the bulk diffusion kinetics for the MIB molecules. The presence of turbidity also reduced MIB adsorption due to formation of larger flocs. The character of NOM was found to have a greater influence on MIB adsorption than the floc structure.

5.3 Ozonation and oxidation

During the last 20 years it has become common practice for many water treatment plants to include prechlorination or preozonation steps in water treatment plants. It was assumed that oxidation can enhance removal of

NOM and turbidity during coagulation. During the 1980s, these coagulation effects were given the name microfloculation or ozone induced particle destabilization. However the mechanisms of ozone effect on coagulation are not fully understood. Some scientists and engineer still question whether there is any effect at all or it is an artefact. Two possible mechanisms of ozone effect on coagulation are proposed (in Rebhun and Lurie, 1993):

- oxidation of adsorbed organics, their hydrophilization and eventual desorption leading to destabilisation of the particles
- catalyzed polymerization of DOC and subsequent adsorption-bridging.

Pre-ozonation effects are dependent on many factors including properties of organic matter, pH, type of coagulant, and concentration of calcium in water. Schneider and Tobiason (2000) found that when alum was used as a coagulant, preozonation hindered turbidity and DOC removal. However, application of cationic polymers and preozonation lead to small but statistically significant increases in the removal of both ingredients. The authors hypothesize that preozonation reduces the surface charge of particulates, leading to better coagulation by charge neutralisation. Chandrakantha et al (1996) also related preozonation effects to DOC which is covering particles, beside they found that ozone-induced particle destabilization occurred only in the presence of calcium.

Ozone has been reported by some to improve coagulation and filtration efficiency (Reckhow et al 1993; Stolarik and Christie, 1997). However, others have found no improvement in filter effluent turbidity due to ozonation (Tobiason et al 1992; Hildebrand et al 1986). Prendiville (1986) collected data from a large water treatment plant showing that pre-ozonation was more effective than pre-chlorination to reduce filter effluent turbidities.

Becker and O'Melia (2001) provided an overview of the effects of ozone on coagulation and filtration processes. Although ozone has many benefits, its expense is significant and its placement in the treatment train should be chosen with a sound understanding of its effects on other unit processes. They stated that the effect of ozone on coagulation is shown to be dependent on the coagulant type and on the water quality characteristic that is setting the optimum coagulant dose. For waters with moderate to high DOC levels, the coagulant dose is set by the DOC. Ozonation converts DOC into smaller, more oxygenated compounds that exert a greater metal coagulant demand than the parent compounds. In this case, ozonation can lead to an increase in the optimal coagulant doses (Edwards and Benjamin, 1992). For low DOC waters, the coagulant dose is set by the particle and the adsorbed organic matter. Ozone may react with the adsorbed DOC and alter the amount and conformation of the adsorbed organics, which can lead to a decrease in coagulant demand.

Bekbolet et al (2005) treated water samples of different origins in Turkey and Italy by coagulation, ozonation, and coagulation followed by ozonation and

photocatalysis. NOM removal efficiencies and DBP formation potentials were compared for the different treatments. The removal efficiencies and the distribution of individual THMs and HAAs were found to be NOM-site specific. Other DBPs were also detected and chloral hydrate ($C_2H_3Cl_3O_2$) was found in significant amounts. The responsible precursor sites could only be reduced by photocatalytic treatment of NOM.

Wei and Yong-Mei (2004) stated that NOM have a very strong influence on the stability of inorganic particles through adsorption, coating, etc., thus making surface water with high NOM concentrations difficult to treat efficiently by coagulation. Jar tests and pilot investigations were used to evaluate the effectiveness of ferrate pre-oxidation in enhancing the coagulation of NOM-rich lake water. A substantial reduction in residual turbidity after sedimentation and filtration was obtained by ferrate pre-oxidation at dose levels of 1-5 mg/L as K_2FeO_4 . A similar improvement of colour, DOC, iron, manganese, and total bacteria was observed. It was suggested that simple addition of ferrate prior to the coagulation step can reduce the coagulant dose demand in order to maintain acceptable residual turbidity. It was believed that additional coagulant (Fe (III)) was formed as a result of ferrate decomposition.

Chandrakanth and Amy (1996) studied the effects of ozone on the colloidal stability and aggregation of particles coated with NOM. They found that ozone-induced particle destabilization occurred only in the presence of calcium and it was proposed that the destabilizing effect was due to a reduction in particle stability through surface charge reduction.

Edwards and Benjamin (1992) investigated the effects of ozone on several water quality parameters that affect particle behaviour in water treatment systems. They found that ozone did not decrease critical coagulant concentration for $AlCl_3$, $FeCl_3$, or alum coagulants; destabilize particles; or improve particle removal processes if pH was held constant. There was no evidence of disruption or desorption of organic coatings of particles that enhanced particle destabilization. In sum, all ozone-induced particle destabilization phenomena observed were primarily the result of a decrease in pH, an increase in pH, or precipitation of $CaCO_3$. Because these changes are produced more efficiently and at a lower cost with chemical addition, aeration, or both, they must be properly considered when evaluating the true benefits of ozonation with respect to particle removal.

Murray and Parsons (2004) studied the Fenton and photo-Fenton oxidative processes to support conventional coagulation of raw waters with high NOM levels. The performance of both processes was shown to dependent on pH, $Fe:H_2O_2$ ratio as well as Fe^{2+} dose. Under optimum conditions both processes achieved greater than 90% removal of DOC and UV254 absorbance, and were able to reduce the THM formation potential from 140 to less than 10 $\mu g/L$, which is well below UK and US standards.

Hozalski et al (1995) used laboratory-scale biologically active sand filters to study the effects of NOM source and empty bed contact time (EBCT) on the removal of TOC from coagulated and ozonated NOM solutions (2-4 mg O₃/mg TOC). They found that TOC removal in the biofilter ranged from 16 to 33%, and that TOC removal was significantly affected by the NOM source but was independent of EBCT in the range of 4 to 20 min. The contribution of biofiltration to TOC removal was relatively small compared with removals across the entire coagulation and ozonation-biofiltration treatment train. In one biofiltration experiment, however, removal of the biodegradable fraction of TOC was 100%, suggesting that biofilters may be effective in reducing subsequent regrowth in distribution systems.

Urfer et al (1999) investigated the use of enhanced coagulation of a postsedimentation ozonation system at the Mannheim WTP in Ontario, Canada. For the specific water tested, model predictions indicated that operation of the plant under enhanced coagulation conditions could substantially improve the disinfection capacity of the ozone system, compared with conventional coagulation.

5.4 Coagulation as pretreatment to UF or MF

Hankins and Price (2005) stated that the combination of microfiltration (MF) or ultrafiltration (UF) with upstream coagulant dosing is beneficial and synergistic for NOM laden waters. This process benefits membrane flux by reducing the penetration of foulants into the membrane pores, and also by the formation of filter cakes on the membrane surface.

5.5 Interactions with softening or corrosion control processes

Jegatheesan et al (2002) studied the influence of NOM on coagulation and flocculation processes in order to optimize process conditions. They tested pretreated pond water and synthetic water containing humic acids as the source waters. They found a strong relationship between optimum alum dose and source water TOC, and that the high MW organic substances had higher removal affinity in the coagulation process. Divalent cations such as Ca²⁺ and Mg²⁺ had no significant effect on the removal of NOM. The presence of clay however, increased the removal of NOM. The removal of TOC obtained from coagulation of the pretreated natural pond water containing non-homogeneous organic matter was significantly lower (60%) than that obtained under the same conditions with the synthetic raw water containing homogeneous humic acid only (80%).

Gregor et al (1997) studied optimization of NOM removal from low turbidity waters by controlled pH adjustment of alum coagulation. They provided guidelines on how pH should be controlled to maximize NOM removal while at the same time making full use of pH-influencing chemicals such as acids, metal coagulants and lime. For low turbidity waters, essential floc nucleating sites can be provided by the lime that is used for pH and corrosion control,

provided that the lime is added in sufficient quantity and at a point where it retains some of its particulate nature, i.e. immediately after coagulant addition. Adjusting the pH down to between 4 and 5 prior to alum coagulant addition allows the formation of soluble NOM-aluminium complexes that link to each other, thereby forming large insoluble bridged complexes (micro-flocs) that also act as nuclei for macro-floc development (flocculation).

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/or automatically. The automatic process control requires instrumentation including water quality sensors that 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 rely mostly on manual operation routines.

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

6.1 Principles of automation and control

6.1.1 Automated control system components

The basic principles of advanced automated control systems are shown in Figure 6.1. Depending on the objective of the automation (e.g. flow control, quality control), different sensors (e.g. flow meters, turbidity meters) 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, dosage systems) which adjust the operating conditions of the plant towards the optimal operation conditions (set points).

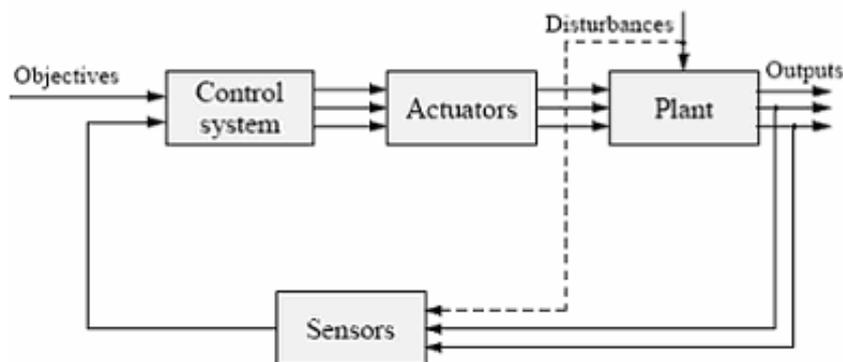


Figure 6.1 Basic principle of an automated control system.

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.

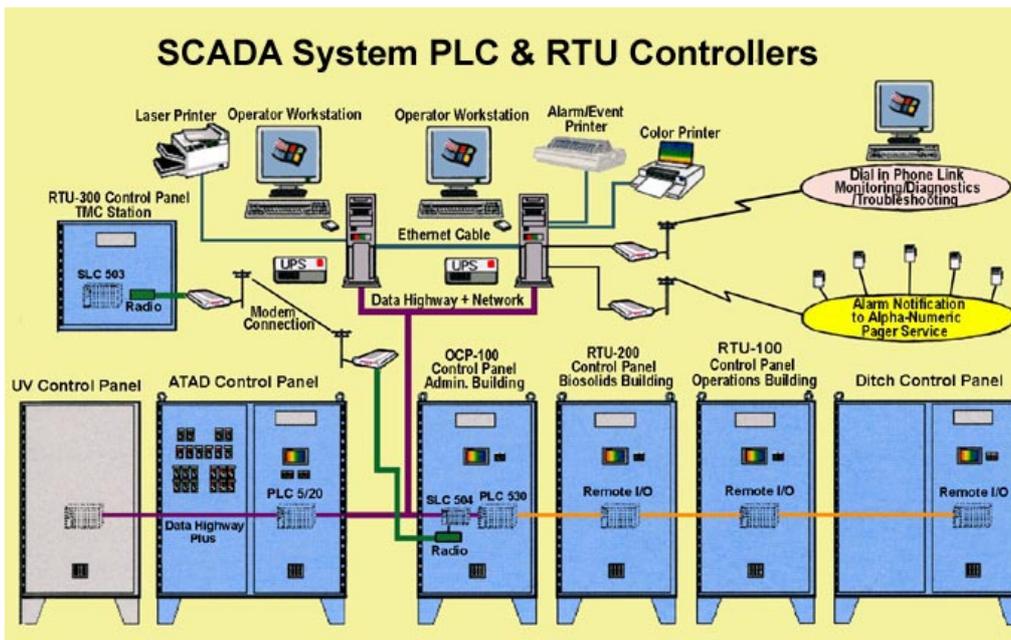


Figure 6.2. Automated control system scheme (www.princeton-indiana.com)

6.1.2 Control types and algorithms

There are three types of automatic controls that are commonly implemented at 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 Enhanced coagulation process control

An enhanced coagulation process control system is a part of an overall water treatment process control system, however in this report it is described separately. The most commonly used monitoring parameters (with sensors) and controlled parameters (with actuators) are given in Table 6.1.

Logsdon (2000) states that careful control of coagulation chemistry and of filtration rate increases, continuous monitoring of filtered water turbidity, and proper management of backwash water are keys to successful filtration. Attaining the proper coagulant dose and pH is crucial to process optimisation. Some treatment plants use jar tests frequently, and others use pilot filters. The use of streaming current monitors as a guide to controlling coagulant dosage has become more common, especially in the United States at plants where raw water quality can change rapidly. In-line turbidity and particle monitoring at each filter is common practice in more and more countries.

Table 6.1 Common monitored and controlled items in enhance coagulation processes

	Mixing, coagulation, flocculation	Clarification, sedimentation, filtration
Monitored parameters (with sensors)	Water level, flow, temp., turbidity, pH, UV-abs, colour, TOC, mixer status	Water/sludge levels, flows, turbidity, UV-abs, colour, TOC, disinfectant and coagulant residuals, valve status, head loss, head loss distribution
Controlled parameters (with actuators)	Chemicals feed, flows, pumps, blowers, valves	Water/sludge levels and flows, chemicals feed (e.g. polymers), blowers, pumps, valves, run times

Coagulants and pH adjusting chemicals (acid/base) are generally fed by positive displacement chemical metering pumps, using flow feedforward control. Coagulant dosing is sometimes automated using feedforward control for turbidity and UV-abs (surrogate for TOC) measurements in combination of with feedback control for the pH value (Fig. 6.3). Depending on the UV and turbidity the algorithm integrated in the PLC estimates the optimal dose of coagulant and acid/base (feedforward control). The turbidity can be measured at the outflow at the plant and dosing is adjusted to obtain optimal TOC and turbidity removal. PID type of controls is normally used to compensate the lag time for the reactions/pH changes following acid or base dosing.

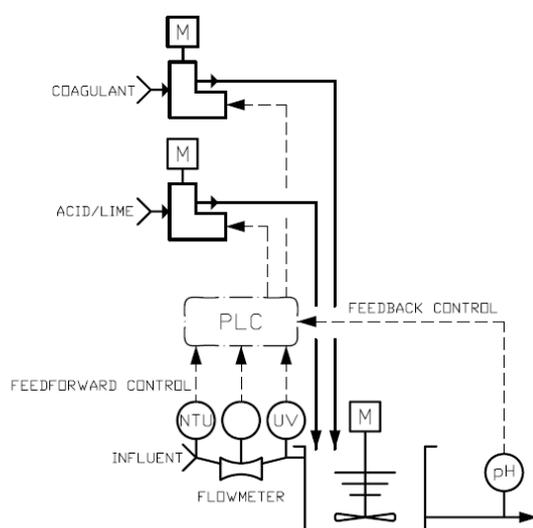


Figure 6. 3. Process control principles of coagulation

6.3 Controls in lab-scale

Coagulant dosage is traditionally determined according to results from jar-tests or operator's experience, which often lead to coagulant overdosing or insufficient dosing resulting in reduced water treatment performance and increased treatment costs. In order to monitor the state of aggregation of coagulation/flocculation and to explore the effects of turbidity, pH and rapid mix intensity on coagulation performance, Kan and Huang (1998) used a fibre optical monitor, i.e. a photometric dispersion analyzer (PDA) first introduced by Gregory and Nelson (1984) and further described by Gregory (1985). The monitoring principle is based on measurements of fluctuations in the intensity of light transmitted through a flowing suspension as recorded by a detector. It was shown in plant operations that the PDA provided an effective index for the floc size and settling velocity, and that the PDA-determined coagulant dosage (PACI) was significantly lower than the daily operating dosage at various raw water turbidities (2.2-125 NTU), with average dosages of 2.0 mg/L and 2.4 mg/l as Al_2O_3 , respectively. Settled water turbidity and sludge production was reduced as a result of the PDA application.

Ebie et al (2006) studied the importance of optimized mixing intensity and time in order to decrease head loss and treated water turbidity in direct filtration of low turbidity source waters. They employed a batch type coagulation experiment apparatus (Figure 6.4) where the particle concentration was measured continuously after injection of coagulant in order to clarify the fundamental coagulation and microfloc formation dynamics.

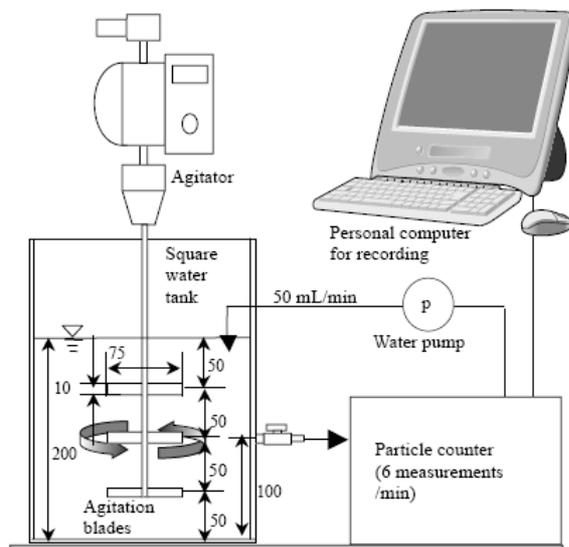


Figure 6.4. Schematic drawing of the batch-type coagulation experimental apparatus (measures in mm). Ebie et al (2006).

7 Process modelling

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, because with their help parameters of the treatment train (e.g. filter area and 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 coagulant doses, filter loading rates, backwashing conditions and frequency, etc. to optimal levels. Models can also be applied to improve automation and process control systems because they allow predictions and use of computers to simulate different patterns of control actions to find the most appropriate one. Up-to-date, however, 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. Empirical models are simpler and often obtained by fitting mathematical equations to a set of experimental data ("black box" approach).

7.1 Modelling of DOC removal

The removal of natural organic matter (NOM) by coagulation is impacted by a number of factors, e.g. NOM character and concentration, turbidity and alkalinity, other organic as well as inorganic constituents of the raw water. High levels of variation can occur in a range of water quality parameters such as turbidity, alkalinity, colour, NOM, algae and micro-organisms.

Mathematical models that relate the character and concentration of dissolved organic matter in the raw water to inorganic coagulant dosing that maximize the removal of DOC have been developed. The models can also be used to predict the required coagulant dosage when treating raw waters of different quality. Van Leeuwen et al (2005) used models to predict alum coagulant dosage that were subsequently applied to treat two Australian raw waters in jar tests and in pilot plant trials. DOC removals of 50-60% were obtained with application of the model predicted alum doses for maximizing DOC removal when coagulation was performed at pH 6. Much higher coagulant doses at similar pH resulted in comparatively minor additional DOC removal. THMFP was found to be proportional to the residual DOC and appeared to be linearly related. Formation of individual THMs was consistent in each water source but different between the two sources.

Harrington et al (1991) developed an interactive, user-friendly computer program to simulate inorganic water quality changes, THM formation, disinfectant decay and removal of NOM in water treatment processes. Furthermore, they discussed the selection, development and verification of empirical models to include in the program.

Tseng and Edwards (1999) presented a Langmuir model for prediction of full-scale removal of TOC during enhanced coagulation. Case studies of 27 full-scale utilities showed accurate prediction of TOC removal by coagulation at a range of utilities using alum, ferric, or poly aluminium chloride coagulants.

Edwards (1997) predicted the concentration of DOC remaining after enhanced coagulation with a standard error of about 10% or 0.4 mg/L. Model inputs were coagulant dosage, coagulation pH, raw water UV₂₅₄-abs, and raw water DOC. When calibrated to a specific site, the standard predictive error could be improved to 4% or 0.27 mg DOC/L. Performance differences between equimolar dosages of alum and ferric coagulants were attributed to: equal or better removal of DOC using ferric at very high coagulant dosages; equal or better removal of DOC using alum at lower coagulant doses, or differing acidity of coagulants, producing a performance advantage for the more acidic coagulant.

7.2 Mechanisms for DOC removal used in the models

In order to develop mechanistic models, an understating of the mechanisms involved in DOC removal is important. The three mechanisms most commonly referred to as important for DOC removal during chemical coagulation are charge neutralization, entrapment, and adsorption.

Charge neutralization is the mechanism used to explain the removal of NOM in operational regions where aluminium hydroxide precipitation is minimal. Cationic aluminum species interact with anionic NOM to form insoluble charge-neutral products. A linear increase in the required cation concentration (e.g. Al, Fe) with anion concentration is expected.

In operational regions where aluminum hydroxide precipitates are formed, NOM can be removed by entrapment (sweep coagulation) in the hydroxide or adsorption to hydroxide surfaces. The concentration of coagulant has to be high to cause rapid precipitation of Al (OH)₃. Colloids, including colloidal NOM can act as nuclei for precipitate formation, and can become entrapped during floc aggregation.

These mechanisms apply mainly for removal of colloidal NOM, typically higher molecular weight HA. These acids generally have low charge densities and therefore require low coagulant doses to induce destabilization. However, the more soluble fractions of NOM (fulvic acids) have higher anionic charge densities that facilitate their dissolution. The sweep coagulation which operates most effectively on colloidal NOM, is unlikely to

be effective for these soluble FA. Charge neutralization may remove soluble FA, but high doses of coagulant will be required to neutralize the high anionic charge, and the high coagulant dose required by soluble FA is likely to correspond to overdosing of humic acid colloids resulting in restabilization of the colloids.

A fourth mechanism that is occasionally alluded to is the chemical interaction of soluble NOM with soluble coagulant metal ions such as aluminium. The metal cation and the chemically bound (complexed) NOM remains in solution until either the binding capacity of NOM has been satisfied, or the solubility of the metal-NOM complex is exceeded. The complex does not need to be charge-neutral to precipitate.

Most of the models developed for enhanced coagulation (e.g. by Edwards (1997)) takes into account only adsorption mechanisms. At acidic pH (used for enhanced coagulation) aluminum species are positively charged, therefore negatively charged organic matter is effectively adsorbed by electrostatic interactions. Other assumptions included in the model are as follows:

- part of the DOC consists of non-sorbable fractions that are not removed by coagulation
- the adsorption capacity of flocs is a function of pH
- the characteristics of DOC do not change with pH
- adsorption can be described with a Langmuir isotherm

7.3 Predicting DOC removal

A model for predicting the DOC concentration remaining after enhanced coagulation was developed by Edwards (1997). This model takes into account only adsorption mechanisms, described by a Langmuir isotherm. The model uses only parameters that are routinely measured, and it can readily be applied at water treatment plants.

The fraction of non-sorbable DOC that is not removed by coagulation is calculated by:

$$\text{Fraction non-sorbable DOC} = K_1 (\text{SUVA})_{\text{raw water}} + K_2 \quad (7.1)$$

where K_1 and K_2 are empirical fitting constants.

The sorbable DOC can then be calculated by:

$$\text{Sorbable DOC} = \{1 - \text{fraction non-sorbable DOC}\} \text{DOC}_{\text{initial}} \quad (7.2)$$

The model is based on the Langmuir equation:

$$x / M = (a b [C]_{eq}) / (1 + b [C]_{eq}) \quad (7.3)$$

where x is DOC removed (mg/L DOC), M is coagulant added and metal hydroxide formed (mmole/L), C_{eq} is sorbable DOC in solution at equilibrium, and a and b are sorption constants. The constant a can be determined by:

$$a = x_3 \text{pH}^3 + x_2 \text{pH}^2 + x_1 \text{pH} \quad (7.4)$$

where x_1 , x_2 and x_3 are fitting constants. Combining these equations gives:

$$\frac{\{(1 - \text{SUVA } K_1 - K_2) \text{DOC}_{\text{initial}} - [C]_{\text{eq}}\} / M}{\{x_3 \text{pH}^3 + x_2 \text{pH}^2 + x_1 \text{pH}\} b [C]_{\text{eq}} / \{1 + b [C]_{\text{eq}}\}} \quad (7.5)$$

This equation can be solved in an Excel sheet if the six empirical constants (K_1 , K_2 , x_1 , x_2 , x_3 , b) are known. The DOC concentration remaining after coagulation is then:

$$\text{DOC (mg/L)} = C_{eq} \text{ (mg/L)} + \text{non-sorbable DOC (mg/L)} \quad (7.6)$$

Edwards (1997) also determined the values for the empirical constants, still keeping the standard error below 10%. These general values for the empirical constants are listed in Table 7.1 below.

Table 7.1 Parameter values used for the model for predicting DOC removal (Edwards, 1997).

Parameter	Fe coagulant	Al coagulant	General
			DOC < 10 mg/L
Standard error, %	9.3	9.5	10
x_3	4.96	4.91	6.44
x_2	- 73.9	- 74.2	- 99.2
x_1	280	284	387
K_1	- 0.028	- 0.075	- 0.053
K_2	0.23	0.56	0.54
b	0.068	0.147	0.107

Several other empirical models predicting DOC or TOC removal by coagulation can be found in the literature. Some of these are summarized in Table 7.2. However, according to Tseng and Edwards (1999) the accuracy of these models is lower than the Langmuir adsorption model described above.

Table 7.2 Models developed to predict DOC or TOC removal during coagulation.

Coag	Equation	Reference
Al a)	$TOC_{final} = 0.405TOC_{raw} - 0.55dose + 0.688pH_{coag} - 3.32$	Moomaw et al (1992)
Fe a)	$TOC_{final} = 0.52TOC_{raw} - 1.49dose + 0.740pH_{coag} - 3.50$	Moomaw et al (1992)
Al b)	$TOC_{final} = 1.076[TOC_{raw}]^{0.923}[dose]^{-0.298}[Alk_{raw}]^{0.173}$	Zhu (1995)
Fe b)	$TOC_{final} = 0.839[TOC_{raw}]^{1.094}[dose]^{-0.355}[Alk_{raw}]^{0.150}$	Zhu (1995)
Al c)	$\ln(TOC_{final}) = -0.33 + 1.31 \ln(TOC_{raw}) - 0.54 \ln(dose) - 0.08 \ln(TOC_{raw})\ln(dose) + 0.077 pH_{coag} \ln(dose)$	Harrington et al (1992)
Fe c)	$\ln(TOC_{final}) = -0.16 + 1.25 \ln(TOC_{raw}) - 0.71 \ln(dose) - 0.05 \ln(TOC_{raw})\ln(dose) + 0.092 pH_{coag} \ln(dose)$	Harrington et al (1992)
Al or Fe d)	$TOC = a + b (2.717)^{-c(dose)}$	Chowdhury and Owen (1996)
a) dose in meq/L Al or Fe; b) alk. in mg/L CaCO ₃ and dose in mg/L as Al ₂ (SO ₄) ₃ · 18H ₂ O or FeCl ₃ · 6H ₂ O c) dose in mg/L as Al ₂ (SO ₄) ₃ · 18H ₂ O or FeCl ₃ · 6H ₂ O d) a, b and c are different empirical constants depending on the TOC, alkalinity and the coagulant used		

7.4 Empirical models and guidelines for optimum operation

Several empirical model equations have been published to predict the required coagulant dose for removal of TOC. Some of these are summarized in Table 7.3. Empiric models have also been developed to predict sludge production, filter run times, etc.

Table 7.3 Model equations for coagulant dosage requirements

Models	Equations	References
Enhanced coagulation alum dose	$Al = 10.9 + 2.63 (TOC_0) + 0.17 (Alk) + 0.74 (pH)$	in Rodrigueaz <i>at al</i> (2000)
Enhanced coagulation alum dose ¹⁾	$Al_{min} = 0.043 \cdot Color + 0.30$ $Al_{pract} = 0.054 \cdot Color + 0.37$	Eikebrokk (2004b)
Enhanced coagulation ferric dose ¹⁾	$Fe_{min} = 0.107 \cdot Color + 0.58$ $Fe_{pract} = 0.134 \cdot Color + 0.72$	Eikebrokk (2004b)
Alum dose depending on water color of water	$Al = 4 \times Colour^{-1/2}$	Soviet Building Norms (1984)
TOC ₀ : concentration of TOC before treatment (mg/l), Al : coagulation dose (mg/l), Alk: water alkalinity (mg/l CaCO ₃), 1) Absolute and practical minimum dosages determined by the residual coagulant concentration standard (< 0.15 mg Me/L).		

Based on numerous pilot plant experiments using 3 model waters with low turbidity and increasing NOM and SUVA levels, Eikebrokk (2004b) developed several simple operational models for the enhanced coagulation – contact filtration process. The models presented below were developed for water with color 15 – 50 mg Pt/L, TOC 2 - 6 mg/L, turbidity < 0,5 NTU and SUVA 3,8 – 4,8 using 2M anthracite-sand filters. The models describe required coagulant dose, sludge production and filter run time to break through, which in turn can be used to determine net water production and backwash water production for various operational conditions.

7.4.1 Required coagulant dosage

Minimum required coagulant dose:

$$\text{Dose (mg Me/L)} = A \cdot \text{Raw water Color (mg Pt/L)} + B \quad (7.7)$$

where A and B are constants depending on the coagulant and operational conditions.

It is also defined a practical minimum coagulant dose which is 25 % higher than the absolute minimum dose determined from the above equation. For granulated aluminium sulphate as a coagulant the absolute minimum and the practical minimum coagulant doses at optimum coagulation pH are given by:

$$\text{Al}_{\min} = 0.043 \cdot \text{Color} + 0.30 \quad (7.8)$$

$$\text{Al}_{\text{pract}} = 0.054 \cdot \text{Color} + 0.37 \quad (7.9)$$

For ferric chloride as a coagulant these doses are given by:

$$\text{Fe}_{\min} = 0.107 \cdot \text{Color} + 0.58 \quad (7.10)$$

$$\text{Fe}_{\text{pract}} = 0.134 \cdot \text{Color} + 0.72 \quad (7.11)$$

7.4.2 Sludge production

Sludge production at optimum coagulation:

$$\text{SS(mg/L)} = \text{SS}_{\text{RW}} + k \cdot \text{Dose} \quad (7.12)$$

where SS_{RW} is the suspended solid concentration in the raw water, k is a constant depending on the type of coagulant and Dose is the coagulant dose (mg Me/L).

For granulated aluminium sulphate and ferric chloride as coagulant, respectively, the sludge production (mg SS/L) is then given by:

$$\text{SS}_{\text{Al}} = \text{SS}_{\text{RW}} + 4.2 \cdot \text{Dose} \quad (7.13)$$

$$SS_{Fe} = SS_{RW} + 2.5 \cdot \text{Dose} \quad (7.14)$$

7.4.3 Filter run time to break through

Filter run time to breakthrough at optimum coagulation:

$$t_{BT} = a (v_f \cdot SS)^b \quad (7.18)$$

where t_{TB} is time of filtration until breakthrough (hrs), v_f is rate of filtration (m/hr), SS is the suspended solids concentration in coagulated water, i.e. sludge production (mg SS/L), and a and b are constants specific to the filter and coagulant.

For alum without any polymer as filter aid and $SS_{RW} = 0$, the time of filtration is:

$$t_{TB} = 298 (v_f \cdot \text{Dose}_{Al})^{-1.29} \quad (7.19)$$

7.4.4 Net water production

The net water production would depend on the operational mode of the process. If ripening water is used as clean water, and the filter is backwashed with raw water, the net water production is:

$$Q_{Net} = [24 / t_p] [v_f \cdot t_f] \quad (7.20)$$

If ripening water is used as clean water, and the filter is backwashed with clean water, the net water production is:

$$Q_{Net} = [24 / t_p] [v_f \cdot t_f - v_s \cdot t_s] \quad (7.21)$$

If ripening water is wasted, and the filter is backwashed with raw water, the net water production is:

$$Q_{Net} = [24 / t_p] [v_f \cdot (t_f - t_r)] \quad (7.22)$$

Finally, if ripening water is wasted, and the filter is backwashed with clean water, the net water production is:

$$Q_{Net} = [24 / t_p] [v_f \cdot (t_f - t_r) - v_s \cdot t_s] \quad (7.23)$$

Where Q_{Net} is the daily water production (m^3/m^2 day), v_f and v_s are the rates of filtration and backwash (m/hr), respectively, t_f , t_s , and t_r is the time of filtration, backwash and ripening (hrs), respectively, and t_p is the total time between each backwash ($t_f + t_s +$ time for valve operation, etc) in hrs.

7.4.5 Backwash water production

As for net water production, the production of backwash water is given by:

$$Q_{BW} = [24 / t_p] [v_s \cdot t_s] \quad (7.24)$$

where Q_{BW} is the daily production of backwash water.

By combining these and similar simple empirical operational models it should be possible to develop powerful tools and software for operation control, optimization and automation. However, the models have to be further developed by i.e.:

- Verify and make the models applicable for other raw water qualities, operational conditions, process configurations, etc.
- Extend the models to include other coagulants, polymer addition, etc
- Investigate the effect of polymer addition and why the polymer is very efficient in some cases and far less effective in other cases.

8 Treated water characteristics

Enhanced coagulation is employed in order to obtain a more efficient removal of TOC without compromising removal efficiency of turbidity, pathogens, etc. Removal of TOC from drinking water is also necessary to provide a stable disinfectant residual and to minimize formation of disinfection by-products (DBPs) within the distribution network that have free chlorine residuals. High TOC in drinking water may also increase taste and odour problems and could be linked to higher BOM levels which in turn may promote growth of bacteria, including opportunistic pathogens.

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 NOM) 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 maximum concentration limit 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 not very precise term “*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 - DBP

One of the reasons for the TOC removal requirements is the need for reduction of the 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 as main precursors of DBPs. Enhanced coagulation has been evaluated as best available technology for the removal of TOC/THM precursors by the USEPA.

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 Examples of mathematical models used for prediction of chlorination by-products formation

Model	Reference
$HHA5 = 4.8 \times 10^4 [OH^-]^{0.35} (C_o[1-\exp\{-kt\}])^{0.43} (UV_{254})^{0.34}$	Sung <i>et al</i> (2000)
$TTHMs = a [OH^-]^j (C_o[1-\exp\{-kt\}])^m (UV_{254})^n (\text{algae})^p$	Sung <i>et al</i> (2000)
$TTHMs = b (\text{DOC})^{b1} (\text{Cl}_2)^{b2} (\text{Br}^-)^{b3} (\text{temperature})^{b4} (\text{pH})^{b5} (\text{Time})^{b6}$	Westerhoff <i>et al</i> (2000)

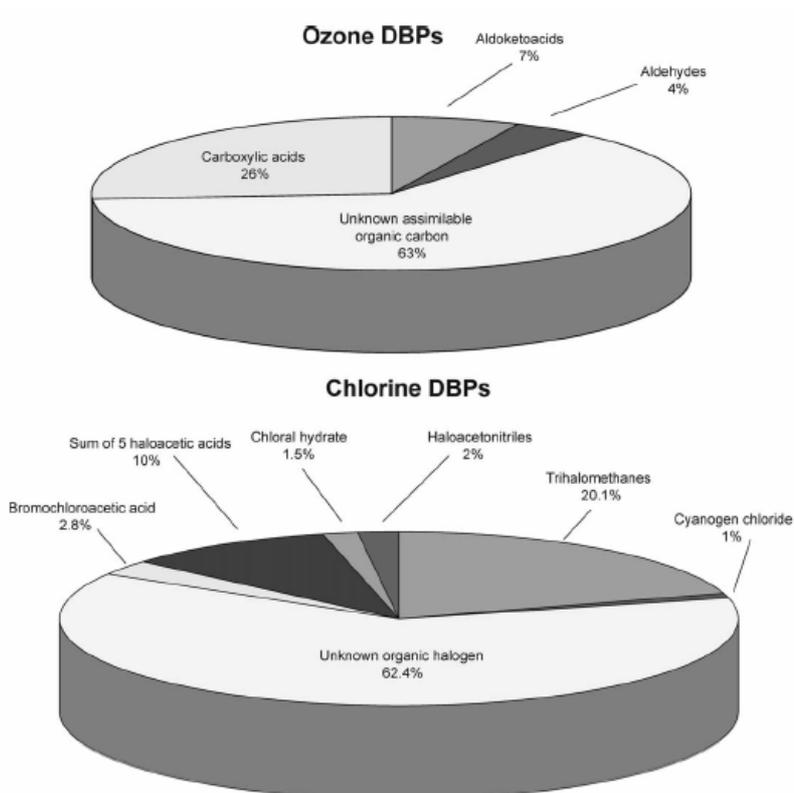


Figure 8.1. a) Relative amounts of ozone DBPs in drinking water as a proportion of the total assimilable organic carbon (AOC) from an ozone demonstration plant. b) Relative amounts of halogenated DBPs as a proportion of total organic halogen (TOX) in chlorinated drinking water from a representative chlorination plant (source: Richardson 2003)

8.3 BOM in treated water

Efficient removal of BOM (AOC or BDOC) is important to provide biostability of drinking waters. Several studies have been carried out to check the efficiency of enhanced coagulation and conventional coagulation with respect to BOM removal.

Volk *et al* 2000 showed that using enhanced coagulation instead of conventional coagulation, the removal of DOC could be improved from 29% (plant conditions termed baseline coagulation) to an average of 43%. Similarly, removal of BDOC was improved from the baseline level of 30% to 38% through the application of optimized coagulation. At lower pH, the ferric coagulants generally performed better with respect to organic carbon removal than the alum or poly aluminum chloride coagulants. In this case, the assimilable organic carbon (AOC) was not affected by coagulation, probably because the AOC fractions were composed of small molecular weight, non-humic compounds that were not amenable to coagulation.

Eikebrokk and Saltnes (2002) reported however from pilot-scale enhanced coagulation-filtration trials that BDOC was removed more effectively than TOC during contact filtration (both exceeding 50 %).

Since BOM can be released from algae during water disinfection, it is important to remove algae (picophytoplankton) before the disinfection step. The removal efficiency of picophytoplankton in conventional drinking water treatment processes, and the contribution of picophytoplankton to AOC were investigated by Okuda *et al* (2006). The removal ratio during coagulation-sedimentation was determined by jar tests using PAC (poly-aluminium chloride). Lower coagulation pH showed better picophytoplankton removal in coagulation-sedimentation. The optimum coagulant dosage for picophytoplankton removal was twice or more compared to that for turbidity removal. The removal efficiency of picophytoplankton was 44-60% at the lowest pH allowed in the water quality standard (5.8), and at an optimum coagulant dosage for turbidity.

The removal of picophytoplankton in rapid sand filtration was determined by pilot scale column experiments with sand and anthracite. The average removal was 16.3% without PAC addition and chlorination before sand filtration; on the other hand it was 51.5% with PAC and chlorination. The increase in AOC as a result of chlorination of picoplankton including 6800 cells/L of picophytoplankton, was 21 µg acetate-C/L (at 0.1 mg/L of residual chlorine). The AOC increased with increasing residual chlorine concentrations, and leveled off at 0.3 mg Cl/L. From the results, the AOC originating from picoplankton (maximum AOC from picophytoplankton) could increase up to 155 µg acetate-C/L in this reservoir. This study indicates that the removal of picoplankton (picophytoplankton) in drinking water treatment processes is important from the viewpoint of AOC control.

In general, the removal of BOM by enhanced coagulation is likely to show significant variation according to operating conditions and NOM composition. If bacterial regrowth is a problem, combining enhanced coagulation with other processes aimed at removing the lower molecular weight hydrophilic fractions may be an interesting option (e.g. MIEX, GAC, intermediate ozonation).

8.4 Microorganisms and turbidity in treated water

In general, enhanced coagulation is an effective multi-purpose treatment process with respect to NOM, turbidity/particles, and microorganisms including protozoa. Process optimization efforts should also identify possible conflicting optimum operating conditions for the different target substances. Vrijenhoek *et al* (1998) have shown that by adjusting pH and alum dose it is possible to obtain synergetic effects and efficiently remove both NOM and turbidity (<0.1 NTU). The pH should be kept relatively low to allow adsorption of NOM on surfaces of alum hydroxides, while alum dose should be high (20-60 mg/l) to allow sweep coagulation for turbidity removal.

There are numerous investigations showing that protozoan parasites like *Giardia* and *Cryptosporidium* can be efficiently removed from water by enhanced coagulation processes, without compromising the removal of NOM and turbidity/particles. However, in most cases success or not is a matter of treatment process optimization.

9 Case studies

9.1 Rusko WTP, Finland

Figure 9.1 shows the process treatment train of Rusko WPT (Jokela et al 2006) treating a lake water with high content of NOM (2005 average values of raw and treated water turbidity, colour and TOC are 2.4 and 0.11 NTU; 17 and < 2.5 mg Pt/L; and 5.8 and 2.0 mg TOC/L, respectively). Treatment capacity is 45 000 m³/day, and the treatment comprises enhanced coagulation with ferric sulphate (7.6 mg Fe/L as PIX), hydraulic mixing/flocculation, three dissolved air flotation (DAF) units, six sand filtration and eight GAC filter units. Lime and CO₂ is used for integrated pH- and corrosion control, and ClO₂ and Cl₂ is used for disinfection.

The average hydraulic load on the DAF unit is very high, 15-20 m/hr, which is increased up to 27 m/h when one of the three DAF-units is under cleaning. Each of the DAF units is typically cleaned for about 6 hours three times a year. The DAF units use a hydraulic sludge removal system (flooding), removing sludge for 5 minutes every 5 hours from each unit. DAF treated water typically have 1.0-1.5 NTU of turbidity, about 3 mg TOC/L, and about 1.5 mg residual Fe/L.

The six sand filters (1 m of sand depth) are also operated with a high surface load, 10 m/hr on average during normal operation and 12 m/hr during backwash of one of the units. Each filter units is backwashed with air and water for 20 minutes approximately every 24 hours, on the basis of head loss measurements. As a result of the 20% increase in loading to the remaining filters during backwash, an increase in filtered water turbidity can be detected.

The main purpose of the GAC filters is taste and odour control. Each of the 1.4 m deep GAC filter units is backwashed every 2-4 days of operation. The short term turbidity increase that can be detected after sand filtration as a result of the load increase during backwash, is negligible after GAC filtration.

The process is considered excellent for cold and humic surface waters, when both process stability, treated water quality and technical and economic feasibility are taken into account.

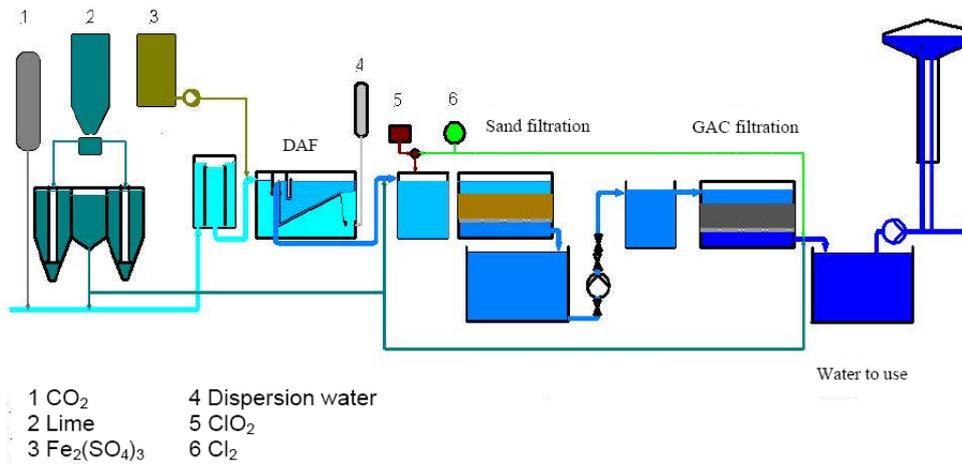
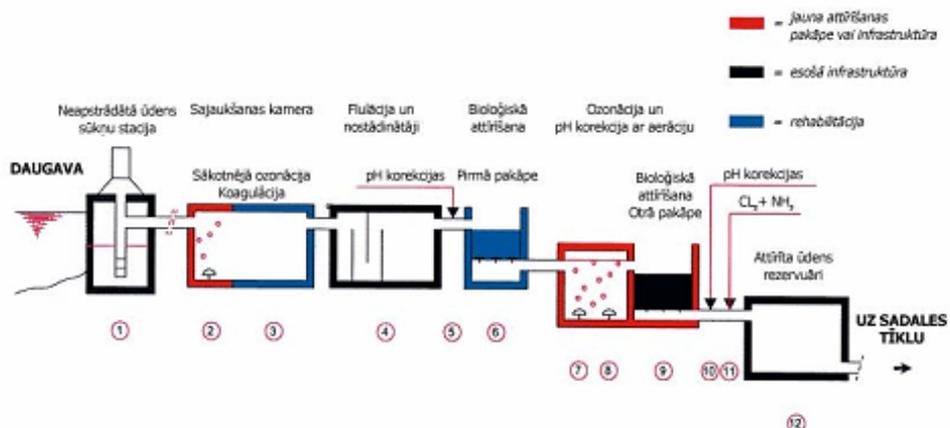


Figure 9.1. Treatment process scheme of the Rusko WPT (Jokela, et al 2006).

9.2 Daugava WTP, Latvia

Daugava water treatment plant is the largest plant in Latvia supplying about 50% of the drinking water for Riga city. Treatment includes coagulation with preozonation, sedimentation, filtration, ozonation and biofiltration, corrosion control and chlorine disinfection. The raw water is taken from the River Daugava (the Riga hydroelectric power station reservoir). Upstream the intake several villages and cities are located (in Russia, Belarussia and Latvia), therefore the raw water is influenced by wastewater discharges.

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 recent study period the concentration of DOC in the river varied between 9 and 16 mg/L with the lower values during autumn and winter.



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. Process scheme of the Daugava WTP in Riga. Newly added treatment processes 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.

The process optimization potentials of introducing enhanced coagulation, and the effect of preozonation should be studied here. In addition, the ozonation-biofiltration process step needs further investigation and optimization efforts.

9.3 Jordalsvatnet WTP, Norway

Jordalsvatnet water treatment plant (Figure 9.3) is located in Bergen at the west coast of Norway. It is an enhanced coagulation-contact filtration plant with integrated corrosion control and UV disinfection. Ferric chloride is used for coagulation, and CO_2 and alkaline filtration is used for pH and corrosion control. The 3-M filter bed is composed of 0.5 m 0.8-1.6 mm anthracite, 0.4 m 0.4-0.6 mm sand, and 1.4 m 1-3 mm crushed calcium carbonate as the bottom layer above the 0.35 m deep support layer.

The design treatment plant capacity is $1900 \text{ m}^3/\text{h}$, with a design rate of filtration of 8 m/h. There are 6 filters with a total area of 281 m^2 . The filters are backwashed with air, followed by water at 60 m/h for 6 minutes. During filter ripening a filter-to-waste procedure is employed for 40 minutes. In addition to the control of coagulation pH, finished water pH is controlled at pH by addition of NaOH. The UV-disinfection units are designed for a dose $> 40 \text{ mJ}/\text{cm}^2$. There is no use of filter aid polymers, a polymer is however applied in the spent backwash water/sludge thickening process. Thickened sludge is transported by the sewer system to the wastewater treatment plant.

9.4 Skullerud WTP, Norway

Skullerud WTP in Oslo, Norway (Figure 9.4) is a direct filtration plant with a treatment capacity of $1800 \text{ m}^3/\text{hr}$ in two parallel lines. The treatment includes alum coagulation at pH about 6, flocculation (2 units), and filtration in 3M filters (3 units per line). Non-ionic polyacrylamid is used as a filter aid in small quantities (0.1 mg/L). This polymer is also used in the spent backwash water/sludge treatment process. The 3-M filter bed has an upper layer of 3-3.5 mm low density PE granules, a second layer of 2-2.5 mm high density PE granules, and a bottom layer of 0.8-1.2 mm sand. Each layer is 60 cm deep. Corrosion and pH control is performed by $Ca(OH)_2$ and CO_2 . Sodium hypochlorite is used for disinfection, but UV-disinfection units are now under

installation. The sludge from filter backwash thickening is sent through the sewer system to the Bekkelaget wastewater treatment plant, while decanted water is equalised and returned to the raw water inlet.

The design filtration rate is 9 m/hr., and the actual rate is 6-8 m/hr. Filter run times are typically 14-15 hours for filtration rates of 8 m/h, and raw water colour and TOC levels of 20-30 mg Pt/L and 3.5-5 mg TOC/L, respectively., and alum dose levels of about 2.1 mg Al/L. Typical treated water quality levels are: colour 2-6 mg Pt/L; turbidity <0.1 NTU; residual Al < 0.15 mg/L.

Backwashing is performed with air (6-7 minutes) followed by water for fluidisation (45 m/h in 9-10 minutes). During filter ripening a filtrate-to-waste procedure is used for a time period of 10 minutes.

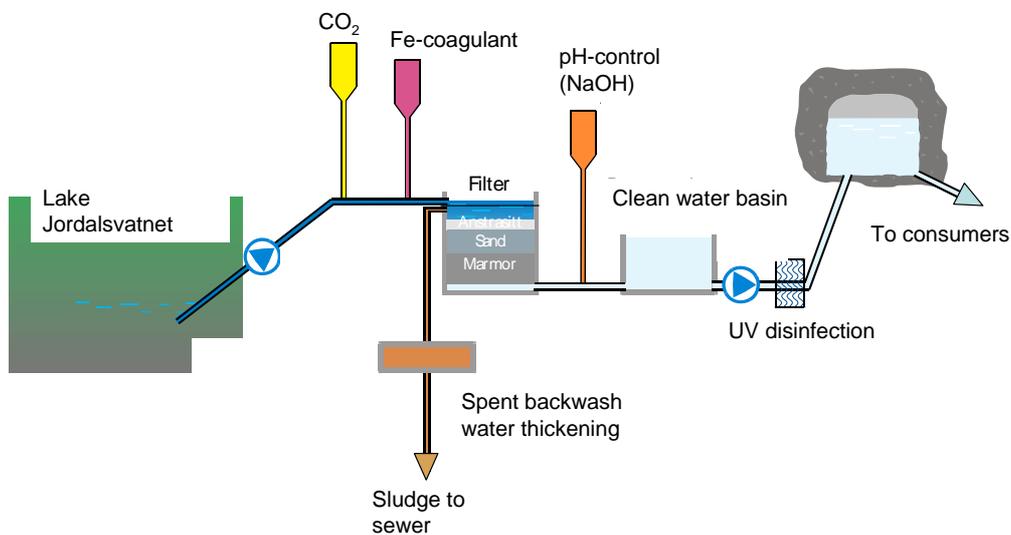


Figure 9.3. Schematic of the Jordalsvatnet WTP in Bergen, Norway

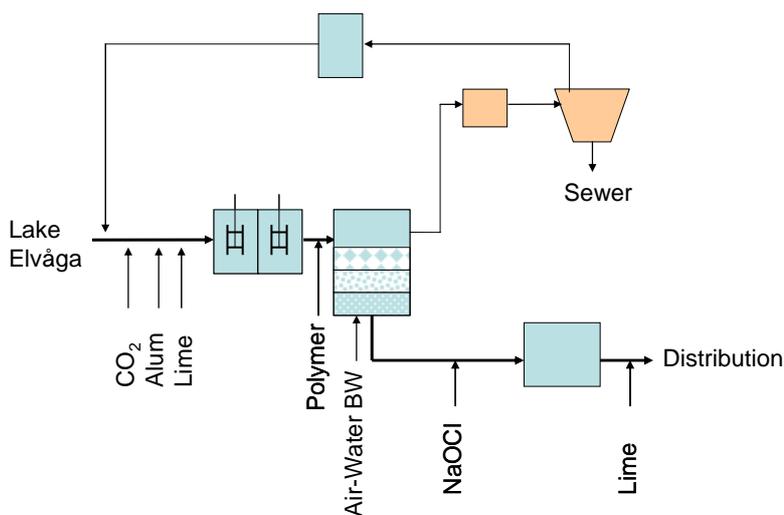


Figure 9.4. Schematic of the Skullerud WTP in Oslo, Norway

10 Knowledge gaps and research needs

Because natural organic matter (NOM) interferes with most water treatment technologies, controls enhanced coagulation conditions and treatment process performance, NOM should be offered more attention with respect to treatment plant operation.

NOM characterization and treatability, enhanced coagulation process optimization, major operational challenges and bottlenecks, best overall operation practices, and process control options are factors that need further addressing:

1. The climate-related trends towards increasing NOM content (link to WA1), as well as the great seasonal variability in NOM composition and the differences in treatability of the different NOM fractions, represent significant challenges with respect to process optimization and process control. These issues need further addressing.
2. There are still remaining challenges regarding the linkage of raw water characterization and best possible enhanced coagulation process operation and performance, and more knowledge is needed on how to remove NOM-fractions that are recalcitrant to removal by conventional enhance coagulation processes. The rapid NOM-fractionation procedure should be further tested and developed into a valuable operation tool; e.g. for identifying and resolution of non-optimum enhanced coagulation conditions and performance
3. The neutral fractions of NOM represent a major challenge with respect to efficient removal. A two-step enhanced coagulation process with intermediate ozonation should be tested in order to increase removal of non-humic NOM, i.e. hydrophilic neutrals (HPN), and to avoid decreased removal of humic NOM, i.e. fulvic acids (FA) as a result of ozonation.
4. Enhanced coagulation operational models should be developed and adopted and verified in pilot and full-scale applications. The models should be extended to other raw water qualities, other operational conditions and process configurations, other coagulants, polymer addition, etc. Enhanced coagulation and metal-based coagulants yields early breakthroughs and short filter runs. Filter aid polymers have demonstrated their ability to act as efficient operational tools to allow for relatively high filtration rates while still maintaining relatively long filter run times and high treatment capacity. The impacts and mechanisms of filter aid polymers on enhanced coagulation and direct or contact filtration should however be further investigated. The effects of polymer on filter run time and breakthrough do not seem to be consistent. The effect of polymer addition and why the polymer is very efficient in some cases and far less effective in other cases should be addressed.
5. The widespread manual coagulant dosage control routines should be replaced by control options based on in-line raw water monitoring,

e.g. by the adaptation of an in-line Delta UV spectra sensor for coagulant dosage control.

6. In a multi-objective enhanced coagulation treatment perspective, the best overall as well as possible conflicting operation conditions with respect to optimum removal of turbidity/particles, micro organisms and parasites, and NOM/DBP-precursors should be addressed.
7. The important role of metal hydroxides in NOM removal/NOM adsorption in enhanced coagulation processes should be further investigated, including identification of the relative importance of adsorption of NOM to metal hydroxide flocs (in conventional and direct filtration processes) versus adsorption to metal hydroxide deposits (in contact filter beds). It should be investigated if the mechanisms and efficiencies are different.
8. Optimum backwash techniques for different types of filters should be identified and tested, e.g. for 3M filters with an integrated deep layer of crushed CaCO₃ for the purpose corrosion control.
9. Optimization routines and best operation practices should be demonstrated in cooperation with end-users (links to WA1, WA3, WA4 and WA7).

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