Assessing Pretreatment Methods to Mitigate Nanofiltration Fouling

Enhancing Efficiency and Performance of Nanofiltration Operating in a Modular Drinking Water Treatment System

Master's thesis in Infrastructure and Environmental Engineering

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Gothenburg, Sweden 2025
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Master's thesis 2025

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Master's Thesis 2025
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Cover: A visualization of a Modular Drinking Water Treatment System, constructed as a container with nanofiltration. Generated through Canva AI Design Tool.

Typeset in LaTeX
Printed by Chalmers Reproservice
Gothenburg, Sweden 2025

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Abstract

Globally, water scarcity is a critical issue affecting millions of people, exacerbated by increasing population and climate change. In countries like Sweden, where water availability is sufficient, this vital resource is often taken for granted. However, the extensive reliance on centralised water infrastructure in Sweden has its limitations. To address these challenges, modular drinking water treatment systems (MDWTS) offer a viable solution. These systems can serve as a complement to, or in the absence of, centralised water infrastructure, ensuring sustainable water management and availability. Membrane filtration is a compact treatment method applicable for various raw water qualities, which makes it suitable to implement in a MDWTS. However, membrane modules face an obstacle due to the risk of fouling caused by organic and inorganic compounds in the raw water. Fouling results in a reduction of treatment efficiency, while increasing the need of operational and economic resources. The aim of this thesis is to suggest suitable pretreatment methods for raw water to mitigate membrane fouling in a MDWTS, where nanofiltration acts as the main treatment step. The issue was investigated through a study of literature, identifying key foulants and possible pretreatment methods. The key foulants identified in the study of literature are particles, natural organic matter, iron, manganese, calcium, magnesium, aluminium and silica. In addition, bench studies on a biological activated carbon filter, an alkaline filter and a charged sieve followed by a disc filter were performed to evaluate their potential to operate as pretreatment. Further, the findings were applied to three water types representing water qualities from three water sources in Sweden, aiming to suggest pretreatment chains to reduce key foulants. The pretreatment chains suggested in the result for the three water types are all spatially manageable in a MDWTS. Although, for one of the water types, the pretreatment may be challenging to implement due to operational complexity. To include additional water qualities, a general procedure connecting key foulants to suitable pretreatment methods is presented. Due to the complexity of nanofiltration fouling and the possible interactions between key foulants and the different steps in the pretreatment chain, pilot studies are necessary to evaluate the fouling mitigation. By validating the framework through pilot testing, this work can contribute to enhancing nanofiltration performance and further the development of MDWTS.

Keywords: MDWTS, Nanofiltration, Fouling, Pretreatment.

Acknowledgements

This master thesis was carried out during the spring of 2024, as the final work within the master of Infrastructure and Environmental Engineering, at Chalmers University of Technology. First, we would like to address our gratitude towards our supervisor at Chalmers University of Technology, Oskar Modin, for giving us valuable insights and expertise to proceed this project. Further, we would like to highlight our appreciation to our colleagues at Norconsult AB for the opportunity to perform this project among an engaging and supportive team. We want to address our greatest gratitude to our supervisors at Norconsult AB, Bo Berghult, Stephan Köhler and Debora Falk, for sharing their excellence and enthusiasm within the field. In addition, we are thankful for the opportunity to conduct this project in collaboration with Björks Rostfria AB, and to be a part of developing their vision of MDWTS. Lastly, we want to show our appreciation to the people working at the drinking water treatment plant, for facilitating the performance of our bench study.

Beate Hallset and Emma Jakobsson, Gothenburg, June 2024

List of Acronyms

Below is the list of acronyms that have been used throughout this thesis listed in alphabetical order:

BAC	Biological Activated Carbon
COD	Chemical Oxygen Demand
EBCT	Empty Bed Contact Time
EC	Emerging Contaminants
EPS	Extracellular Polymeric Substances
DBP	Disinfection By-Products
DOM	Dissolved Organic Matter
HMW	High Molecular Weight
LMW	Low Molecular Weight
MDWTS	Modular Drinking Water Treatment System
MF	Microfiltration
MWCO	Molecular Weight Cut Off
NF	Nanofiltration
NOM	Natural Organic Matter
PFAS	Per- and Polyfluoroalkyl Substances
POM	Particulate Organic Matter
RO	Reverse Osmosis
TMP	Trans Membrane Pressure
TOC	Total Organic Carbon
UF	Ultrafiltration

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Introduction

In 2015, the United Nations set up the 2030 Agenda for Sustainable Development, including 17 goals, whereof one goal is to offer clean water and sanitation for all. In the Sustainable Development Goals Report, the United Nations (2023) states that even though the drinking water field has experienced a great progress, access to secure drinking water is still a scarcity for billions of people. Simultaneously, challenges concerning water quantity and quality have arisen. The world's population is increasing, which affect the water scarcity because there is a strong connection between population growth and water availability. Climate change is another challenge within the drinking water sector. The human lifestyle including the use of fossil fuels and changed land use leads to an increase of greenhouse gases resulting in global warming (Calvin et al., 2023). As a consequence, climate and weather are affected with changed temperatures and precipitation, leading to flooding and droughts.

These problems influence the infrastructure in the cities including drinking water treatment plants, therefore the water security is threatened which could imply devastating consequences connected to human health (Calvin et al., 2023). In order to maintain a secure water production, the drinking water treatment plants need to be climate adapted (Swedish NFA, 2019). The process of climate adaption includes the development of a new water treatment plant or the selection of a new raw water source, which are time-consuming processes. In the absence of, or as a complement to centralised water plants, decentralised water treatment can contribute to the production of clean water (Stoler et al., 2022). Decentralised water treatment includes smaller solutions which can be located near the water supply or where the treatment should operate (Kulkarni et al., 2022). Modular drinking water treatment systems (MDWTS) are a type of decentralised treatment, that is applicable for various flows and water qualities.

A drinking water treatment method that has gained popularity in recent years is membrane filtration (Lidén, 2020). Membrane filtration is a physiochemical separation method, which utilizes the differences in permeability of the components in raw water (Crittenden et al., 2012). Membranes possess the ability to remove particles, pathogens, organic matter. Using a denser membrane can additionally retain ions and dissolved compounds. When using membranes, the need for coagulation, flocculation and sedimentation facilities are reduced. As a result, membrane plants are allowed to be compact while suitable for different sources of raw water, making them applicable for MDWTSs.

However, membrane modules face an obstacle due to the risk of fouling caused by the accumulation of organic and inorganic compounds in the raw water. Fouling results in a reduction of treatment efficiency while increasing the need for maintenance and operational costs. To ensure that membrane facilities constitute a secure option to offer clean water, the issue with fouling needs to be investigated and countered. In order to reduce the fouling problems, pretreatment of the water can be implemented to decrease the microorganisms, organic and inorganic matter prior to the nanofiltration system.

1.1 Aim

The aim of this thesis is to suggest suitable pretreatment methods for raw water to mitigate membrane fouling in a MDWTS, where nanofiltration acts as the main treatment step.

1.2 Objectives

To achieve the aim of this thesis, the following objectives are outlined:

- To describe the function of nanofiltration used in drinking water treatment and account for the key foulants causing membrane fouling.
- To suggest pretreatment chains for three raw water types. The pretreatment should be applicable prior to nanofiltration in a MDWTS and the water types characterizes surface water and groundwater occurring in Sweden.
- To create a general solution, connecting key foulants with suitable pretreatment methods, serving as a framework to facilitate the development of an effective pretreatment chain.

1.3 Limitations

The raw water qualities investigated within this work is geographically limited to Sweden. Therefore, the raw water sources which are analysed are limited to surface water and groundwater, which in the current state constitutes the potential sources for drinking water production in Sweden during regular circumstances. Seawater, brackish water and other types of potential water sources are excluded in this study.

Further, this study is based on membranes using nanofiltration. The requirements of the pretreatment method are specifically investigated for nanofiltration, which may differ from requirements for other types of membranes, such as microfiltration, ultrafiltration, and reverse osmosis. In addition, the pretreatment should be applicable in a MDWTS, which also affects the selection of pretreatment methods.

The treatment steps posterior the membrane filtration, distribution of the drinking water as well as discharge and waste flows from the treatment steps are not included in this work. In order to achieve a good water quality and quantity, it is important to consider all the steps and understand the interaction between them.

Since this project consider the planning stage of a treatment chain and is time limited to 20 weeks, the actual performance of the chosen pretreatment method in the real case scenario is not validated. To assure if the proposed pretreatment method is sufficient, pilot testing should be implemented. To take account for seasonal variations, the period of the testing for membrane facilities should last for a year at minimum (Lidén, 2020). The time limitation also affects the experiments, since a stabilization period may be required to obtain accurate results.

Specific use of material and design of a membrane module is used as a basis for the investigation within this work. Since the physical and chemical characteristics of the material affects the performance of the membrane (Crittenden et al., 2012), the requirements of the pretreatment method may vary depending on the type of material.

Lastly, economical and ecological aspects related to the pretreatment methods are not investigated to a further extent within this work.

2

Theory

2.1 From raw water to drinking water

This first theory chapter will give a brief overview of the raw water used for drinking water production in Sweden, raw water components, water quality parameters, regulations and guidelines for drinking water treatment.

2.1.1 Raw water in Sweden

In general, water availability in Sweden is sufficient, using both surface water and groundwater as raw water sources for drinking water production (Svenskt Vatten, 2016). Water found in lakes and watercourses is classified as surface water, whilst the water present in cracks, pores and voids in the soil- and rock layers is classified as groundwater (Svenskt Vatten, 2016; Svenskt Vatten, 2010a). Because of this, surface water and groundwater chemically differs. Groundwater could also be artificially made through various of processes, such as induced recharge, injection wells, infiltration ponds and basins (USGS, 2024). In order to classify the artificial groundwater as unaffected, it has to possess a residence time of at least 14 days (Swedish NFA, 2024). Around half of the produced drinking water in Sweden is extracted from surface water sources, while natural and artificial groundwater constitute the other half (Svenskt Vatten, 2016).

Usually, surface water may contain high concentrations of organic matter, whilst the inorganic content is lower (Svenskt Vatten, 2010a). The natural organic matter (NOM) origins from plants and living organisms (Zheng et al., 2024). Human activities, which include agriculture, industries, and wastewater treatment plants, also affects the concentration of organic matter and microorganisms in surface water (Swedish EPA, 2024). Different conditions concerning nutrient and oxygen availability, sunlight exposure, flow and mixing conditions, temperature etc. affect the concentrations of microorganisms in surface water (Svenskt Vatten, 2010a). In general, surface water possess favourable conditions for microorganisms, resulting in high microbial activity. Further, the temperature of surface water varies with the seasons. It is important to consider the effect of the thermocline occurring during the summer in lakes in Sweden. As a result, the levels of iron and manganese increases during the late summer and winter. Shallow groundwater is also classified as surface water. The conditions around shallow groundwater generate a high content of natural organic matter and mineral salts, which origins from the soil- and rock layers.

In comparison to surface water, groundwater infiltrates the soil layers, resulting in a natural self-cleaning effect (Svenskt Vatten, 2010b). However, the different soil layers affect the organic and inorganic content in the water throughout the infiltration process. If the organic content is high, the groundwater is most likely affected by shallow groundwater (Geological Survey of Sweden, 2024). Artificial infiltration of surface water in order to recharge groundwater could also increase the organic content. Since organic content interact with metals, a high organic content enables increased metal transport.

In Sweden, the most common soil types are podzol and brown soil (Kullberg, 2014). Groundwater recharge in podzol starts with rainwater entering the humus layer, where accumulation of organic matter takes place, resulting in a low organic content (Svenskt Vatten, 2010b). Further, in the ash grey layer, carbon dioxide is generated and soluble humic and fulvic acids create acidic water. These conditions affect the mineral particles present in the next layer. This process leads to an increased pH value since the mineral particles contains basic compounds. Another occurring process is the water uptake of ions, resulting in precipitations of iron, aluminium, humic and fulvic acids in the following layer called reddish brown layer. The amount of ions which does not precipitate in this layer remains in the water, explaining the high inorganic content. The microbial and organic matter is usually low at the end of the infiltration process. Concerning the groundwater temperature, it is more stable than the surface water temperature and reaches lower values all year around.

Most groundwater used for drinking water production in Sweden is extracted from high permeable Quaternary deposits (Kitterød et al., 2022). Depending on the origin of the deposits, the groundwater can achieve various quality parameters. In swell deposits and till, which covers approximately 75 % of Sweden's surface area, the groundwater has a short residence time (Swedish Geotechnical Institute, 2023 ; VISS, n.d.). A short residence time enhances leachate of base cations, making the groundwater sensitive to acidification. In contrast, the residence time is considered to be long for groundwater found in glaciofluvial deposits and till located below a layer of clay, enhancing high concentrations of base cations and high alkalinity. Further, anaerobic conditions are often found in confined aquifers and consequently high concentrations of iron and manganese can be found in the groundwater. Glaciofluvial deposits are commonly used for extensive groundwater extractions. The groundwater in glaciofluvial deposits often have a long residence time.

Further, the chemical quality of the groundwater is governed by the mineral composition of the underlying bedrock (Kitterød et al., 2022). Sedimentary rock, which can be found in Skåne, on the islands Öland and Gotland and in some lots on the inland of Sweden, affects the water quality significantly (VISS, n.d.). The richness of lime content generates a well buffered water with high concentrations of calcium and magnesium. In addition, a high content of sulfate, which can occur in sedimentary rock, creates a reducing environment where iron and manganese can dissolve.

In crystalline rock, existing extensively in Sweden, groundwater is most commonly found in shallow fractures, where it obtains high alkalinity and low concentrations of iron. High concentrations of iron can be found in fractures found at greater depths where reducing conditions occur, or as a result of peat affected groundwater during large water extractions.

Traditionally and in the current state, the surface water treatment plants operating in Sweden are few but large, mainly located by bigger cities (Svenskt Vatten, 2016). These treatment plants consists of different treatment steps depending on the quality of the influent raw water. Commonly, conventional drinking water treatment plant consists of coagulation, flocculation, sedimentation, filtration and disinfection (Naser et al., 2021). In contrast to surface water treatment plants, groundwater treatment plants in Sweden are smaller and more simple, and mainly supplies smaller communities with drinking water (Svenskt Vatten, 2016)

2.1.2 Raw water components

Water may contain a wide range of compounds with different structures, properties and behaviours. In this section, microorganisms, natural organic matter (NOM), and inorganic compounds are further explained.

Microorganisms are present everywhere, for instance in soil, water, air, food and the human body. A big part of the microorganisms are harmless and important for human health and other living organisms (Svenskt Vatten, 2024b). Another part of the microorganisms can cause waterborne diseases and outbreaks. These microorganisms are called pathogens and origin from sources such as infected faeces, food, water and soil environments (Crittenden et al., 2012;World Health Organization, 2017). Exposure to these pathogens usually results in gastrointestinal illness. Pathogens can be divided into bacteria, algae, viruses, protozoa and helminths. In order to discover microbial contamination, regular sampling and analyses of water samples are required (Svenskt Vatten, 2024b). Since some of the microorganisms are hard to detect, indicator organisms are commonly used. In addition to the indicator organisms, risk assessments are conducted to ensure safe drinking water.

Bacteria are simple organisms, consisting of one single cell. The size of bacteria varies from approximately 0.1 μm to 10 μm (Crittenden et al., 2012). They can be categorized into different groups based on their properties, some examples of bacteria from different groups are Escheria coli (E.coli), Salmonella, Vibrio cholera and Legionella pneumophilia. Viruses possess a parasitic behaviour, which implies that they must feed on their host cell to persist. Compared to bacteria and protozoa, the size of viruses are small, generally ranging from 0.001 μm to 0.01 μm. Viruses are also divided into different categories, for instance Adenoviruses, Astroviruses, Enteroviruses and Caliciviruses (World Health Organization, 2017). Protozoa are single cell animals and possess a relatively complex structure (Crittenden et al., 2012). They can function on their own without a host, however they often take advantage of the host. Protozoa vary in size from 1 μm to 100 μm. Some examples of protozoa from different categories are Giardia lamblia, Cryptosporidium parvum and Entamoeba dispar.

Blue-green algae consists of Cyanobacteria and differs in size from 1 μm to 100 μm (Crittenden et al., 2012). High abundance of Cyanobacteria in algal blooms can lead to creation of toxic substances, resulting in adverse consequences for human and animals. Parasitic worm or helminths are present in the size interval 1 μm to 10 000 μm. Compared to the earlier mentioned pathogens, algae and helminths are detectable without a microscope.

The risk of infection varies for the different pathogens. For instance, exposure to only a dozen of Cryptosporidium oocysts can result in infection (Institutet för hälsa och sjukvård, 2024). Giardia lamblia also requires a low number, approximately ten cells, to develop an infection (Leggett et al., 2012). In comparison, at least the number of 1000 Vibrio cholera cells need to be present in order to develop an infection. To effectively remove microorganisms from raw water, it is important to consider their properties, including shape, size, surface charge, requirements for oxygen/-carbon/energy, and motility (Crittenden et al., 2012). Their stability in different environments is also important to consider. For bacteria, viruses and protozoa, the surface charge is negative.

NOM are carbon-based compounds which originates from degradation of aquatic organisms and plants (Zheng et al., 2024). The various types of NOM have different characteristics since they are structured in different ways, with varying proportions and comes from different raw water sources (Mallya et al., 2023). Moreover, the characteristics of high importance for NOM are surface charge, molecular weight and existing functional groups (American Water Works Association., 2011). NOM occur in different forms in water, either as dissolved organic matter (DOM) or particulate organic matter (POM), distinguished around the size of 0.45 μm (Docter et al., 2015). Colloidal NOM can vary in size from around 0.01 μm to 0.1 μm (American Water Works Association., 2011). However, for a particle to be considered as a colloidal particle, it needs to have a dimension $<$0.5-1 μm.

Functional groups are groups of atoms in a molecule which affect the outcome of the molecule's chemical reactions. NOM consists of functional groups, for instance humic acids constitutes of carboxylic acids, phenolic alcohols, and methoxy carbonyls (Gao et al., 2023). These functional groups affect the properties of NOM, such as the reactivity, adsorption, solubility and the interactions with other components. NOM may be divided into hydrophobic, hydrophilic and transphilic portions. The hydrophobic portion compose of humic and fulvic acids which have high molecular weight, whilst the low molecular weight hydrophilic portions for instance include proteins, polysaccharides and carbohydrates (Mallya et al., 2023). The transphilic portion are possessing characteristics in between the hydrophobic and hydrophilic portions. The transphilic and hydrophilic portions cover for 20-40% of the DOM (L. Fan et al., 2001). Humic substances constitute more than 50% of the DOM, and is the greatest proportion of NOM.

When particles occur in water and solutes, their surface often becomes electrically charged (American Water Works Association., 2011). During the majority of conditions, the electrical charge is negative. The particulate charge of NOM could be described by the reaction of anions such as NOM combined with functional groups. As a consequence, interactions between NOM and the particles of the functional groups occur. Different types of interactions, including covalent bonding, hydrophobic interactions, hydrogen bonding and ligand exchange, enables the bonding between anions and functional groups. The negative surface charge of particles could usually be explained by the NOM coating of particles. In addition, functional groups are able to ionize in contact with water molecules. This phenomenon also affects the surface charge of particles and is strongly connected to the pH-value in the water. A low pH results in a positive surface charge, and a high pH results in a negative surface charge. However, natural particles possess a negative charge at the majority of the pH levels in natural waters. According to Mallya et al. (2023) NOM with LMW have a greater negative charge than HMW NOM. Charge interactions are vital for the removal of LMW NOM.

NOM by itself is not damaging, however during disinfection processes it can potentially result in disinfection by-products (DBP) with adverse health effects (Svenskt Vatten, 2010a). Other consequences which arise during water treatment is the emergence of synergy between NOM and pollutants. These pollutants among other things include emerging contaminants (ECs), per/polyfluororoalkyl substances (PFAS), heavy metals, and pharmaceutical compounds. Further, NOM emphasizes the creation of biological growth and aesthetic complications.

Inorganic components, such as salts, nitrites, heavy metals and phosphates, have been found in various ground- and surface waters (S. Singh et al., 2020). Accounting for these components are of significant importance during drinking water production, due to their potential impact on the treatment process and distribution network, as well as health-related aspects. The content of calcium and magnesium ions affects the hardness of the water. While high hardness generally does not pose a health hazard, it is of concern since calcium carbonate can precipitate, leading to disturbance in treatment processes, distribution networks and household installations (Svenskt Vatten, 2008). Similarly, high concentrations of iron and manganese are rarely harmful for human health, but can cause technical issues in treatment processes and during distribution, as well as aesthetically issues, such as colour and odour. In Guidelines for Drinking-Water Quality, the World Health Organization (2017) states that some heavy metals in drinking water, such as arsenic and cadmium, pose a threat to human health. Consequently, the guidelines provide regulations of accepted concentrations of heavy metals in drinking water.

2.1.3 Water quality parameters

Along the way of producing drinking water from raw water, the monitoring of several physio-chemical parameters is significant to examine the water quality. A selection of water quality parameters relevant within this work is presented briefly in this subsection.

The pH accounts for the concentrations of hydrogen ions in a solution and is used as a measurement of a solution's acid-base relation (Crittenden et al., 2012). Usually, pH has no direct influence on consumers, but it is mentioned as one of the water quality parameters that has the most operational significance during drinking water production (World Health Organization, 2017). pH has a direct impact on chemical processes, such as dosage of chemicals to coagulate particles and managing the hardness of the water (Crittenden et al., 2012). In addition, a low pH in the effluent water can cause corrosion of the pipes in the distribution network.

The ability of water to resist changes in pH is measured as alkalinity (Crittenden et al., 2012). In the presence of a weak acid, biological and chemical activities can occur without changing the pH of a water, due to the acid's ability to consume or produce hydrogen ions supporting other reactions. The carbonate system is the primary source of alkalinity, yet carboxylates, silicates, phosphates and borates can also impact the alkalinity. The alkalinity and pH are the parameters that primary governs the corrosion rates of pipes in the distribution network.

Another water quality parameter to consider during drinking water production is the turbidity, which describes the cloudiness of water. The turbidity is increased due to the presence of organisms, organic particles (e.g. plant debris), suspended particles (e.g. silts and clay) and chemical precipitates (e.g. calcium, iron and manganese) in the water (World Health Organization, 2017). Changes in turbidity are commonly occurring due to variations of source quality, but variations of the effluent turbidity can also indicate on disturbance during the treatment process. For example, high turbidity can limit disinfection effects on microorganisms, stimulate microbial growth and indicate the presence of microbial and chemical contaminants. While particles that increase the turbidity rarely are significant to health, it is of importance to manage, thus high turbidity generally reduces the acceptability of drinking water.

High turbidity can enhance an apparent colour of water, which can be measured on unfiltered samples. When measuring the colour of filtered samples, the true colour of water, affected by the presence of dissolved organic content (e.g. humic and fulvic acids) and metallic ions (e.g. iron and manganese), can be determined (Crittenden et al., 2012). The origin of colour in drinking water should be determined and is of particular importance in the event of substantial changes (World Health Organization, 2017).

Organic content exist in various shapes, sizes and composition in water and to measure the concentrations of each of the various types is comprehensive. Therefore, a water quality parameter frequently measured is the total organic carbon (TOC), accounting for the total concentration of carbon occurring in organic matter in a sample (Crittenden et al., 2012). TOC can further be distinguished by measuring the dissolved organic carbon (DOC) and particular organic carbon. Another method to gain knowledge of the organic content in water is to measure the chemical oxygen demand (COD) and biochemical oxygen demand (BOD). COD accounts for the concentration of oxygen required to degrade the total content of organic matter, while BOD accounts for the concentration of oxygen required to decompose the biological degradable organic matter.

In conformity with organic matter, it is not feasible to measure all microorganisms that exist in a water sample. Therefore, some indicator organisms, e.g. E. coli and coliform bacteria, are monitored during drinking water production. The indicator organisms are chosen due to their ability to indicate the presence of fecal material in the water and their property of being easy to analyse (Svenskt Vatten, 2024b). Even though the presence of indicator organisms not necessarily imply presence of pathogens, it is a significant indicator on a water's level of pollution (Lindberg and Lindqvist, 2005).

Hardness, total dissolved solids (TDS) and electrical conductivity constitute inorganic quality parameters relevant to measure during drinking water production (Crittenden et al., 2012). Hardness is the content of polyvalent cations, where calcium and magnesium primary governs the hardness, even though other ions can contribute. However, hardness is generally calculated as the sum of calcium and magnesium concentrations, and measured in German degrees of hardness, classified as shown in Table 2.1. Hardness can be either carbonate or noncarbonate. Carbonate hardness is the total amount of polyvalent ions connected to alkalinity such as HCO_{3-}, whilst the noncarbonate hardness is the amount of polyvalent ions connected to non-alkalinity, for instance SO_4^{2-}. While the hardness accounts for the content of polyvalent cations, TDS accounts for the total concentration of ions in a solution. Further, the electrical conductivity accounts for the concentration of dissolved constituents, measured by their capacity to conduct electrical charge. The presence of dissolved ions in a solution generates a high conductivity.

Table 2.1: The classification of hardness (Svenskt Vatten, 2010b)

Classification	Calcium content (mg/l)	Degree of hardness (°dH)
Very soft	0-15	0-2
Soft	15-35	2-5
Medium hard	35-70	5-10
Hard	70-140	10-20
Very hard	> 140	< 20

Lastly, the temperature of water is important to keep under observation since it has the ability to impact physical, chemical and biological processes, such as solubility and rates of reactions. High temperature can for instance enhance growth of microorganisms, corrosion and increase issues related to colour, taste and odour (World Health Organization, 2017).

2.1.4 Regulations and guidelines for drinking water production

The Swedish regulations and guidelines are based on international goals, requirements, and directives. One of the UN's sustainable development goals, more precisely goal number 6, is to "Ensure access to water and sanitation for all" (United Nations, 2023). To achieve this goal, a list of targets has been set up (United Nations, 2024). By 2030 the drinking water supply and sanitation should be globally equal. An improvement of the worldwide water quality with a decreased amount of pollution and untreated wastewater is desired. It is important to implement cross-border cooperation's to reach a sustainable water production, especially in developing countries with water stress and sanitary problems. The World Health Organization has published Drinking Water Guidelines, the main goal of this is to ensure good drinking water quality and thereby protect the public health (World Health Organization, 2017). The drinking water guidelines works as a basis for development of national drinking water policies by including topics such as the effect of climate change, chemical and other key contaminants, microbial hazards etc.

A major part of the WHO's guidelines is the concept of Water Safety Plans with a risk assessment approach, this is strongly connected to the UN's goals number 6. Water safety plans are made to manage the chemical and microbial quality of the drinking water and are carried out by doing an extensive risk assessment analysis which consider all the steps in the drinking water production (World Health Organization, 2024). In order to provide a risk assessment, the potential risk of hazardous events has to be identified. World Health Organization (2017) define a risk as "...the likelihood of identified hazards causing harm in exposed populations in a specified time frame, including the magnitude of that harm and/or the consequences." where a hazard is defined as "...a biological, chemical, physical or radiological agent that has the potential to cause harm;". By predicting and ranking the potential risks of the drinking water production, it is possible to take preventative actions.

In Sweden, drinking water quality is governed by the Swedish National Food Agency's regulations called LIVSFS 2022:12 (Swedish NFA, 2022). The regulations include treatment requirements and specific limit values for microorganisms, some chemical and radioactive compounds. There are no limit values for TOC mentioned in LIVFS 2022:12. According to Svenskt Vatten (2009), the content of TOC is classified as low when TOC <2 mg/l, which further is a desirable value to achieve for outgoing drinking water. These regulations are based on EU's Water Framework Directive. The objective of the water framework directive is to provide good water quality and quantity (European Commission, 2024). It applies to both surface water and groundwater.

Table 2.2: Limit values for drinking water in Sweden from LIVSFS 2022:12

Parameter	**Unit**	**Outgoing drinking water**	**Drinking water at the tap**
Turbidity	FNU	0.5	1.5
Colour	mg/l Pt	15	30
pH	-	10.5	6.5-9.5
Conductivity	mikroS/cm	-	2500
Alkalinity	mg HCO_{3-}/l	5-50	-
COD	mg/l	-	-
TOC	mg/l	-	No change
Aluminium	mg/l	-	0.2
Iron	mg/l	0.1	0.2
Manganese	mg/l	-	0.05
Calcium	mg/l	20-60	100
Magnesium	mg/l	-	30
Hardness	dH	-	-
Silica	mg/l	-	-

In order to achieve the limit values for microorganisms, microbial barriers are used to inactivate or separate microorganisms from the raw water. Depending on the raw water quality, the microbial barriers requirements differs (Svenskt Vatten, 2014). Usually, groundwater requires one barrier, whereas two if it is affected by surface water and contain a certain amount of coliforms, see table 3.6. Moreover, surface water requires two or three barriers, depending on the amount of coliforms in the water. Microbial barriers include:

- Artificial infiltration
- Chemical precipitation with filtration
- Slow filtration
- Primary disinfection
- Membrane filtration, including RO, NF and UF with a pore size <0.1μm

The Water Safety Plan's risk assessment procedure mentioned in the WHO's guidelines works as a complement to the microbial barriers to prevent microbial contamination (Svenskt Vatten, 2024a).

Table 2.3: Number of microbial barriers depending on the microbiological water quality

Parameter	Unaffected groundwater (in 100ml)	Surface water, groundwater affected by surface water (#/100ml)	Surface water, groundwater affected by surface water (#/100ml)	Surface water, groundwater affected by surface water (#/100ml)
E. coli or enterococcus	Not detected	Not detected	1-10	>10
Coliform bacteria	Not detected	Not detected - 10	>10-100	>100
Least numnber of barriers	1	1	2	3

2.2 Membrane filtration

Membrane filtration is a physio-chemical separation method, which separates components from water through differences in permeability (Crittenden et al., 2012). Water is pumped against a semipermeable membrane, allowing permeable components to pass through the membrane while retaining impermeable components on the feed side.

Based on their density, membranes are divided into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), ranked from the lowest density to the highest as illustrated in Figure 2.1. The limits differing the classes from each other are not completely unified (Lidén, 2020). MF and UF is defined by their pore size, and a pore size of 0.1 μm is a recognized limit classifying the filters. An alternative division between MF and UF is the ability to retain viruses, which is achieved by UF to full extent. When the density increases, the pore size becomes less significant to describe the membrane. For dense NF and RO membranes, it is instead diffusion through connected molecule chains that constitute the main mechanism transporting the water, enhanced by high pressure. Furthermore, the retention of divalent ions acts as a classification limit between UF and NF, while the retention of monovalent ions acts as the classification limit between NF and RO. Molecular weight cut off (MWCO), measured in Dalton, is connected to the size exclusion removal mechanism and is a measure of membrane filtration capacity.

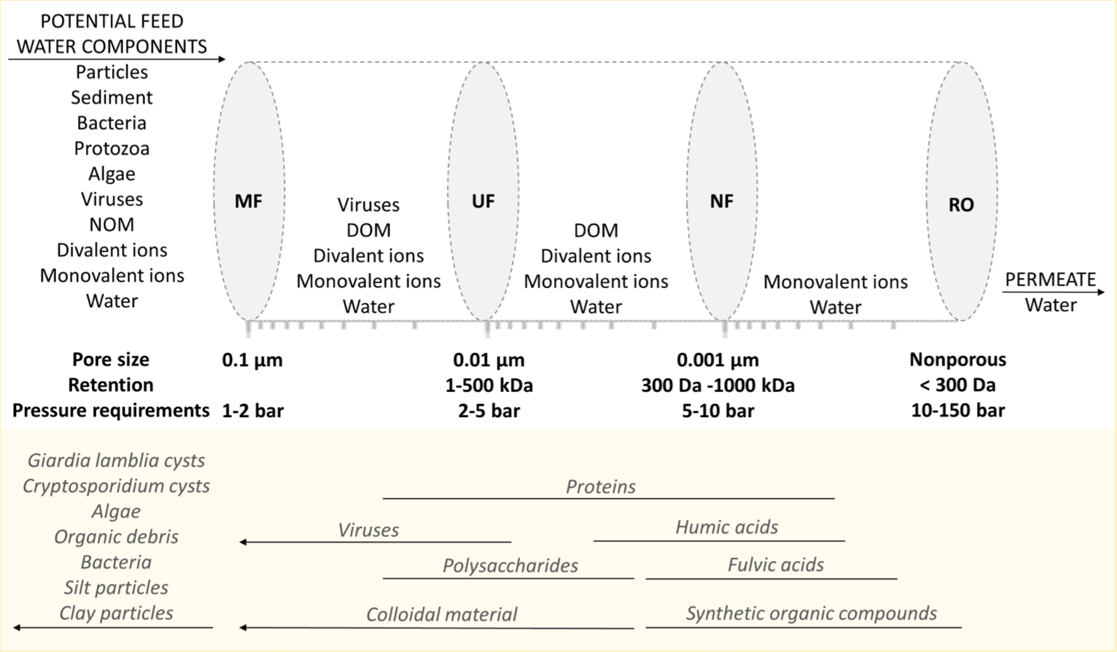


Figure 2.1: Illustration of the membrane types MF, UF, NF and RO, with description of their pore size/retention ability and pressure requirements. In addition, the water compounds which passes through the membranes are shown. The information presented is obtained from Crittenden et al. (2012) and Lidén (2020)

The primary removal mechanism during membrane filtration is straining, where components are removed through size exclusion (Crittenden et al., 2012). In addition, components are removed by adsorption and cake formation, due to concentration polarization. Adsorption is especially operative as a removal mechanism for a clean membrane, during the early stages of filtration. Material in the feed water, such as NOM, adsorbs to the membrane, resulting in a reduction in size of the voids. This leads to an increase in the membranes' ability to retain smaller components and soluble materials, even though their physical size is smaller than the theoretical retention rating. In addition, a clean membrane will promptly accumulate a cake of components at the membrane surface. High concentration components will, due to their size or charge, accumulate close to the membrane surface, with a decreasing concentration further away from the surface. This concentration polarization affects the resistance for water to pass through the membrane.

In membrane processes, the water that is pressured towards the membrane surface is termed feed water (Crittenden et al., 2012). Permeable components pass by through the membrane surface, creating the permeate. The permeate is addressed as flux and given in liters per membrane surface and hour (l/m^2h). On the feed side, impermeable components accumulate as retentate, reaching higher concentrations during the operation. For the process to proceed, the retentate needs to be discharged continuously, partly or entirely depending on the concentrations in the feed water and the membrane's retention ability. The retentate also depends on the design of the flow conditions within the membrane module. Dead-end flow, which particularly is used for MF and UF, represents a process where the feed water is pumped perpendicular to the membrane surface.

Due to the higher density of NF and RO membranes, the concentration polarization becomes excessive during the use of dead-end flow, increasing the resistance which further obstruct the treatment process (Lidén, 2020). When using denser membranes, cross-flow is used for the process to proceed effectively. During cross-flow, water is pumped parallel to the membrane surface with high pressure, which decrease the effect of the concentration polarization. In addition, the retentate during cross-flow can be designed to recirculate, allowing a higher recovery rate for NF and RO membranes. Still, due to the need of discharging retentate as its concentration increases, a significant loss of recovery rate will occur. Dead-end and cross flow are illustrated in Figure 2.2.

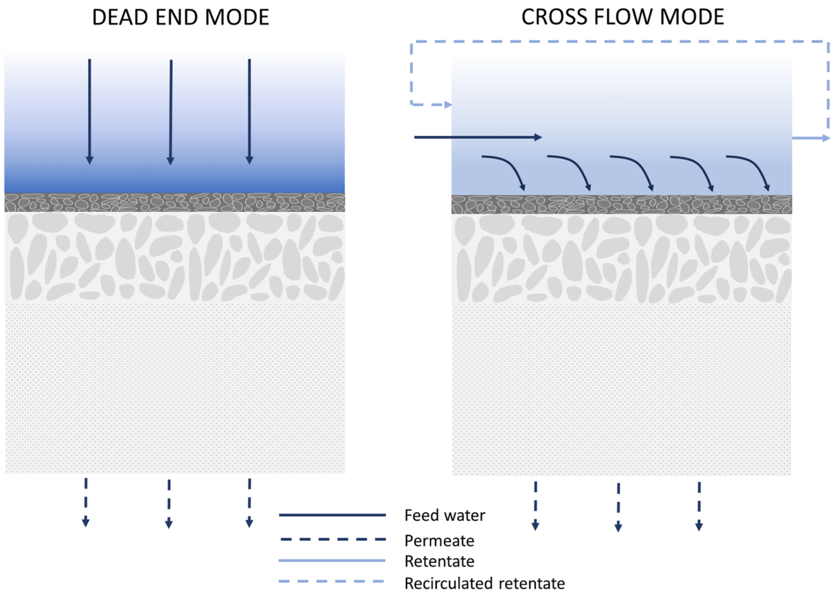


Figure 2.2: Membranes operating in dead end mode (left) and cross flow mode (right)

When operating at a constant flux, an important parameter to consider is the transmembrane pressure (TMP) drop that accounts for the differential pressure between the feed and permeate lines, which appear due to the frictional resistance over the membrane (Crittenden et al., 2012). Since the flux in a membrane process is dependent on pressure and temperature, the operational parameter specific flux is calculated, accounting for a standard temperature flux divided by the TMP. During cross flow, shear forces in the feed channel will cause a feed channel pressure drop, causing lower pressure in the retentate line than in the feed line. An additional pressure to consider during retention of ions and salts is the osmotic pressure, which is affected by the concentration polarization.

2.2.1 Membrane modules

Membranes are usually constructed as thin-film composites, meaning that the active membrane surface is very thin, stabilized by a thicker layer underneath (Lidén, 2020). The retention rate is determined by the thin-film, while the supporting layer is porous. This construction leads to a decreased need for pressure, contributing to making membranes more economical. The active membrane surface can be placed on the feed side, permeate side or on both sides, but most commonly it is placed on the feed side. The most commonly used material for both the membrane surface and the supporting layers are polymers. The material will constitute a resistance, affecting the transmembrane pressure. Moreover, Lankes et al. (2008) states that surface parameters of the membrane, such as morphology, hydrophobicity, roughness and charge will affect the performance of the membrane.

Originally, membranes were designed as flat sheet modules, where sheets of membrane are placed in a module in several layers separated by feed spacers (Lidén, 2020). This construction allows the membrane to manage relatively high pressures, while requiring much place due to low packing density. As a development of the flat sheet modules, spiral wound modules became an alternative, allowing a higher packing density. Spiral wound modules are constructed with sheets of active membrane surface and sealed layers separating feed water and permeate. The sheets are rolled and placed in a pressure resistant cylinder, with feed spacers separating the sheets from each other, allowing feed water and permeate to flow through separate channels. Due to the ability to manage high pressure, spiral wound modules are commonly used for dense NF and RO. In Figure 2.3, the construction of a spiral-wound RO membrane element from FilmTec^{TM} is shown.

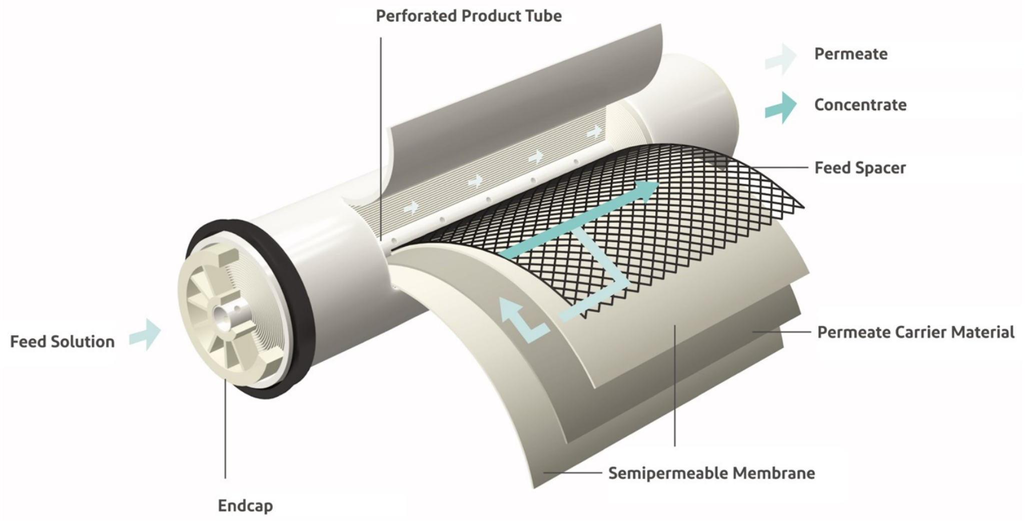


Figure 2.3: Spiral-wound RO membrane element from FilmTec, obtained from FilmTec^{TM} Reverse Osmosis Membranes Technical Manual (Dupont, 2023)

For UF, which does not require as high pressure, hollow-fiber modules are the most common module used for drinking water production (Lidén, 2020). This type of membrane module consists of a collection of thin hollow-fibers. The water is filtered when the water passes the membrane surface, which can be placed either on the inside of the fibers or the outside. Additionally, tube membrane is a type of module, consisting of fewer channels with bigger diameter compared to the hollow-fiber membranes. Both hollow-fiber membranes and tube membranes have adequate washing characteristics, where tube membranes can manage higher pressure and particle load, with the disadvantage that they require more space. A combination of hollow-fiber and tube modules is a relatively new type on the market, having better strength than pure hollow-fiber modules and better packing density than regular tube membranes.

For MF and UF, the membrane filtration process is commonly designed to operate in a cycle of two phases (Lidén, 2020). During the filtration stage, impermeable components are accumulated on the feed side, while permeable components passes through the membrane. During the second step, backwashing, the accumulated components and particles are flushed from the system and removed as retentate. Backwashing is an approach used to manage the decline in flux occurring due to the accumulation of particles close to the membrane surface. The loss of recovery rate for MF and UF membranes mainly originates in the washing and backwashing of the system. For processes based on NF and RO, which commonly uses spiral wound modules to manage high pressure, backwashing can not be performed as easily due to the risk of harming the feed spacers in the structure. The feed spacers in spiral wound membranes constitutes an additional risk, by compounding an obstacle for washing. Consequently, places where organic matter can stack are created, enabling biofilm to accumulate.

2.2.2 Fouling in membrane facilities

Fouling is a common problem within membrane facilities used in different sectors, including drinking water production (Zheng et al., 2024). The process of membranes fouling implies that particles, organics, colloids and solutes create formations generated by various mechanisms. Quality of the raw water source, the characteristics of the membrane material and the conditions during operation are affecting the fouling outcome. Fouling in membranes results in a lower flux due to a higher differential pressure which appears over the membrane. Further, this affects the function of the membrane, leading to shorter intervals between the replacement of the membrane and a reduced robustness of the system. As a consequence, the energy consumption and economic costs for the process increases. Membrane fouling results in a loss of the membrane permeability, which requires a higher operating pressure (Park et al., 2022).

The division of fouling types can differ, and in this report the fouling is divided into biological fouling, organic fouling and inorganic fouling, depending on what is causing the problem (Zheng et al., 2024). The different fouling mechanisms, illustrated in Figure 2.4 include pore blocking, pore constriction and cake formation (Zheng et al., 2024). Pore blocking and pore constriction are more likely to cause fouling problems in MF and UF membranes, whilst cake formation is more common for NF and RO.

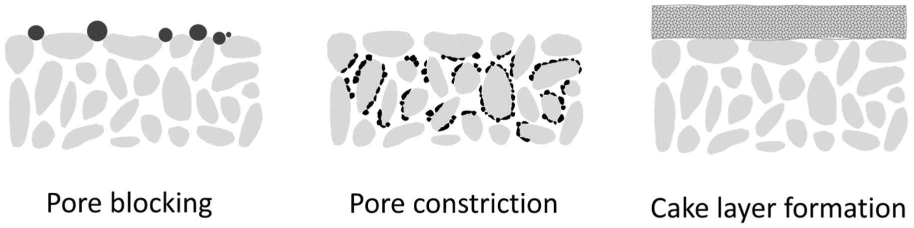


Figure 2.4: Fouling mechanisms that can occur during membrane filtration, including pore blocking, pore constriction and cake layer formation

Pore blocking occurs when compounds in the same size range as the pore size streams through the filter, resulting in a decrement of flow (Lidén, 2020). This mechanism is not commonly occurring in commercial membrane facilities since there are other possible flow paths because the membrane depth is much larger than the pore width (Crittenden et al., 2012). Pore constriction implies a reduced pore size caused by adsorption of particles and colloids. The adsorption take place both in the pores and on the membrane surface. The particles are transported via diffusion or hydrodynamic forces. In order for these processes to occur, the particles need to have certain properties which make them attach to the membrane surface. Cake formation is a result of the concentration polarization, which accumulates material from the incoming feed water on the membrane surface (Lidén, 2020). The cake formation has filter properties as well, which contributes to the retention effectiveness of the membrane process. However, the cake layer also affects the flow conditions by an increased resistance.

Fouling could be classified as reversible or irreversible, depending on how the fouling is managed (Lidén, 2020). The fouling is classified as reversible if the fouling can be removed during backwashing, which can be the case when particles cause the fouling. An influencing factor during reversible fouling is the concentration polarization existing on the membrane surface (Mallya et al., 2023). Further, a gradual loss of membrane performance can be observed during the operation, which occurs due to slow adsorption and an excess of components which are unable to get removed by backwashing (Crittenden et al., 2012). This type of fouling is classified as irreversible fouling and can partly be managed by adding chemicals (Lidén, 2020). Commonly, acids are added to the system aiming to reduce deposits of limescale and deposits of other metals, while alkali wash is used when the aim is to reduce deposits of organic matter.

To predict the fouling tendency of a membrane, the Silt Density Index (SDI), which accounts for the content of particulate matter in water, can be measured (Dupont, 2023). The SDI is obtained from calculating the filtration rate when water is fed through a 0.45 μm filter at constant pressure. The guidelines for membranes are to control the SDI to below 5, while an SDI below 3 is recommended to minimize fouling. Modified Fouling Index (MFI) is a more specific index, compared to SDI, used for RO and NF membranes. The colloidal and particulate fouling is assumed to be controllable at a MFI <1, which approximately equals to a SDI <3.

2.3 Fouling in nanofiltration

To understand the fouling process, it is important to consider the relation between the occurring contaminants, and their relation to the surface of the NF membrane (Zheng et al., 2024). Multiple factors affect the fouling outcome, including membrane properties and design, water quality of influent raw water and operational conditions (C. Fan et al., 2024).

2.3.1 Organic fouling

In order to successfully remove NOM from incoming water, several factors including NOM and solution characteristics, hydrodynamic conditions, MWCO and chemistry of the membranes needs to be studied (Fane et al., 2006). Size exclusion is applicable to NOM with molecular sizes in the same range as the membrane pores, and is therefore more common for removal of HMW hydrophobic NOM (Mallya et al., 2023). Hydrophobic NOM could also be removed via hydrophobic interactions. In addition, the process of size exclusion is potentially intensified by hydrogen bondings occurring between the surface of the membrane and NOM.

If the NOM possess hydrophobic or hydrophilic characteristics affects the removal of NOM (Mallya et al., 2023). A higher NOM removal is possible for hydrophobic NOM, since the hydrophobicity makes the NOM more prone to deposition or adsorption. The membrane could also be hydrophilic or hydrophobic, depending on the characteristics, the ability of detainment of NOM differs. For hydrophilic membranes, a layer of water molecules is created at the membrane surface, resulting in a high removal of hydrophobic NOM (Huang et al., 2021). In contrast, hydrophobic membranes show greater effects in rejection of hydrophilic NOM (Mallya et al., 2023). During the membrane operation, the hydrophobic/hydrophilic properties changes. Another factor affecting the NOM removal is electrostatic repulsive forces which exists between NOM and the membrane surface. The negatively charged build-up of NOM on the membrane surface could affect the surface charge, and as a result, the removal of organic matter increases. The effect of charge is vital for the removal of LMW hydrophilic NOM.

NF is used to address the issue of NOM present in raw water sources, as a result NOM is decomposed onto the membrane surface resulting in organic fouling (Zheng et al., 2024). NOM is the most common contributor to fouling for treatment of surface water. According to Uhl et al. (2007), biopolymers which includes aminosugars, proteins and polysaccharides has been identified as a major contributor to flux decline. Uhl et al. (2007) also mention the the HMW hydrophobic fraction as another contributor to flux decline. Crittenden et al. (2012) also states that HMW constituents probably one of the biggest contributors of fouling in NF. In addition, Crittenden et al. 2012 mention the problematic effect of colloidal NOM. For high pressure fouling, including fouling in nanofiltration, the fouling usually occurs on the surface, creating a cake formation (Zheng et al., 2024). The process of NOM fouling implies a cake formation resulting in an intensified concentration polarization which affect the membrane efficiency (Park et al., 2022).

Several mechanisms contribute to the formation of organic fouling (Zheng et al., 2024). A possibility is attachment of NOM to the membrane surface resulting in electrostatic forces, this affects the membrane surface properties. Further, adsorption of NOM in the pores and on the membrane surface results in pore blocking behaviour, it also affects the charge and hydrophobic properties. As a consequence, the membrane flux decreases. A common functional group for NOM is carboxyl groups, these groups appeal with ions which leads to the creation of a cake layer. In addition, a gel layer can form under the influence of concentration polarization, which could lead to irreversible fouling. In a multistage NF system, NOM and large particles usually accumulate in the primary steps of the system (C. Fan et al., 2024).

2.3.2 Biofouling

Biofouling can be viewed as a biotic form of organic fouling, occurring as a result when microorganisms, plants, algae and animals, accumulate on wet surfaces (Ismail et al., 2019). Water columns obtaining parameters indicating on an abundance of microbes, nutrient availability and favorable environmental conditions for microbial growth, possess the potential of biofouling. Microorganisms exist in all water systems and biofouling starts to develop as microbial cells attach to membrane surfaces, where they grow at the expense of nutrients available in the water column or organic matter adsorbed on the membrane surfaces. Biofilms, consisting of microorganisms such as bacteria, protozoa, algae and fungi etc, are formed as the attached microorganisms excrete extracellular polymeric substances (EPS), in which the populations of microorganisms are embedded.

All surfaces in spiral wound membrane modules hold a risk for accumulation of biomass as a result of complex interactions between microorganisms, dissolved substances in the water, the material of the membrane and flow parameters (Ismail et al., 2019). Vrouwenvelder et al. (2009) declared that biofouling in the feed spacer is a complication of greater importance than membrane biofouling. To control biofouling, it is essential to remove the biomass accumulated on the elements of the membrane module. Studies showed that membrane cleanings could inactivate biomass to a certain extent, but difficulties to remove the biomass remained (Vrouwenvelder et al., 2008). The residual inactivated biomass serves as a food supply for both the remaining active microorganisms within the system and those introduced by the feed water. As a consequence, rapid regrowth of biomass can occur.

2.3.3 Inorganic fouling

Inorganic fouling is a result of deposition of minerals, colloidal matter and hardness scales from inorganic salts, present in the feed water (Lin et al., 2020). The presence of ions in the feed water is significant for the occurrence of inorganic membrane fouling. Due to the concentration polarization occurring in NF, high concentration of dissolved salts exists close to the membrane surface (Shirazi et al., 2010). When the solubility product of ions is reached or exceeded, the salt precipitates on the membrane surface. Gradually, microcrystals are formed, which progressively results in inorganic fouling (Zheng et al., 2024). The presence of divalent cations has been found to increase the risk of membrane fouling. Further, alkaline earth metal cations, for instance Ca^{2+} and Mg^{2+} ions, lead to additional risk of severe fouling when coexisting with polyanions, such as carbonate (CO_3^{2-}), sulphate (SO_4^{2-}) and phosphate (PO^{3-}) (Jarusutthirak et al., 2007). Shirazi et al. (2010) accounts for some of the most common inorganic salts causing scaling being $CaSO_4$, $CaSO_3$, SiO_2 and $BaSO_4$.

The concentration polarization promotes precipitation scales from inorganic substances such as calcium, magnesium, iron, silica etc., while the hardness ions further form a gel layer (Lin et al., 2019). In addition, inorganic fouling occurs due to colloidal particles affecting the structure and permeability of the gel and cake layer. The most frequent inorganic colloids causing fouling when nanofiltration is used for drinking water production are aluminium silicate clay, iron, calcium, aluminium and silicon colloids (C. Fan et al., 2024).

In anaerobic groundwater, which often is used for NF facilities, iron and manganese exist in their reduced soluble state (Crittenden et al., 2012). When the groundwater is exposed to oxygen, iron and manganese can oxidize and precipitate in the water system, constituting a risk of membrane fouling. Fouling caused by iron is more frequently occurring, thus iron is prone to oxidize when air enters the system, forming iron hydroxides possessing very low solubility. In studies performed by Beyer et al. (2014), nanofiltration plants performed more unstable for feed waters classified as oxic groundwater than anoxic groundwater. Although, fouling can occur rapidly when anoxic groundwater is exposed to oxygen, and oxidation should be prevented to reduce the risk of fouling (Crittenden et al., 2012). Another approach to manage fouling by iron and manganese is to allow oxidation and remove the components prior to the membrane. This alternative may be favourable when high concentration of iron occurs ($> 50\mu g Fe/L$), due to the risk that oxidation can cause irreversible damage to the membrane surface (Ismail et al., 2019).

Electrostatic repulsion is the central mechanism of rejecting inorganic salts (Thanuttamavong et al., 2002). The membrane surface carries a charge varying between positive, negative or even neutral, when exposed to different pH conditions. The rejection characteristics of the membrane will therefore be dependent on the molecular charge of the target solutes. This electrostatic interaction between the membrane surface and the contaminant affects the process of membrane fouling (Zheng et al., 2024). If the surface charge is equivalent to the charge of the contaminant, electrostatic repulsion will occur, leading to reduced adhesion and deposition of contaminants. As a result, the membrane fouling will be delayed. Due to the fact that most contaminants in the raw water column carry a negative charge, an effective approach to enhance anti-pollution of the membrane is to increase the electrostatic repulsion by enhancing the negative potential on the membrane surface. Thanuttamavong et al. (2002) investigated the average rejection of inorganic pollutants by NF membranes in long-term operations, showing that divalent ions such as Ca^{2+}, Mg^{2+} and SO_4^{2-} gave higher rejection than monovalent ions such as Na^+, K^+, Cl^-,

NO_3^- and HCO_3^-.

To manage the formation and precipitation of crystallized mineral salts, antiscalants can be added to the process (Ismail et al., 2019). The antiscalants can be used to prevent salt precipitation on membranes in three ways. Firstly, antiscalants can inhibit of the threshold to maintain the supersaturation of salt in the water. Secondly, antiscalants can alter the shape of the salt crystals. Lastly, antiscalants can project high anionic charges on the surfaces of colloids and particles, aiming to keep the crystals separated. It is of high importance that the dosing of antiscalants are proper, since underdosing will not interfere the salt precipitation enough, resulting in scaling and fouling. In contrast, overdosing risks to create a deposition of antiscalants on the membrane, also resulting in fouling. In Sweden, the allowed dosage of antiscalants are restricted to 5 mg/l in LIVSFS 2022:12 (Swedish NFA, 2022).

2.3.4 Interactions between foulants

The division of membrane fouling caused due to organic, biological or inorganic origin is an approach to gain theoretical understanding of the fouling mechanisms. In practice, membrane fouling in NF facilities for drinking water production are significantly more complicated (Guo et al., 2024). Since various fouling types can take place at once, interactions of the foulants arise and creates synergy effects affecting the fouling extent (Vrouwenvelder et al., 2008). A possible interaction is the combination of organic and inorganic content present in the water. Interaction occur due to the chemical and psychical characteristics of the concerned substances and include complexation, adsorption, or exclusion (Gao et al., 2023). Organic and inorganic content comprise a wide range of different substances, resulting in a wide range of outcome interactions.

Membrane fouling occurs when the functional groups present in the humic acids come in contact with inorganic content. The functional groups of humic acids in combination with silica, Ca^{2+}, or Mg^{2+} result in bridging of the molecules in the humic acid creating accumulation leading to membrane fouling. The structure and characteristics of the humic acids get affected by the inorganic interactions. According to Gao et al. (2023), Mg^{2+} and Fe^{3+} have a higher charge density compared to Ca^{2+} as a result, the interaction effect is less extensive resulting in decreased fouling problems. Through adhesion, silica interacts with humic acids, which leads to the creation of larger structures which accumulate on the membrane.

For polysaccharides, transparent exopolymer particles is the main contributor to fouling (Gao et al., 2023). Transparent exopolymer particles binds to inorganic ions, creating a larger structure which is strongly abundant in water. The inorganic ions which binds to the carboxyl groups in the polysaccharides and create transparent exopolymer particles are following; Ca^{2+}, Mg^{2+}, Al^{3+}. Moreover, silica interacts with both carboxyl and hydroxyl groups. Y. Wang et al. (2023) also concludes that Ca^{2+} combined with organic content contributes to a higher fouling severity.

An additional aspect to consider when viewing nanofiltration in a drinking water treatment system, is the interactions that can occur between the components in the raw water and additives from possible pretreatment methods. Zheng et al. (2024) mention residual flocs of metal ions, such as Al^{3+} and Fe^{3+}, as an additional component risking causing fouling when nanofiltration is applied in a treatment chain. Further, Guo et al. (2024) identified residual aluminium from coagulation as a key foulant during bench- and pilot tests performed on a NF. When aluminium occurs in active form, Al-NOM complexes can be produced, due to interactions with fulvic and humic substances, in particular but not exclusively. In addition, P could be identified in the fouling layer. Thus, Al and P concentrations usually occur in low concentrations in surface water, the occurrence of the components in the fouling layer is believed to be caused by residual Al-coagulant and phosphate-containing antiscalants added to the feed water.

2.3.5 pH and temperature

Water parameters such as pH and temperature affects the membrane performance and the occurrence of fouling. A decrease in temperature affects the contraction of the membrane pores as well as increasing the viscosity of water (Zheng et al., 2024). The pH has a significant effect on the charge of the nanofiltration membranes. Neutral or alkaline environments primarily generates negative charge on the membrane, while the membranes possess a positive charge during highly acidic conditions (Mallya et al., 2023). Low pH conditions causes a reduction of the contaminant's surface size and an increase in hydrophobicity (Zheng et al., 2024). These factors enhance deposition of contaminants on the surface of the membrane, due to a decrease of the electrostatic force between the surface's charges. The deposition leads to blockage of the membrane pores, causing fouling. In addition, the membrane's adsorption of DOM increases during low pH conditions (Crittenden et al., 2012).

Temperature and pH also have an effect on the fate of NOM. The various groups of NOM have different solubility at different pH levels. Humic acids are soluble in water at pH values higher than two, while fulvic acids are soluble at all values (Darko et al., 2014). In contrast, humins are not soluble at any pH value. Temperature affect the solubility as well, a higher temperature results in a higher solubility (Shoukat et al., 2020). As mentioned earlier in the organic fouling section, pH influence the ionization of the functional groups present in the NOM molecules, resulting in the surface charge (American Water Works Association., 2011). During low pH conditions, the particle charge is positive, while the charge is negative at higher values. In addition, natural particles usually have a negative charge in most conditions.

Waters with pH levels ranging from 6.5 to 8.5 are generally the most favorable conditions for bacteria to thrive, but most bacteria can operate during pH conditions ranging from 4 to 9 (Crittenden et al., 2012). Further, some microorganisms can tolerate values outside this range. Mostly, biological activity increases with a higher temperature, leading to an increased degradation of NOM present in the water (Zeng et al., 2021). The chemical reactivity also gets boosted by rising temperatures. For NOM concentrations, the DOM fraction increases.

The solubility, precipitation and reaction rate for salts and ions varies during different pH and temperature conditions. In general, lower pH values increases the solubility of the metal, whereas metals have a tendency to precipitate during high pH conditions (Saalidong et al., 2022). In addition, low pH conditions in water systems can cause corrosion of metal pipes. As temperature increases, so does the solubility, mobility and reaction rate for most inorganic components.

The stability of colloidal contaminants in the water column decreases as a result of high ionic strength combined with low pH levels (Zheng et al., 2024). This allows the colloids to form gel at the membrane surface, contributing to the concentration polarization. Mallya et al. (2023) further explain that a combination of high ionic strength and low pH levels contributes to severe fouling.

2.4 Pretreatment methods for NF

Pretreatment is usually required for nanofiltration to function well, especially when treating surface waters. In addition, pretreatment can be essential to protect the membrane from damage. In this section, possible pretreatment methods to implement in a MDWTS are presented.

2.4.1 Microfiltration

A MF membrane consist of symmetric pores in the size range 0.05-10 μm (R. Singh, 2014). The size of the pores enhance removal of particles, sediment, algae, bacteria and protozoa, while small colloids, viruses, DOM and ions transmit with water through the membrane. Due to the deficient removal of viruses, MF does not count as a microbial barrier according to LIVSFS2022:12 (Swedish NFA, 2022). MF membranes have historically operated in dead-end mode for small volume applications, with the advantage of operating solely with a feed stream and a permeate stream (R. Singh, 2014). Due to the ability to backwash MF membranes, the treatment method gained popularity in large scale applications, thus it manages treatment of big water volumes with high turbidity and removal of pathogens, such as Cryptosporidium and Giardia cysts. Further, MF membranes have the advantage of being easily operated under relatively low pressure, around 1-2 bar.

Additionally, MF membranes can be implemented as pretreatment to improve the performance of NF and RO, having a lower footprint than conventional pretreatment. Using MF as pretreatment prior to NF was investigated by Lee and Lee (2006) for a surface water, which showed that the NF flux increased with a decreasing nominal pore size of the MF. It was further shown that cake resistance on the NF decreased using MF, which was explained by the MF's ability to remove large particles. However, the external fouling resistance on the NF, occurring due to irreversible surface deposition of small particles and colloids, was not reduced. Compared to operating the NF without pretreatment, the total resistance on the NF and the SDI decreased with 25-50% respectively 28-50%, when applying MF prior to the NF.

Further, Lee and Lee (2006) found that a MF membrane within the pore size range of 0.08-0.45 μm reduced the turbidity to below the detection limit of 0.1 NTU. Another study performed on surface water showed a reduction of turbidity by 94% when applying MF (Podaru et al., 2008). Podaru et al. (2008) further comments that no reduction of the hardness ions was observed for neither MF nor UF and that an oxidant or coagulant is required for MF to remove iron and manganese.

2.4.2 Ultrafiltration

UF membranes are porous, with asymmetric pores in the size range of 0.001-0.05 μm and operational pressure requirements are generally 2-5 bar. Through size exclusion of bigger compounds than the mentioned sizes, UF possess the ability to retain suspended and colloidal matter, macromolecules, protozoa, bacteria and most viruses

(R. Singh, 2014). Without the use of a coagulant, the retention of NOM is limited (Lidén, 2020). UF possess the advantages of being a compact process with easy automation, operation and maintenance, while being independent of the microbiological activity in the feed water (Arnal et al., 2009). When applying UF, it is important to consider the risk of fouling and the relatively high discharge (10-15%) of water the process generates. In addition, the water used for cleaning the membrane may require treatment before being discharged. When used for drinking water production, UF can be applied in different parts of the treatment chain, such as pretreatment, main treatment or final treatment to remove protozoa, bacteria and viruses as disinfection.

It has been shown that pretreatment with UF can improve NF flux and mitigate NF membrane fouling. In a study performed on surface water, HMW NF foulants were efficiently rejected by UF, while the retention of humic and fulvic acids were low (Lee and Lee, 2006). When UF was applied as pretreatment prior to NF, the cake resistance and external fouling resistance of NF decreased. The remaining portion of cake resistance was thought to owe to small colloidal particles and dissolved organics which passed through the UF membrane. In comparison to using UF as pretreatment with solely using NF, the resistance was decreased to about 10-20%. In addition, the SDI was reduced to under detection limit when using UF as pretreatment, implying that nearly all particles and colloids were removed.

Another study, made on surface water, showed that only partial rejection of TOC occurred when applying UF (Mijatović et al., 2004). In accordance with the previous mentioned study, UF was shown to be deficient in removing humic substances. A TOC rejection of 22% was observed when UF was applied, and the potential of increasing the removal efficiency by adding a coagulant in the feed water was mentioned. Sun et al. (2012) stated that UF alone showed a removal of 62.2% for turbidity, 68.7% for DOC and 62.2% for Fe^{2+}. It was also stated that UF resulted in an increase of NF permeate flux, from 46% to 95%.

The utilization of UF as pretreatment has also been investigated for groundwater in a study performed by Kasim et al. (2016). When feeding the treatment chain, consisting of UF prior to NF, with synthetic and natural groundwater, a removal of 92-99% of the influent Fe^{2+} and 68-94% of the influent Mn^{2+} was observed. Additionally, a synthetic water where iron and manganese coexisted with humic acids was investigated, resulting in removal of 95% of the influent Fe^{2+} and 45% of the influent Mn^{2+}. However, it was not shown how this affected fouling of the NF.

2.4.3 Rapid sand filtration

Rapid sand filtration (RSF) is a mechanical filtering process where water is passed through a filter of granular media by pressure or gravity flow (Crittenden et al., 2012). Suspended and colloidal components in the water are retained in the pores between the sand and by attaching to the surfaces of the sand grains. As a result, particles which are less than the openings between the sand grains are removed. RSF affects the turbidity, odour, taste and organic matter. Further, RSF possess a high efficiency and flexibility to treat waters with different turbidity levels. In general, RSF is used in centralised water treatment plants, where it is used after the conventional treatment chain or directly after coagulation.

The filtering process usually occurs from top to bottom, with a flow rate of 5-25 m/h (Svenskt Vatten, 2010a; Akinwunmi and Adebanwi, 2024). In order to proceed the process of an open RSF, a flow rate of 5 m/h is preferable. If higher flow rates are desired, the turbidity of the raw water has to be low or an increase of filtration resistance is needed. A common height of the filtering bed is 1m. Backwashing is needed after 1–7 days, and can be done with water, requiring about 10-15% of the influent water, or air (Crittenden et al., 2012). Hoslett et al. (2018) claim that RSF is an alternative for SSF when the land use is a limiting factor. RSF alone does not mitigate the key foulants significantly, as a consequence RSF requires prior and/or post treatment to prevent NF fouling.

Used as pretreatment to large scale SWRO facilities, due to the robustness of the method to decrease turbidity/remove particles larger than 0.35 mm. In these facilities, coagulation and flocculation are often used prior to the RSF to enhance removal of particles. Biological impact on water passing through RSF is minimal (assumption) (Bar-Zeev et al., 2012) Monitoring the microbial maturation process in an RSF. By monitoring and optimization, RSF had potential to function as a biofilter after two and half months. Stage one single medium, removal of TOC 17 % at day 7 and 37% at day 27. Stage two 55% removal at day 42, 21% at day 65 dual medium. Decrease of removal efficiency assumed to be due to influent water quality.

2.4.4 Activated carbon (AC) filtration

Activated carbon (AC) filtration consists of an active material which is highly porous and works as a powerful adsorbent (Korotta-Gamage and Sathasivan, 2017). Several properties of the filter media surface, including the chemistry, pore structure and surface area, affect the outcome of the adsorption ability. Further, AC can be divided into granular activated carbon (GAC) and powdered activated carbon (PAC). GAC is used to remove toxic organic compounds, compounds causing taste and odour, disinfection by-products precursors and DOC (Crittenden et al., 2012). PAC also has the ability to remove taste and odour, but to a less extent than GAC. Further, PAC adsorbs pesticides and low concentrations of herbicides.

Regarding the operational aspects, GAC is more preferable since it requires a lower carbon usage rate per volume of treated water compared to PAC. A disadvantage of GAC is the reduction in adsorption capacity which occur after a time of operation, which could be explained by the shortage of adsorption sites (Korotta-Gamage and Sathasivan, 2017). When these sites are occupied, the filter gets exhausted. As a result, organic compounds in the water are able to get by the filter, which has severe consequences for the water quality. In order to regenerate the adsorption efficiency, the filter has to be restored or regenerated (Korotta-Gamage and Sathasivan, 2017). GAC is more easily reactivated compared to PAC (Crittenden et al., 2012). It is not possible to remove all the materials adsorbed in the filter media, and therefore the filter has to be restored at some time (Korotta-Gamage and Sathasivan, 2017). According to Svenskt Vatten (Svenskt Vatten), the effect of the AC reduces significantly after 1–3 years. For organic content, the GAC removal effect reduces significantly already after three months of use.

AC is most commonly applied after chemical flocculation or posterior conventional treatment (Svenskt Vatten, 2016). It has also been shown that AC can be implemented as a pretreatment method prior to membrane filtration, with the potential to mitigate fouling for UF, NF and RO (Gur-Reznik et al., 2008). In a study conducted by Sun et al. (2012), the removal efficiency of GAC as pretreatment before NF was investigated through adsorption tests in an up-flow plastic column. For turbidity, DOC and Fe^{2+} the removal efficiencies obtained were 21.2%, 17.8% respectively 21%.

2.4.5 Biological filtration

The process of biological activated carbon (BAC) filtration involves an AC filter, which function as a media for growth of microorganisms (Korotta-Gamage and Sathasivan, 2017). At the beginning of the process, physical and concurrent adsorption are the main removal mechanisms. Further, biological degradation occur due to the present of organic matter and nutrients in the filter media, creating a biofilm. In this step, biological degradation is the main removal mechanism, and the adsorption decreases. BAC reduces the concentration of manganese, iron, LMW NOM, trace level organics and the taste and odour causing compounds.

Multiple physiochemical properties affect the adsorption of bacteria in the filter media. The retention ability increases with a larger surface area and pore volume (Quintelas et al., 2010; Korotta-Gamage and Sathasivan, 2017). Surface hydrophobicity is favourable for the adsorption of bacteria (van Loosdrecht et al., 1990; Rijnaarts et al., 1996). Further, bacteria adsorbs to metallic oxides present in the water (Jucker et al., 1997; Rivera-Utrilla et al., 2001). During biofilm formation, the negative surface charge of the filter increases (Korotta-Gamage and Sathasivan, 2017). As a result, the filter is capable to attract positively charged compounds.

A bioreactor filled with sand, e.g. DynaSand Oxy, is another type of biological filtration. DynaSand Oxy is an upstream bed filter, applied to reduce taste and odour causing compounds, BOD, turbidity, iron and manganese (Nordic Water, 2023). Raw water enters the inlet at the top of the reactor and are led down to the bottom, filled with sand. The cleaning process takes place as the water moves upwards through the reactor. For the DynaSand Oxy an aerator is installed as well to enhance the reduction of iron and manganese.

An important parameter for biological filtration processes is the empty bed contact time (EBCT), which is defined as the duration a bed volume of water stays in contact with an adsorbent such as carbon or sand (Korotta-Gamage and Sathasivan, 2017). A longer EBCT results in a higher removal rate, which can be accomplished by decreasing the flow rate or increasing the water bed volume. However, a longer EBCT leads to greater footprint and more material use. Svenskt Vatten (2010a) states that a EBCT between 15–30 minutes is common, but that a longer EBCT is beneficial.

Several parameters such as oxygen levels, pH, micronutrients and nutrients concentrations are important to monitor the biofilm in the biological processes (Korotta-Gamage and Sathasivan, 2017). The growth rate of microorganisms is strongly affected by the oxygen concentration and pH value of the incoming water. According to Scholz and Martin (1997), a high oxygen concentration > 8 mg/L and a low pH value between 5.5 to 6.5 is beneficial to create and maintain the bioactivity in the filter media. Another factor to consider is the temperature dependency of biological processes. According to Svenskt Vatten (2010a), the biological activity decreases with 50% when the temperature decreases from 10 to 0 degrees (Svenskt Vatten, 2010a). On the contrary, higher temperatures 15–20 degrees requires higher flows for backwashing the filter. Material possess different adsorption capacities, for activated carbon has a higher adsorption capacity than sand and anthracite (Korotta-Gamage and Sathasivan, 2017).

Accumulation of organic, inorganic compounds and microorganisms results in clogging of the BAC filter media (Korotta-Gamage and Sathasivan, 2017). The clogging of the filter leads to a decrease in efficiency and eventually results in a replacement of a new filter. The clogging effect occurring in the filter media can be prevented by periodic backwashing. In comparison to a BAC filter, bioreactors are continuously backwashed during the treatment process (Nordic Water, 2023). Bioreactors possess a surface load between 12-15 m/h, whilst BAC filters have a surface load of 5 m/h (Svenskt Vatten, 2010a). As a result, bioreactors have a lower footprint. Another aspect to consider is that the effluent water from both BAC filtration and bioreactors requires further treatment, such as a rapid filter, since it contains microorganisms, particles and EPS (Korotta-Gamage and Sathasivan, 2017).

Uhl et al. (2007) states that BAC as a pretreatment method can reduce organic-, inorganic-, and biological fouling in a NF membrane. Compared to a rapid media filtration, the TMP increase for a BAC filter was significantly low. Sun et al. (2012), investigated the removal efficiency of BAC with the same methodology as for the GAC tests. BAC by itself removed 22.8% of the turbidity, 29.2% of the DOC and 22.8% of the Fe^{2+}. BAC was also tested in combination with UF, this combination showed the great effect of fouling mitigation. In another study conducted by Yang et al. (2023), the effect of BAC as a pretreatment step before UF was investigated. To conclude the study, it was obtained that BAC reduced the transmembrane pressure by 49%, decreased the irreversible membrane fouling with 78.7% and limited the formation of DBPs.

In a pilot study (unpublished) performed on surface water, the removal potential of organic content, turbidity, manganese and iron for a bioreactor with sand was investigated. Since the study was performed during a year, the obtained results varied. As an effect of decreasing temperatures during the winter, the biological activity was reduced, which affected the removal mechanisms. After a stabilisation period, the turbidity removal was >50%. For the organic content, the highest obtained reduction was 10%. The majority of the degraded organic content is easy-degradable. When the levels of manganese were at its highest, a reduction of 60-80% was observed. The removal rate reduces during winter, however the ingoing levels of manganese reduces as well. According to the study, both particulate and dissolved manganese was reduced to low concentrations in the reactor. The dissolved manganese is gathered by the biomass. For iron, the reduction is approximately 10-30%. In comparison to manganese, the dissolved iron is not bonded to the biomass. As a result, the removed iron is particulate.

2.4.6 Ion exchange

Ion exchange (IEX) is primary used for water softening and demineralization of groundwaters (Crittenden et al., 2012). The process involves the use of natural or synthetic resin, typically composed of polymeric materials. The resin contains fixed charged functional groups covalently bonded to the external and/or internal surfaces of the resin. The functional groups carry ions with opposite charges, known as counterions, aiming to reach electroneutrality within both the exchange material and the surrounding solution. As water flows through a fixed bed of resin, ions present in the raw water are exchanged for the insoluble ions bound to the functional groups. The exchange capacity and selectivity of the resin can differ and depending on the charge of the functional group, the counterion can either be a cation or an anion. When the counterion is a cation, exchange of ions such as Ca^{2+}, Mg^{2+} and Na^{+} is enhanced, while ions such as Cl^{-}, SO_4^{2-} and NO_3^{-} are exchanged when the counterion is an anion. In addition, anion exchange is capable of removing DOM (Apell and Boyer, 2010).

The exchange continues until the desired treatment objective is achieved or until the resin becomes saturated with accumulated ions (Crittenden et al., 2012). Once the resin is saturated, it undergoes a regeneration process to restore the resin's capacity for further ion exchange. This involves adding a regenerating agent, typically various salts or acid solutions, which is capable of displacing the accumulated ions on the resin. The regenerating agent is passed through the resin bed, typically in the same flow direction as the solution being treated (commonly downwards), or in the opposite direction, depending on the specific requirements of the process. When applying IEX, Chaturvedi and Dave (2012) proposes cautiousness in cases where high concentrations of iron and manganese occurs, thus these risks to rapidly clog the process when exposed to oxygen.

While Crittenden et al. (2012) indicate that the IEX process is not widely used in large-scale water treatment plants, Amini et al. (2015) indicate that due to the scalability of the process, IEX can be employed in centralised as well as decentralised treatment systems. Further, IEX is described as a flexible method that can operate during different operational modes, for various water qualities and treatment objectives, as well as regarding positioning in a treatment chain. Several studies have been made on hybrid solutions of IEX and membranes processes in water treatment. In a review, Abdulgader et al. (2013) investigate the utilization of IEX as pretreatment prior to NF membrane filtration to prevent fouling, showing potential of reducing the risk of organic and inorganic fouling. However, this conclusion was mainly based on results from short-time laboratory tests.

In another study, the efficiency of a process combining magnetic anion and cation exchange resin was investigated, applied on a groundwater containing high levels of DOM and hardness (Apell and Boyer, 2010). The results in the study showed a removal of 70% of the DOC and $> 55\%$ of the hardness in the groundwater when combined ion exchange treatment was applied. Additionally, the theoretical fouling potential was calculated, showing that the individual cation and anion exchange treatment reduced the potential, while combined ion exchange treatment could reduce the fouling furthermore. While the fouling potential likely correlates to the removal of key foulants, it is not possible to draw conclusions without testing the hybrid process in full-scale or pilot-scale. This issue is addressed by Humbert et al. (2007) in a study of IEX as pretreatment prior to UF. While treating the feed water with IEX removed 80% of the DOM, the fouling rate of the UF was unaffected.

2.4.7 Chemical flocculation

The process of coagulation starts with a chemical coagulant, which is added in order to destabilize the solution or suspension. Dissolved, colloidal and suspended materials present in the water are affected by the process (Bratby, 2016). During coagulation, two mechanisms, including sweep coagulation and charge neutralisation, occur simultaneously (Svenskt Vatten, 2010a). The charge of colloids is usually negative, resulting in a zeta potential larger than the van der waals bondings. The aim of the coagulation is to neutralize the negative charge and enable the creation of flocs. The mechanism of sweep coagulation gather colloids and particles in the created coagulant floc. The addition of a coagulant implies the start of the flocculation (Bratby, 2016). Flocculation includes the formation of larger agglomerates and flocs consisting of the destabilized particles and the particles created during coagulation. If the water temperature is too low and the water viscosity is too high, a coagulant aid may be required in order to proceed the process (Svenskt Vatten, 2010a). The effluent water after coagulation and flocculation undergoes further treatment to remove the created flocs, either by filtration or sedimentation.

Traditionally, the process consists of three units for flocculation, sedimentation/flotation and rapid filtration. However, this requires large volumes and results in low surface loads. In order to minimize the footprint, units which combine different steps of the process have been evaluated (Svenskt Vatten, 2010a). The combination of flocculation and separation in the same unit is called contact filtration. The chemicals are dosed directly to the pipe, and the flocculation occur in the water above the filter material. The flocculation time for this process is between 4–10 minutes, compared to 30–60 minutes for the conventional process.

Since raw waters contain a wide variety of different constituents, it is hard to optimize and operate the process of coagulation and flocculation (Bratby, 2016). Several parameters affect the performance of the process such as the water composition, coagulant properties and how the coagulant is added. Different coagulant are used depending on the goal of the treatment, properties of the water, cost- and environmental factors. Iron- and aluminium-based coagulants are commonly used, since they have good adsorption properties and are relatively cheap. For aluminium-based coagulants the optimal pH is 5.5-6.5 whilst iron-based coagulants are most effective at a pH of 5-5.5 (Svenskt Vatten, 2010a). As a result, more chemicals are required for the process of precipitation with iron-based coagulants.

According to Svenskt Vatten (2010a), chemical flocculation or membrane filtration has to be implemented for raw water with high NOM content. A majority of the constituents that exist in surface water possess a particle size which is too small to be separated with granular filtration. Chemical flocculation is especially effective for removal of HMW hydrophobic NOM, and after the process LMW NOM are still present in the water. Iron-based coagulants enhance the precipitation of NOM. As the pH decreases, the positive charge of the $Fe(OH)_2$ increases, resulting in a neutralisation of the NOM charge. This especially applies for hydrophobic NOM, such as humus and fulvic acids.

In a paper, Sillanpää et al. (2018) present several studies where iron- and alum-based coagulants were compared (Sillanpää et al., 2018). For $Al_2(SO_4)_3$ the optimum pH for the different jar tests varied from 5-7, resulting in DOC removals between 43.2%-83%. When $FeCl_3$ was used as coagulant, the DOC removal varied between 51-57.9%. Zhao et al. (2014) stated that $Al_2(SO_4)_3$ resulted in a lower flux decrease compared to $FeCl_3$. Jarvis et al. (2012) achieved a turbidity reduction of 86% when using $Al_2(SO_4)_3$ as coagulant.

In an experimental analysis made by J. Wang and Wang (2006), UF in combination with in-line coagulation was compared to UF alone, in order to remove natural humic acid. At a pH value of 7, this process increased the DOC removal from 28% obtained with UF alone to 53%. More specifically, the in-line coagulation and UF combination was effective for the removal of humic acid with a molecular weight less than 6 kDa. In addition, after 10 hours, the TMP did not increase significantly compared to UF alone, which resulted in a TMP increase. For the in-line coagulation and UF the permeated flux was relatively constant whilst UF alone resulted in a decline of 75%. In another experiment made by Listiarini et al. (2010), the combination of hybrid coagulation and NF was tested with the purpose to remove humic acid. Both NF270 and NF90 were tested, for the coagulant alum and ferrous were used. This method achieved a high removal rate of humic acid, the permeate flux was almost unaffected.

Chemical flocculation can also be used to remove heavy metals such as zinc, lead and iron (Pang et al., 2011). If the level of colloidal silica or silicates are too high, chemical flocculation is a great removal alternative (Dupont, 2023). Sun et al.

(2012) tested coagulation by itself, in combination with UF and in combination with BAC. For coagulation alone, the removal efficiencies for turbidity, DOC and Fe^{2+} were 45.6%, 59.6% respectively 45.6%. When combining coagulation with UF the removal of turbidity, DOC and Fe^{2+} increased to around 70%. The same result applies for the combination of coagulation and BAC. Further, these combinations resulted in an increase of the permeate flux from 46% when using UF alone to approximately 95%.

The complex process of chemical flocculation requires regularly operation and maintenance work. In addition, there is a need for storage of chemicals (EPA, 2000). A major disadvantage of the process is the risk of residuals causing fouling. It is important to control the alkalinity in order to maintain a stable process (Svenskt Vatten, 2010a). If the alkalinity is too low, the removal mechanism reduces and the risk of residual chemicals in the drinking water increases. In a study conducted by Ohno et al. (2010), the effect of residual aluminium sulphate and ferric chloride was evaluated. The membrane permeability was decreased due to the use of aluminium coagulant. In comparison to ferric chloride, which did not show any significant permeability decrease. However, the concentration of residual is important to consider, Ohno et al. (2010) observed that the permeability decreased when the water contained 18 mikrog/l or more. Further, the process of chemical flocculation results in creation of sludge which needs to be handled (Svenskt Vatten, 2010a).

2.4.8 Lime softening

Softening can be achieved with chemical precipitation (Crittenden et al., 2012). The softening process is based on the removal of calcium, through calcium carbonate precipitate ($CaCO_3$) and magnesium, through magnesium hydroxide precipitate ($Mg(OH)_2$). It is a complex process, which requires jar tests to determine the optimum dosage of chemicals (EPA, 2000). Overdosing can result in the opposite effect. Both $CaCO_3$ and $Mg(OH)_2$ requires increased pH-values (Crittenden et al., 2012). More specifically, to remove $Mg(OH)_2$ efficiently, a pH of 10.5 is favourable.

The choice of chemicals used for precipitation mainly depends on the quality of the raw water. For softening, lime and caustic soda are often used. Depending on if the carbonate hardness is sufficient or not, different approaches are used. If the carbonate hardness is not the problem, softening can be done with pH-adjustment. As a result, both $CaCO_3$ and $Mg(OH)_2$ are precipitated. Further, if the carbonate hardness is too low, addition of soda ash/sodium carbonate is needed. When there is a high noncarbonate hardness, it can be beneficial to use ion exchange instead of chemical precipitation softening.

Lime softening can also remove heavy metals, iron, manganese, turbidity and NOM (Crittenden et al., 2012). According to U.S. EPA, the use of lime softening can result in a TOC removal of 40-80%, strongly affected by pH.

2.4.9 Oxidation

In drinking water treatment, oxidation is a commonly used method due to its ability to enhance the removal of organic and inorganic chemical constituents that occur in a reduced state (Crittenden et al., 2012). Oxidation can be applied at different places in the treatment chain, whereas it is most commonly applied as preoxidation in the earlier part of the chain or as disinfection in the later part. The principal use of oxidation is for taste and odour control, removal of hydrogen, colour, iron and manganese, or as disinfection. Conventional oxidizers for the mentioned targets are oxygen, chlorine, chlorine dioxide, hydrogen peroxide, ozone and permanganate, which are listed in Table 2.4 along with a comment on their removal efficiency of Fe(II), Mn(II) and sulphide. As can be seen, the removal efficiency and oxidation rates vary for the oxidants and conditions, such as pH and occurrence of NOM.

Table 2.4: Conventional oxidants and their removal rate of iron, manganese and sulphide. Information obtained from Crittenden et al. (2012)

Oxidant	**Fe(II)**	**Mn(II)**	**Sulphide**	**Note**
Oxygen	Rapid*	Slow	Slow	*at alkaline pH
Free chlorine	Effective	Effective*	Effective*	*at pH>8
Chlorine dioxide	Likely effective*	Rapid	Likely rapid	*disturbed when complexed with NOM
Hydrogen peroxide	Effective	Effective		
Ozone	Effective	Very rapid		
Permanganate	Effective*	Effective*		*increase of pH accelerates the oxidation kinetics

The oxidants share some similar operational challenges when used in drinking water treatment. The oxidation of Fe(II) with oxygen and chlorine are disturbed by the occurrence of NOM (Crittenden et al., 2012). For free chlorine and ozonation, Mn(II) requires posterior filtering and even in the use of filtering, oxidation of Mn(II) through ozonation occurs so rapidly that it tend to cause clogging or form extremely fine colloids, risking disturbing the following treatment steps. When adding ozone and permanganate, alkalinity is consumed subsequently as Fe(II) and Mn(II) are oxidized, which is an issue especially when alum coagulation is involved, thus it also consumes alkalinity. The dose range of permanganate is critical, and overdosing can cause deposits in the distribution system.

Crittenden et al. (2012) further explain that when NOM occur in the water, the reaction with ozone form compounds of lower molecular weight. If the compounds are deficient removed after the oxidation process, ozonation can cause biofouling in later treatment steps. An operational advantage with ozonation is that the process doesn't generate sludge (Razali et al., 2023). On the other hand, ozonation constitutes a risk since it can cause degradation of biopolymers, which was further studied by Ouali et al. (2021). The conclusion of the study was that the properties and performance of polyamide membranes were highly damaged by contact with ozone, since ozone caused partial removal of the active layer of the membrane.

The application of ozonation prior to membrane filtration was investigated by Yu et al. (2018) for a surface water of moderate quality. One UF, loose NF and dense NF were tested, and the results showed that pre-ozonation slightly decreased the flux decline for the dense NF membrane. In contrast, applying pre-ozonation prior to the loose NF membrane resulted in a slight increase of flux decline. The differences of the NF performance was thought to relate to the new sizes of organic compounds generated through ozonation.

In a study performed on surface water with notable concentrations of iron and manganese, ozone oxidation with filtration reduced the level of iron and manganese to beyond detection levels (Mijatović et al., 2004).

2.4.10 Alkaline filtration

Alkaline filters consist of a container filled with filter mass. This treatment is usually inserted as posttreatment to increase alkalinity, pH and hardness (Svenskt Vatten, 2011). The chemical process can be described by the excess of hydrogen ions in relation to the hydrogen carbonate ions. When carbonate ions are added through the filter mass, the excess of hydrogen ions is consumed, which results in an increased pH value.

Historically, alkaline filters have mostly been used in small scale groundwater treatment plants (Svenskt Vatten, 2011). Two types of filter masses, including calcium carbonate and dolomite, are available. The evolving material, calcium carbonate is classified as a slow mass, whilst dolomite is classified as fast due to the present magnesium which make the mass more easy-soluble. Dolomite is usually chosen, however it is hard to reach an adequate pH with this material. When dolomite is added, the pH rises up to a pH value of 10.5. After some weeks of decreasing pH, the pH is considered as too low, which results in a new addition and the pH rises again. The size of the material varies between 0.5-4 mm, and the bed height is in the range of 1-3 m. For calcium carbonate, a contact time of 20–30 minutes is preferred. In order to maintain the filtration capacity, regular back washes are required.

Except the reaction with soluble CO2 present in the water, it can contribute to the oxidation of iron and manganese, and the filtering of particles (Svenskt Vatten, 2011). The alkaline environment results in the creation of solid basic salts of the ions, rust and brownstone. The manganese precipitate is catalysed by the brownstone, resulting in an enhanced reduction of manganese. In order to precipitate iron, the water preferably is aerated. Despite this, waters with too high concentrations of iron and manganese should not be treated with alkaline filters since this will result in fouling of the filter. The created iron- and manganese hydroxides are hard to flush from the filter. If these compounds clog the filter, the abilities of increasing alkalinity, pH and hardness are reduced. As a guideline, the ingoing water should not have an iron concentration above 0.5 mg/l, whilst the manganese concentration should not exceed 0.05 mg/l.

In a pilot study (unpublished) performed on surface water, the removal efficiency of manganese was investigated. After running the study for two months, a significant reduction of manganese was observed. The manganese concentrations in the raw water was approximately 0.25 mg/l, after the alkaline filter the concentration was below the limit of 0.05 mg/l.

3

Methodology

3.1 Study of literature

The study of literature was performed in three steps, resulting in four theory sections and a matrix accounting for the performance of the potential pretreatment methods identified. The aim of the steps differed from each other and was performed in different ways.

To begin with, the general aspects of drinking water were investigated. The aim was to characterize the raw water commonly used for drinking water production, and account for the components required to be treated and water quality parameters to monitor during the operation. Further, regulations and guidelines relevant for drinking water production in Sweden were presented. The literature used to investigate the topics mentioned was mainly documents found from authorities, such as World Health Organization, United Nations, Swedish NFA and Svenskt Vatten. In addition, scientific literature from the database ScienceDirect and the search engine Google Scholar, as well as books on the topic drinking water treatment, was used to establish a brief, scientific overview of the drinking water topic. Similarly, by using scientific literature from ScienceDirect, Google Scholar and books, a general theory section on membrane filtration was produced. The aim with this section was to provide information of the purpose, function and obstacles with membrane filtration since NF-filtration was chosen to act as the main treatment step in the MDWTS.

The second step of the study of literature aimed to investigate the raw water components and factors influencing fouling in nanofiltration, aiming to work as a basis for investigating relevant pretreatment methods. This procedure was more iterative and precise in the use of keywords. The sources used to gain knowledge of fouling in nanofiltration were obtained from ScienceDirect, Google Scholar and books. In addition, a main approach during this step was citation chaining, i.e. further exploring the sources referred to in literature on the subject.

The third and final step of the study of literature aimed to form the basis for the suggestion of the pretreatment method. Initially, possible pretreatment methods were identified based on their ability to remove key foulants and if they had been implemented prior to NF. At this point, treatment methods with extensive footprint, such as sedimentation and slow sand filtration, were excluded due to their unsuitableness of implementation in a MDWTS. The remaining identified pretreatment methods were approached with a systematic literature search, aiming to present comparable information regarding removal efficiency of key foulants, fouling mitigation, footprint and operability. The information was obtained from scientific reports, reviews on the topic and compilations from organisations.

A brief overview of the keywords used to obtain the aims of the study of literature is shown in Table 3.1. The words within parentheses represent all pretreatment methods respectively key foulants identified.

Table 3.1: Aim and keywords used in the study of literature

Aim	Key words	
Providing general information on drinking water production and membrane filtration	"Drinking water production" "Membrane filtration"	
Gaining knowledge of factors and components affecting fouling in NF	Nanofiltration AND	Organic fouling Inorganic fouling Biofouling
Present information of pretreatment methods' performance	Nanofiltration AND [Pretreatment method]	"Pretreatment method" "Nanofiltration fouling" "Removal of [key foulant]"

3.2 Setting a frame for the pretreatment method

Together with the consultant company Norconsult AB and the membrane facility manufacturer Björks Rostfria AB, the prerequisites for pretreatment were established. First, the properties of the NF used were investigated. This was followed by applying the findings from the study of literature, through identifying the key foulants in three water types. Further, possible pretreatment chains possessing the ability to manage the identified key foulants in respective water type were presented. In addition, a flowchart was created to connect key foulants with suitable pretreatment methods. Lastly, designs accounting for the dimensions of the pretreatment chains applied in a MDWTS were visualised.

3.2.1 Properties of NF

Within this project, the spiral wound membranes NF270 and NF90 from the membrane manufacturer Dupont, were used as the main treatment step in the MDWTS. The specific NF modules used within this work operate for an influent flux of 3.5 m^3/h with 75% recovery rate, resulting in a permeate flux of around 2.6 m^3/h. NF270 is customized to treat raw water with high organic content, whilst NF90 is preferable when treating raw water with high inorganic content. The MWCO varies from 200-400 Da for both NF90 and NF270. Table 3.2 presents additional properties of the membranes, as described in the product data sheets for NF90 respectively NF270 (Dupont, 2021a; Dupont, 2021b).

Table 3.2: Properties of NF90 and NF270 (Dupont, 2021a; Dupont, 2021b)

Properties	**NF90**	**NF270**
Material	Polyamide	Polyamide
Contact angle	27	54
	Hydrophilic	Hydrophobic
Operational pH-range	3-10	2-11
pH-range during short-term cleaning	1-12	1-13
Free Chlorine Tolerance	<0.1 ppm	<0.1 ppm
Used for	Raw water with high inorganic content	Raw water with high organic content

Dupont has established guidelines for the feed water entering their NF and RO membranes, which are presented in Table 3.3. The estimated values in Table 3.3 were based on the study of literature and discussion with experts within the field of drinking water processes.

Table 3.3: Guidelines for RO feed water quality (Dupont, 2023)

Component	**Max level**	**Source**
TOC	3 mg/l	Dupont, 2023
COD	10 mg/l	
Fe(II)	4 mg/l*	
Fe(III)	0.05 mg/l	
Mn	0.05 mg/l	
Al	0.05 mg/l	
Turbidity	1 FNU	Estimated values
Calcium	15 mg/l	

** at pH <6 and oxygen <0.5ppm*

3.2.2 Application on water types

Quality characteristics of water from three different sources in Sweden were selected to represent potential influent water, possessing various challenges during drinking water treatment. The water sources that were chosen possess the main characteristics of groundwater with high hardness (water type 1), surface water currently extracted and treated for large-scale drinking water production in a centralised DWTP (water type 2) and surface water with high levels of humous, turbidity and inorganic content (water type 3). Key foulants were identified for the water types, based on information gained during the study of literature and through discussions with a membrane manufacturer. In order to determine whether a water type needed pretreatment for a key foulant or not, the guidelines for RO feed water quality presented in Table 3.3 were used.

The water quality parameters shown in Table 3.4 represents yearly median values obtained from a groundwater source, extracted from five wells. Values marked in red are exceeding the guidelines from Dupont, whilst the values marked in orange are exceeding the estimated limit values for the other key foulants. The water is classified as a hard groundwater, as it contains high levels of calcium and moderate levels of magnesium. Even at its maximum value, the colour in the raw water is low enough to pass the requirements for outgoing drinking water without treatment. Overall, the groundwater possesses a low turbidity which implies that NOM, iron and manganese are dissolved in larger extent.

The identified key foulants for water type 1 is furthermost the hardness ions. In addition, the levels of turbidity and iron content obtained in Well 4 constitute a fouling risk if not managed. For the posttreatment, it is further of importance to consider the pH and alkalinity, which may be strongly affected by the NF.

Table 3.4: Water type 1: Water quality parameters for a groundwater with high hardness

Parameter	Unit	Well 1	Well 2	Well 3	Well 4	Well 5
Temperature	°C	8.8	8.6	**8.9**	8.6	*8.4*
Turbidity	FNU	*<0.10*	1.0	0.17	**1.5**	0.37
Colour	mg/l Pt	*10*	10	**15**	15	10
pH	-	*7.4*	7.4	7.4	7.4	**7.5**
Conductivity	mS/m	54.7	54	*53.4*	**55**	54.6
Alkalinity	mg/l HCO_3^-	247	244	246	**257**	*241*
COD	mg/l O2	4.0	*3.4*	**4.3**	3.8	4.0
Aluminium	mg/l	*<0.01*	**0.01**	0.01	0.01	<0.01
Iron	mg/l	*<0.01*	0.033	0.016	**0.155**	0.04
Manganese	mg/l	*0.024*	**0.136**	0.068	0.082	0.049
Calcium	mg/l	90.2	89.1	*87.8*	**91**	88.8
Magnesium	mg/l	8.5	*8.2*	8.3	**9.1**	8.9
Hardness	dH	14.6	14.4	*14.2*	**14.8**	14.5
Silica	mg/l	-	-	-	-	-

Water type 2 represents a surface water currently extracted and treated for large-scale drinking water production in a centralised DWTP. The obtained minimum, median and maximum values for the water quality parameters are shown in Table 3.5. The minimum and median levels of turbidity show that particles normally occur in the water to some extent, requiring pretreatment. Occasionally, the turbidity reaches remarkably high values, which requires additional consideration when determining pretreatment. The colour further indicates that the iron and NOM also exist in their soluble state.

The key foulants observed for water type 2 are particles and colloids enhancing turbidity, NOM and iron, occurring in particulate and dissolved form. This water type shows on relatively high content of aluminium, which can increase the risk of NF fouling. However, considering the pH of the raw water, the aluminium is assumed to be complexly bound to NOM and the treatment implemented to target NOM is therefore assumed to simultaneously reduce the aluminium content. An additional aspect to carefully consider is the low alkalinity observed in the raw water, which can affect the choice of pretreatment. The alkalinity is of particular importance related to the use of chemical flocculation.

Table 3.5: Water type 2: Water quality parameters for a surface water

Parameter	**Unit**	**Min**	**Median**	**Max**
Turbidity	FNU	2.9	5.2	21
Colour	mg/l Pt	12	16	39
pH	-	7.1	7.4	7.8
Conductivity	mS/m	0.2	0.5	11.6
Alkalinity	mg/l HCO_3^-	18.9	20.1	21.4
COD	mg/l O2	4	4	8
TOC	mg/l	4.2	4.5	6.0
Aluminium	mg/l	0.11	0.24	0.95
Iron	mg/l	0.1	0.2	0.75
Manganese	mg/l	0.003	0.007	0.019
Magnesium	mg/l	1.5	1.6	2.2
Calcium	mg/l	1.2	1.3	1.8
Silica	mg/l	-	-	-

Lastly, water type 3 represents a surface water, with challenging levels of turbidity, NOM, aluminium, calcium, iron and manganese. Without pretreatment, the levels constitute a severe risk to cause NF fouling for both the ordinarily and maximum values, and even for some minimal values of the parameters. The high levels of turbidity and colour indicate on the occurrence of both particulate and dissolved forms of NOM, iron and manganese. The content of NOM is consistently a concern. In addition, the water's content of calcium generates water of moderate hardness. While the levels of calcium are not an issue for the outgoing drinking water, calcium risks to cause fouling when concentrated at the NF membrane surface. The alkalinity is presumed to be stable enough to not alter the choice of pretreatment.

For water type 3, NOM is consistently identified as a key foulant of particular concern. Iron, manganese and calcium are identified as additional key foulants. In similarity with water type 2, the pH value of this surface water implies that aluminium is complexly bound to NOM. However, if the pH slightly increases, aluminium can dissolve and constitute a problem if applied to NF.

Table 3.6: Water type 3: Water quality parameters for surface water with high levels of humous and turbidity

Parameter	**Unit**	**Min**	**Median**	**Max**
Turbidity	FNU	3.6	16	92
Colour	mg/l Pt	50	80	130
pH	-	7.5	7.8	8
Conductivity	mS/m	27.4	29.5	32.8
Alkalinity	mg/l HCO_3^-	97	100	120
COD	mg/l O2	8.4	21	40
Aluminium	mg/l	0.34	0.56	2
Iron	mg/l	0.32	2.1	12
Manganese	mg/l	0.05	0.22	1.1
Calcium	mg/l	46	50	54
Magnesium	mg/l	3.5	4.3	4.7
Silica	mg/l	3.7	3.7	3.7

Based on the study of literature, pretreatment methods that target the key foulants occurring in the three water types were evaluated based on their removal efficiency and operability. Further, possible pretreatment chains were discussed with experts in drinking water treatment, aiming to create a favourable option for each water type. Further, a flowchart was created aiming to include additional compositions of raw water. The flowchart was designed to evaluate when pretreatment is required and to connect key foulants to possible pretreatment methods.

3.2.3 Design of pretreatment method for the water types

When the pretreatment chains were suggested, calculations on the approximate size were performed. Further, the processes were briefly visualised to get an overview of the proportions of the pretreatment methods in a MDTWS of 40 ft. A 40 ft container (inner dimensions: L=12032mm, B=2438mm) was used for all the three processes. Depending on the height of the pretreatment processes in the chains, containers with different heights were used. The available options were an ordinary container (inner height: H=2390mm) and a high cube container (inner height: H=2695mm). In order to maintain a NF influent flux of 3.5 m^3/h, the design flow for the pretreatment methods was set to 4 m^3/h.

In all treatment chains, a grid and a cartridge filter were included. The purpose of the grid is to filter larger debris and branches to protect the later treatment steps. Before the NF, a cartridge filter was implemented as a safety measure if operational problems earlier in the process would occur. For the design flow of 4 m^3/h three cartridge filter modules were inserted before the NF as a safety measure. The cartridge filter module (dimensions: L=762mm and D=64mm) resulted in a total area of $762 \cdot 192 mm^2$ for three modules. The pore size of the cartridge filter was set to 5 μm based on recommendations from the membrane manufacturer. If colloidal silica pose a risk of fouling, a cartridge filter with a pore size of 1-3 μm was used instead (Dupont, 2023).

The footprint for NF (volume of a module: L=2850mm, B=860mm, H=1860mm) includes space for storage and dosage of antiscalans and chemicals. For posttreatment, UV disinfection and adjustment of pH and alkalinity were used. UV disinfection, required one meter of the permeate pipe. At the end, a 200 litre container was inserted to cover the need for chemicals to adjust the pH of the outgoing drinking water.

The footprint of the pretreatment processes including rapid sand filtration, BAC filtration and bioreactors was calculated by dividing the design flow with the surface loading rate for each method presented in Table 3.7. Coagulants for chemical flocculation and aeration were induced to the pipeline. When chemical flocculation was suggested, the surface loading rate in posterior filter process was set to 5 m/h, since the process requires surface loading rates <5 m/h. For chemical flocculation over UF, the retention time was considered.

There are several UF modules available on the market, which can be designed inside-out or outside-in. For the outside-in modules IP/D-77 and IP/D-51 were considered as possible alternatives for pretreatment. In general, the filtrate flux interval for Dupont´s UF modules is 40-110 lmh (Dupont, 2022). According to experts within the field, a flux of 60 lmh is optimal. When chemical flocculation over UF was suggested, an inside-out module, MB 80 TR, was used. A footprint of 1 * 1,5 m, which includes space for the whole installation, was approximated for the UF in all suggested pretreatment chains.

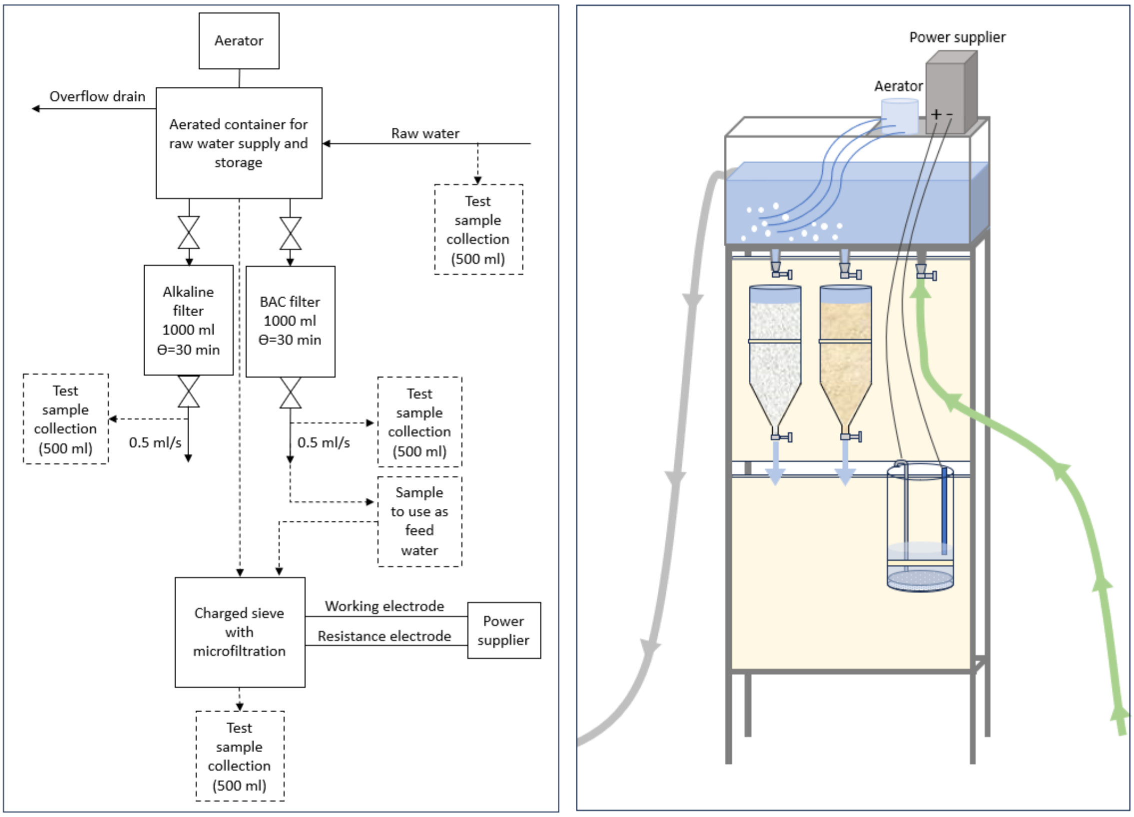
Table 3.7: Surface loading rate for pretreatment methods

Pretreatment	**Surface loading rate (m/h)**
Rapid sand filtration	5-10
BAC filtration	5
Biological reactor	12-15
Chemical flocculation	<5

3.3 Bench study of pretreatment methods

As a complement to the result from the study of literature, three potential pretreatment methods were tested in a bench study. The investigated methods include a biological activated carbon filter, an alkaline filter and a charged sieve followed by a disc filter. As a complement, biological filtration in combination with the charged sieve was tested to evaluate the synergy effects. The purpose of the tests was to evaluate the methods' removal efficiency of the key foulants identified in the literature study, including NOM, turbidity, aluminium, iron, manganese, calcium and magnesium.

A flowchart presenting the components used in the bench study is shown in Figure 3.1, along with an illustration of the physical arrangement of the experiments.



(a) (b)

Figure 3.1: Flowchart (a) and an illustration of the physical arrangement (b) of the bench study. The continuous lines in the flowchart represent continuous flows, whereas the dashed lines represent temporary flows used for the charged sieve and the collection of test samples.

The bench study were conducted at a DWTP, and raw water was taken from the raw water intake pipe. The raw water was led to a container and aerated with an aerator (Superfish Airflow 4) placed on top of the rack. The BAC and alkaline filter was set up and connected to the outlets of the container, aiming to operate continuously, while the charged sieve was set up to operate during three occasions under supervision. In addition, samples of raw water were taken and sent for chemical analysis.

Table 3.8 accounts for at which occasions the different pretreatment methods were started and sampled. The collected samples were sent for chemical analysis during the same day as they were collected. The chemical parameters which were analysed for raw water and posterior the experiments are presented in Table 3.9.

Table 3.8: Overview of the dates when the pretreatment methods were started and/or sampled

Pretreatment method	**30/4**	**7/5**	**21/5**	**28/5**
Raw water	Sampled	Sampled	Sampled	Sampled
BAC filter	Started	Sampled	Sampled	Sampled
Charged sieve				
0 V		Sampled	Sampled	Sampled
+0.5 V		Sampled	Sampled	
-0.5 V		Sampled	Sampled	
+1.0 V				Sampled
Alkaline filter		Started	Sampled	Sampled
Combination			Sampled	

Table 3.9: Chemical parameters analysed for the water samples

Parameter	**Unit**
Turbidity	FNU
Colour	mg/l Pt
pH	-
Conductivity	mS/m
Alkalinity	mg/l HCO_3^-
COD	mg/l O2
Aluminium	mg/l
Iron	mg/l
Manganese	mg/l
Calcium	mg/l
Magnesium	mg/l
Silica	mg/l

3.3.1 Experiment 1 - Biological activated carbon filtration

In this test, a cylindrical glass funnel was fixed on a stand and filled with 1000 ml of activated carbon. The carbon material was taken from a drinking water treatment process, and therefore the material was already biologically activated. By doing this, the startup time to make the carbon biologically active was excluded. A valve was installed to be able to adjust the flow. To ensure that the funnel always was filled with water, the inflow was greater than the outflow of the funnel. The outflow was set to 0.5 ml/s, which resulted in an EBCT of about 30 min.

The setup was connected to the intake pipe and the test was running continuously for 28 days. Water samples were collected and sent for chemical analysis after 7, 21 and 28 days. To ensure that the raw water was not collected for the water samples, a short hose was connected to the outlet of the glass funnel. For this experiment the levels of manganese, iron and NOM were expected to decrease.

3.3.2 Experiment 2 - Charged sieve with a following disc filter

For the charged sieve experiment, a small disc filter construction made of plastic was fixed on the stand, see Figure. The pore size of the used disc filter was 10 μm. A grid made of stainless steel corresponded to the sieve, and was connected to the disc filter construction. Three filter papers (Munktell filter paper 15 cm) were placed beneath the disc filter, aiming to increase the resistance to maintain a relatively stable water level and extend the retention time.

To charge the grid, a power supply apparatus (Power Supply B 300 D manufactured by Oltronix AB) was used. The grid was connected to the cathode, creating the working electrode. A platinum electrode (M24 1 Pt from Radiometer analytical) was connected to the anode, creating the counter electrode. During the first sampling occasion, the sieve was charged with a voltage of 0 V, -0.5 V and +0.5 V. This was repeated during the second sampling occasion. In addition, the sieve was charged with +1.0 V during the last sampling occasion. In this test, the negatively charged particles were expected to be collected in the charged sieve when positive voltage was applied. As a result, the concentration of NOM and turbidity were expected to decrease. In addition, a negative charge was applied to evaluate the potential of affecting the positively charged components in the water.

For the combination of the pretreatment methods, the BAC filtration effluent was collected and fed through the charged sieve which was charged with +0.5 V. One water sample was collected and sent for analysis.

3.3.3 Experiment 3 - Alkaline filter

To create an alkaline environment, the glass funnel was filled with approximately 1000 ml of crushed marble, with a pore size of 7-15mm, which is defined as a slow mass. A connection from the raw water container to the glass funnel was installed, see Figure 3.2. Raw water was continuously led to the funnel, with a greater inflow than outflow, in order to maintain a stable water level in the funnel and keep the material saturated. Samples were collected at two occasions, after operating in 7 respectively 14 days, and sent for chemical analysis.

(a)

(b)

Figure 3.2: Pictures taken during the operation of the bench study, showing the set-up of all three tests (a) and a close up on the charged sieve (b)

4

Results and discussion

4.1 Study of literature

The results from the study of literature are divided into two sections. Firstly, the removal efficiency of the pretreatment methods on the identified key foulants from the study of literature are presented in individual charts. The identified key foulants are turbidity, NOM, iron, manganese, aluminium, calcium, magnesium and silica. For the second part, the pretreatment methods' fouling mitigation, obtained in the study of literature, are presented and discussed. A comprehensive compilation is presented in Appendix A.

4.1.1 Removal efficiency of turbidity

Several of the pretreatment methods show a moderate to effective removal efficiency of turbidity. As can be seen in Figure 4.1, the highest removal is achieved by MF, UF, RSF, alum coagulation and the combined pretreatment chains.

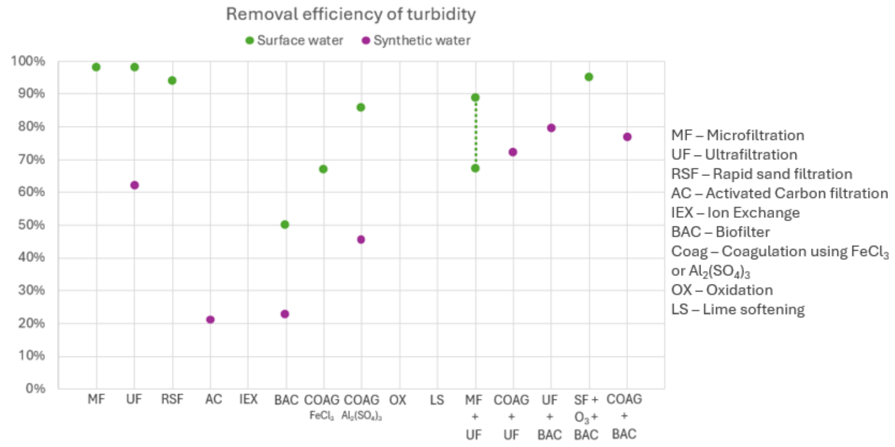


Figure 4.1: Removal efficiency of turbidity by pretreatment methods investigated in the study of literature. The dotted line represents studies where the removal efficiency was described as an interval.

The removal efficiency of turbidity through filtration methods is highly dependent on the sizes of the components in the raw water. Therefore, it is believed that these methods possess a high removal potential, while the range of removal varies depending on the components in the raw water. AC and BAC show a fair to moderate improvement of the turbidity, which is reasonable for methods with adsorption as primary removal mechanism. In the study of literature, it was not clarified whereof ion exchange, oxidation and lime softening target turbidity on their own. In general, the removal of turbidity in natural surface water is higher than in synthetic water.

4.1.2 Removal efficiency of NOM

The pretreatment methods' ability to remove NOM based on the study of literature is presented in Figure 4.2. There is a wide fluctuation of removal efficiency between and within the pretreatment methods. In addition, the results from the studies are obtained for various water types. In general, UF, AC, ion exchange (anion), coagulation, lime softening and the combined pretreatment methods show a good potential in removing NOM. Further, RSF, ion exchange (cation), BAC and the combination of SF, ozone and UF, show a fair removal efficiency of NOM. No removal efficiency was found when MF or oxidation are applied solely, but the methods' potential to remove NOM (particulate for MF) was mentioned in studies.

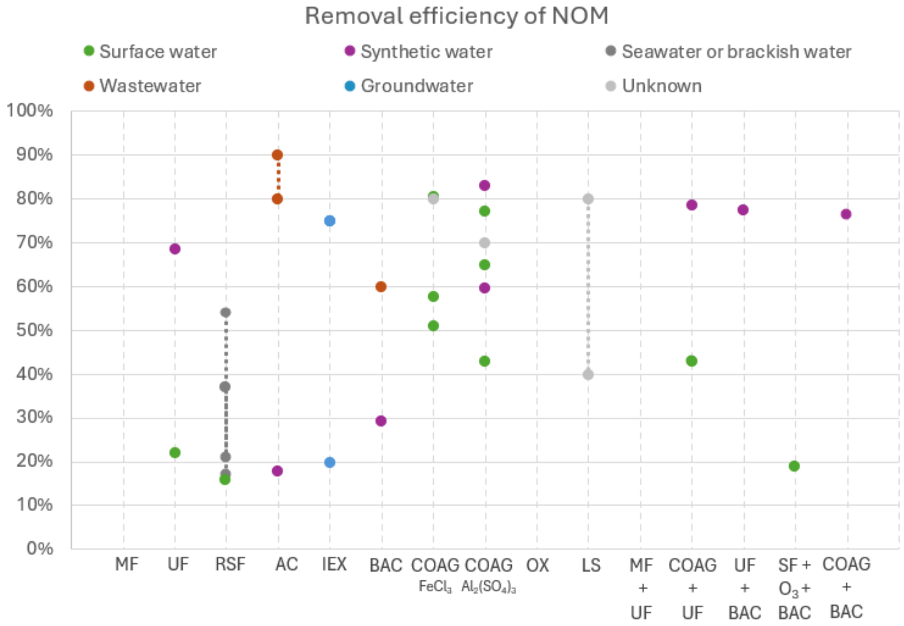


Figure 4.2: Removal efficiency of natural organic matter by pretreatment methods investigated in the study of literature

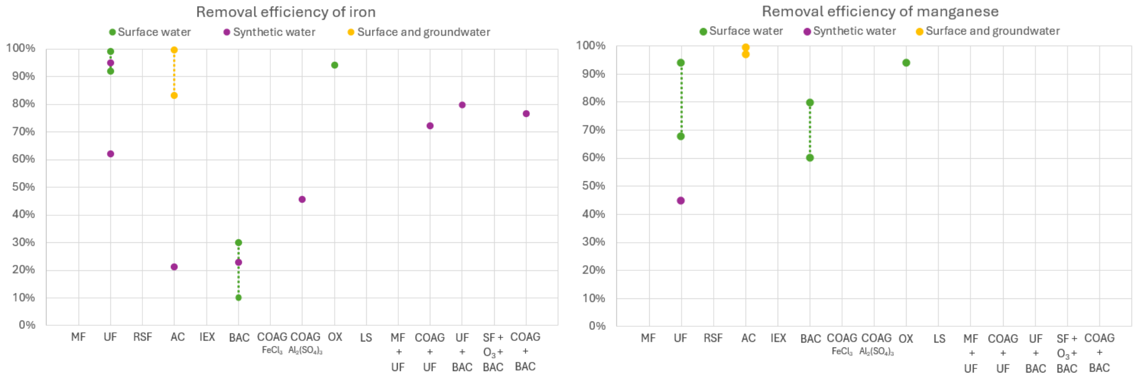
Since NOM consists of different types of substances which have varied properties, it is important to characterize the present NOM in raw water in order to implement a suitable pretreatment method. Liquid Chromatography – Organic Carbon Detection (LC-OCD) is a possible analytical method used to detect different types of NOM in the water. In addition, UV-absorbance can give an indication of HMW NOM concentration. For instance, coagulation and flocculation is more effective against HMW NOM, whilst BAC affects the easy degradable and LMW NOM. The characterization also helps to predict possible chemical consequences, such as creation of by-products. Another factor which needs to be considered is whether the NOM is particulate or dissolved. If the NOM is particulate, it is possible to remove it directly via size exclusion. However, if the NOM is dissolved it needs to be flocculated or removed with other removal mechanisms such as adsorption and cake formation occurring in a NF membrane.

Multiple studies presented in Figure 4.2 have used other water types than surface and groundwater in their studies, for instance the removal effect on wastewater is tested in two studies. In addition, a big part of the studies have used synthetic water for their tests. The use of synthetic water could be problematic, since inadequate results are a possible outcome (Sillanpää et al., 2018). Some of the results presented in Figure 4.2 are questionable. For AC, there is a large interval between approximately 20-90%. However, the high value of 90% was obtained for a wastewater where the water composition differ. The same applies for BAC, where the interval is between 30-60%. Once again, the higher value was determined for a wastewater, and BAC enhances the reduction of easy-degradable NOM. Another large interval of 20-70% is observed for UF. The value of a 70% DOC reduction is questionable, and a possible explanation to this inadequate result is the use of synthetic water. The large interval of 40-80% for reduction of NOM for Lime softening is remarkable. Since the values were obtained from a literature and no further information was declared, it is hard to comment whether the result is valid or not.

The variations of the obtained results for iron- and alum-based coagulants are reasonable, since the conditions such as pH and dosage highly affect the outcome. For ion exchange, the changes in the result can be described by the different resin used in the tests. For the treatment combinations, higher reductions of NOM is expected. Despite this, the result of almost 80% DOC reduction for the combination of BAC and UF is questionable without the addition of a coagulant. However, synthetic water was used for the tests, which could explain the high outcome.

4.1.3 Removal efficiency of iron and manganese

The study of literature accounts that not as many of the investigated pretreatment methods possess the ability to remove iron and manganese, as for turbidity and NOM. The methods that possess the highest removal efficiency of iron and manganese, as shown in Figure 4.3, are UF, AC and oxidation. For iron, the pretreatment chains involving coagulation and posterior filtration as well as the combination of UF and BAC also possess a high removal efficiency, while BAC and coagulation with aluminium possess a fair removal efficiency. Further, BAC shows a higher removal potential of manganese than iron.



(a) (b)

Figure 4.3: Removal efficiency of iron (a) and manganese (b) by pretreatment methods investigated in the study of literature

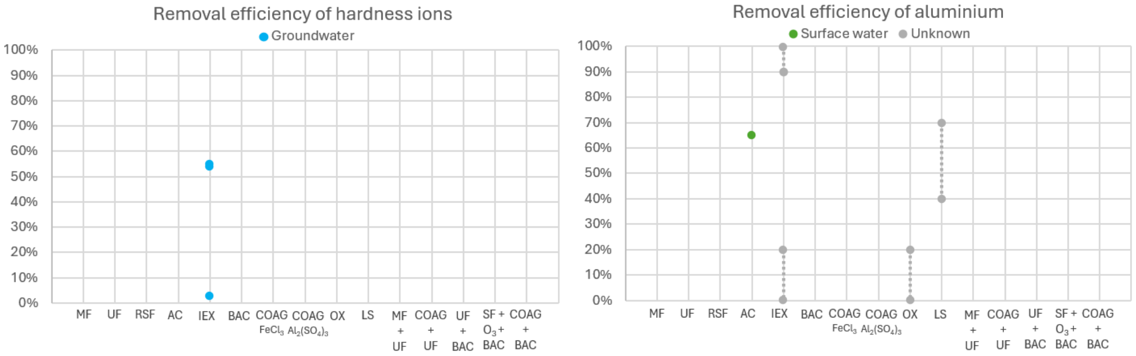
The state of the raw water, such as pH value and redox potential, is important to consider for the removal of iron and manganese. For example, the high removal efficiency received for UF and AC, is believed to account for when iron occur in their oxidised form. For manganese, the results for UF and AC are questionable. AC may possess the ability to remove manganese if biological growth in the carbon is developed. Still, a removal of 90% is unreasonable. A possible explanation to this could be a low initial concentration of manganese. The same applies for the results of manganese removal with UF.

The high removal efficiency obtained from oxidation represent results from a process where ozonation with posterior PAC filtration is operated. The obtained values are considered as accurate, but when suggesting pretreatment methods to operate prior to NF, it is important to consider the risks involved with the different oxidants. Even though ozonation and chlorination possess the ability to target iron and manganese efficiently, these oxidants constitute a risk of destroying the active membrane surface. Therefore, when considering oxidation within this work, aeration is the primary oxidant. Further, a lower removal efficiency than the one shown for oxidation in Figure 4.3 is expected.

4.1.4 Removal efficiency of hardness ions, aluminium and silica

In the scope of this study of literature, the pretreatment method showing potential to remove the hardness ions calcium and magnesium are ion exchange, which Figure 4.4 accounts for. Ion exchange using cationic resin and a combination of cationic and anionic resin show a moderate removal efficiency, while anionic ion exchange generates a minor effect. Further, the ability to remove hardness through lime softening is commonly mentioned in literature. However, the removal efficiency of key foulants by implementing lime softening requires further investigation. Regarding the removal of hardness, one additional method to consider is the NF itself, which possess the ability to reduce the concentration of hardness. In addition, the use of antiscalants are not classified as a pretreatment method within this work, but heavily considered as a method to mitigate inorganic fouling.

Figure 4.4 also presents the pretreatment methods' removal efficiency of aluminium. AC, ion exchange (cation) and lime softening show a moderate removal efficiency, while ion exchange (anion) and oxidation show a fair removal efficiency. The presented values are obtained from a literature review and a preliminary analysis at a water treatment plant, with no further specification of the raw water characteristics or if the methods are used on their own. Therefore, the comparison of the methods becomes more difficult, as well as applying the findings.



(a) (b)

Figure 4.4: Removal efficiency of the hardness ions (a) and aluminium (b) by pretreatment methods investigated in the study of literature

Regarding the removal efficiency of silica, no information was found for the pretreatment methods in the study of literature. However, it is declared that colloidal silica can be removed by the use of coagulants or by filters with a pore size of 1-3 μm. Overall, information regarding the removal efficiency of key foulants was hard to find for the pretreatment methods investigated. The methodology applied was performed through a study of literature, and few studies where the pretreatment methods were investigated on their own were found. The pretreatment methods account for few case studies, making it hard to present representative removal performances. An explanation to this can be that some of the key foulants are rare in high concentrations, and therefore they are not deeply investigated. In addition, the results represent different water sources, complicating the comparison of the methods. To generate more accurate results for the comparison, conducting pilot studies on the pretreatment methods would be valuable. However, since pilot studies are time-consuming both regarding planning and operation, the methodology used within this work gives an overview of which key foulants the investigated pretreatment methods target.

4.1.5 NF fouling mitigation

In Table 4.1, pretreatment methods that have shown NF fouling mitigation are presented. As can be seen, the practice of measuring fouling mitigation varies widely between the studies. The fouling mitigation accounts for lab- and pilot studies on NF resistance and NF permeate flux, as well as calculations and estimations. While the aspect that NF fouling is measured through different procedures restrict the comparison, some conclusions can be drawn from the obtained result.

No adequate NF fouling mitigation was observed when using sand filtration, whilst MF, AC and BAC filtration showed on moderate fouling mitigation. When UF filtration and UF-combined pretreatment was implemented prior to NF, efficient fouling mitigation was observed. Efficient fouling mitigation also applies for pretreatment methods that include the addition of a coagulant. For ion exchange, the fouling potential is presented which was obtained through calculations, with varying results depending on resin used. Ozonation is the only one of the investigated pretreatment methods that shows a negative impact on NF fouling, which was observed when a loose NF was used. For the dense NF, ozonation showed a slight positive effect on NF fouling mitigation. No information was found regarding the effect on NF fouling when lime softening or aeration was implemented as pretreatment.

Pretreatment method	**Specification**	**Observed NF fouling mitigation**
MF	-	Total resistance decreased with 17.4-27.5%
UF	- Pore size 0.02 μm	Total resistance decreased with 31.5-45.7% Increase of NF permeate flux with 49 %-points
Sand filtration	-	No adequate fouling mitigation
AC	- GAC	Assumed mitigation of organic fouling Increase of NF permeate flux with 2.8 %-points
BAC	-	Increase of NF permeate flux with 4.8 %-points
Ion exchange	Combined ion exchange Anion exchange Cation exchange	Calculated NF fouling potential Org=0.3 Inorg=0.1 Calculated NF fouling potential Org=0.25 Inorg=0.2 Calculated NF fouling potential Org=0.8 Inorg=0.4
Chemical flocculation	$Al_2(SO_4)_3$, jar test	Increase of NF permeate flux with 21.7 %-points
Oxidation	Ozonation - Dense NF Ozonation - Loose NF	5% decrease of NF flux decline Slight increase of NF flux decline
Coagulation + UF	$Al_2(SO_4)_3$ Pore size 0.02 μm	Positive effects on NF fouling Increase of NF permeate flux with 49 %-points
UF + BAC	Pore size 0.02 μm	Increase of NF permeate flux with 49 %-points
Coagulation + BAC	$Al_2(SO_4)_3$	Increase of NF permeate flux with 52 %-points
SF + Ozone + BAC	-	Fouling mitigation is estimated but not proved

Table 4.1: Fouling mitigation in NF observed for the pretreatment methods

When conducting the study of literature and investigating NF membranes, fouling and the importance of pretreatment was frequently mentioned. However, not many studies were found where the efficiency of the pretreatment to mitigate fouling was investigated. The studies which had observed a fouling mitigation used different measurements to present the results. Due to this, it was hard to compare the different studies which had noticed a fouling mitigation. Overall, few studies which evaluated a pretreatment method prior to NF were found. As a consequence of the mentioned difficulties, the results presented for NF fouling mitigation are considered as too uncertain to be used as a basis for the suggestion of the pretreatment method chain. Instead, pilot studies are proposed to gain knowledge on the pretreatment methods effect on NF fouling mitigation.

4.2 Pretreatment for the water types

In this section, the pretreatment chains conducted for each water type are presented. The suggestion is a result of considering the ability of a pretreatment method, investigated in the study of literature, to target the key foulants occurring in the specific water type. As a complement, a possible design of each process including the footprint of the treatment steps is included.

4.2.1 Suggested pretreatment for water type 1

The groundwater investigated obtains pH values varying from 7.4 to 7.5 and an alkalinity above 230 mg/l HCO_3^- . The primary key foulants in this water are calcium and magnesium. The levels of manganese observed in most of the wells can constitute a fouling risk, according to the guidelines for NF feed water quality. Further, one of the wells contains levels of turbidity and iron that may contribute to the risk of NF fouling. The key characteristics of water type 1 are listed in Table 4.2 and Figure 4.5 shows the suggested pretreatment chain for water type 1.

To manage the levels of turbidity shown in one well, implementation of a low pressure membrane is suggested. The type of membrane suitable depends on the particle sizes that enhance turbidity, where MF is suitable if the particles are bigger than approximately 0.1 μm and UF is suggested if the particles are smaller. In this solution, UF is implemented to target the turbidity. The iron levels are targeted by aerating the water prior to the filtration. Addition of antiscalants prior to NF is proposed to mitigate the scaling risk of calcium and magnesium, which can occur when high concentrations accumulates close to the membrane surface. The antiscalants modifies the scaling properties, while the NF has a good ability to retain the divalent ions.

pH (-)	Alkalinity (mg/l HCO_3^-)	Turbidity (FNU)	Fe (mg/l)	Mn (mg/l)	Ca (mg/l)
7.4 - 7.5	230 - 257	1.5	0.155	0.136	91

Table 4.2: Intervals of pH and alkalinity for water type 1, along with maximum values for the key foulants identified

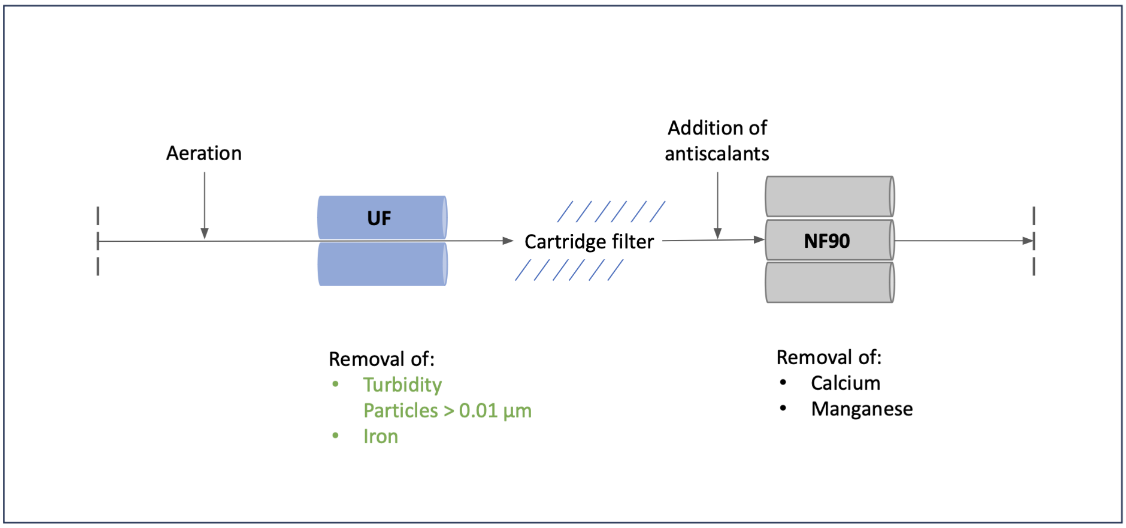


Figure 4.5: Pretreatment chain suggested for water type 1, including aeration, UF, cartridge filter and addition of antiscalants. The removal potential of key foulants is listed below the pretreatment methods, and the removal efficiency is characterised in different colours, where green, yellow and orange characterise good, moderate respectively fair removal.

Type water 1 represents a relatively stable groundwater, which has advantageous conditions to be treated by NF in a MDWTS. The water contains low levels of NOM and the high levels of calcium and magnesium can be managed with the use of antiscalants. In Sweden, the allowed dosage of antiscalants are restricted to 5 mg/l in LIVSFS 2022:12 (Swedish NFA, 2022). Further, the dosage of antiscalants can be limited by the concentrations of calcium, magnesium and antiscalants occurring in the retentate in relation to what concentration is allowed to discharge to recipients.

One of the five wells obtains levels of turbidity and iron that can concern the NF treatment efficiency. Therefore, it's taken into consideration for the suggested treatment chain, where aeration and low pressure membrane filtration are proposed. One alternative approach is to consider the possibilities to exclude the well with challenging values. However, since iron is a commonly occurring challenge for groundwater treatment, the suggested pretreatment chain accounts for application at more locations.

As mentioned above, the levels of manganese comes with a risk of NF fouling, according to guidelines from the NF membrane manufacturer. However, the removal of manganese is not heavily considered in the suggested pretreatment chain. Although some studies showed that UF have fairly good removal efficiency of manganese, manganese may require stronger oxidizers than iron to fully precipitate on an UF membrane. Implementing a BAC-filter to manage the levels of manganese will possibly result in higher removal efficiency but without the addition of a filter posterior the BAC-filter, discharged biomass risks to enter the feed water and NF system. To include a BAC-filter and an UF membrane to a groundwater might be necessary, but for water of this quality it might be too extensive. Therefore, a pilot study on the pretreatment chain presented is required to substantiate the choice.

Lastly, by implementing the proposed treatment chain and the posttreatment mentioned earlier, two microbial barriers are achieved. An additional barrier could be added if a dense UF membrane is chosen. However, this amount of barriers is only required for groundwaters affected by surface water and for considerable occurrence of pathogens.

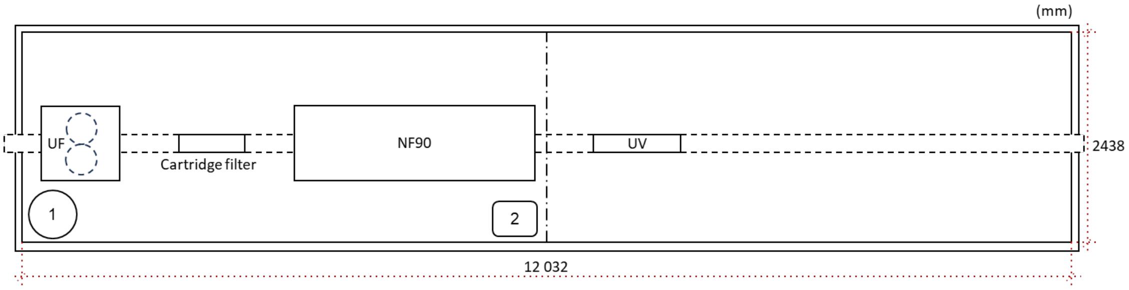


Figure 4.6: Drawing of the suggested pretreatment chain for water type 1 in a 40 ft container, with approximate dimensions given in mm. Tank 1 and 2 represents storage of oxygen/aeration and caustic soda, with sizes corresponding to approximately 125 and 200 liters.

4.2.2 Suggested pretreatment for water type 2

For the surface water representing water type 2, the pH values varies within the range 7.1-7.8. The low alkalinity is of significant concern when designing a pretreatment chain, due to operational aspects. Particles, colloids, NOM and iron, are identified as key foulants and furthermore the targets for the pretreatment. The characteristics of water type 2 is presented in Table 4.3 and the suggested pretreatment chain is shown in Figure 4.7, consisting of an aerated biofilter and an UF membrane. The aerated biofilter aims to target particles causing turbidity, iron and manganese. In addition, the biofilter has the ability to remove easily degradable NOM. Posterior the aerated biofilter, the water is pumped through an UF which further enhances the removal of turbidity and NOM. A MF is applied prior to the UF to ensure the operability and performance of the UF. In addition, the placement of the UF membrane allows it to work as catchment of biomass, potentially released from the biofilter. This solution has the potential to contain three microbial barriers, if a dense UF membrane is chosen and UV-treatment is implemented as posttreatment.

pH (-)	Alkalinity (mg/l HCO_3^-)	Turbidity (FNU)	Fe (mg/l)	TOC (mg/l)	Al (mg/l)
7.1 - 7.8	19 - 22	21	0.75	6	0.95

Table 4.3: Intervals of pH and alkalinity for water type 2, along with maximum values for the key foulants identified

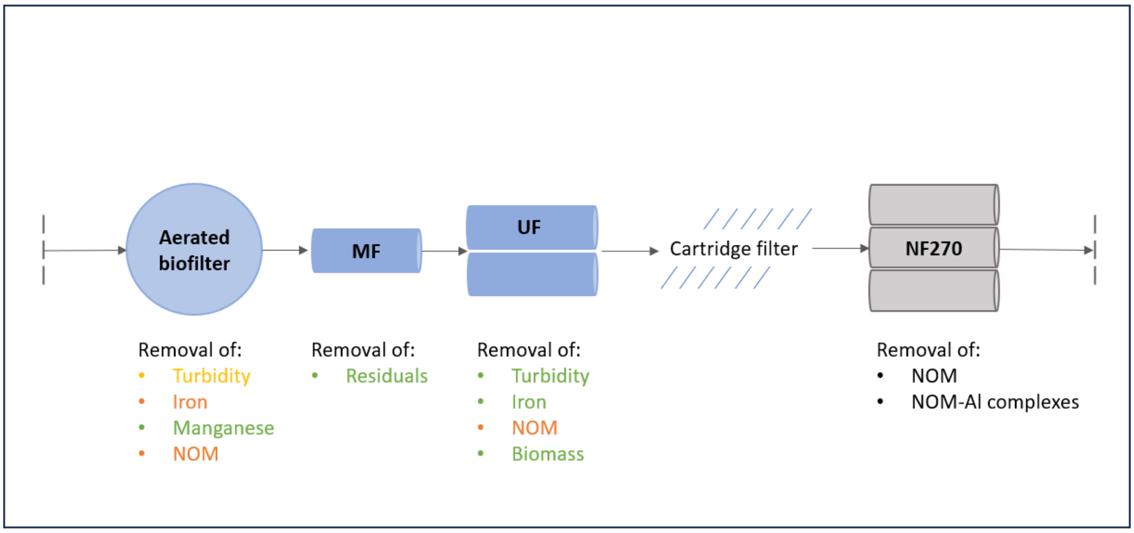


Figure 4.7: Pretreatment chain suggested for water type 2, including an aerated biofilter, MF, UF and cartridge filter.

Since the biofilter and UF membrane conduce limited removal of NOM, a large portion of NOM is allowed to enter the NF membrane. The amount of raw water components allowed to enter the NF membrane is a complicated trade-off. While the NF efficiency can get reduced due to organic fouling, the NF270 is developed to manage waters with high organic content and to remove NOM. While the biofilter and UF membrane has limited removal efficiency of DOM, the implementation of a biofilter in the pretreatment chain prevent unstable forms of NOM to enter the NF and can further reduce the risk of biofouling. The choice to let NF act as the main treatment step for organic matter is further motivated by the challenge to operate chemical flocculation due to the low alkalinity in this water.

Based on discussions with experts in drinking water treatment processes, both BAC filters and bioreactors are biological processes suitable for application in a MDWTS. These processes achieve approximate equivalent removal efficiency of key foulants, but have some operational differences. BAC-filtration requires a lower surface load than bioreactors, resulting in higher footprint. Conversely, bioreactors have a lower footprint but, due to the operation at higher velocities, there is a risk of sand being discharged to posterior processes. Potential discharge of biomass applies for both BAC-filtration and bioreactors, implying that both processes requires additional filtering prior to NF. Another aspect to consider regarding the operability of both the BAC-filtration and bioreactors is the importance of maintaining the biological activity to reach the expected removal efficiency. To summarise, the choice of biofilter depends on the spatial availability and the other pretreatment methods applied in the treatment chain.

In this water, the aluminium is assumed to be complexed to NOM. Depending on the influence the aerated biofilter and the UF membrane has on the Al-NOM complexes in the feed water and how the NF can manage the potential fouling of these, additional treatment steps may be required to target aluminium. However, for the aluminium levels shown for this surface water, the pretreatment chain shown above is suggested to be tested in pilot scale to determine the effect before taking any further measures.

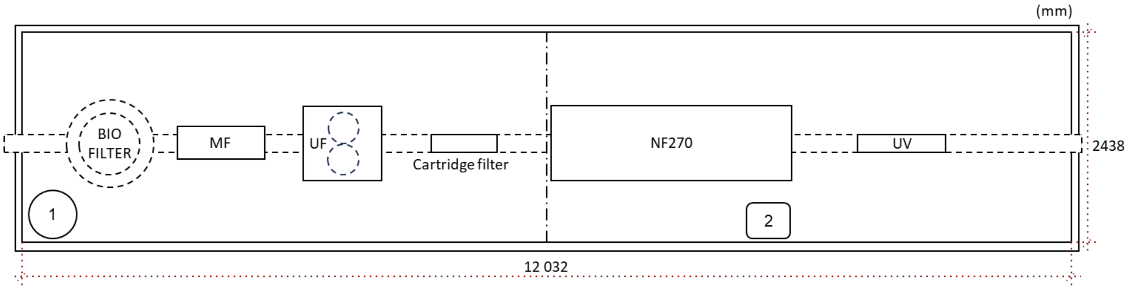


Figure 4.8: Drawing of the suggested pretreatment chain for water type 2 in a 40 ft container, with approximate dimensions given in mm. Tank 1 and 2 represents storage of oxygen/aeration and caustic soda, with sizes corresponding to approximately 125 and 200 liters

4.2.3 Suggested pretreatment for water type 3

The pretreatment suggested for the last water type is the most extensive, as can be seen in Figure 4.9. The pH-values for the surface water varies within the range 7.5-8 and the alkalinity is around 100 mg/l HCO_3^- . NOM is identified as a major key foulant for this water, alongside turbidity, iron, manganese and aluminium to a slightly smaller extent. The suggested pretreatment chain starts with an aerated biofilter, which mainly targets manganese and iron. Further, particles causing turbidity and partial NOM are removed as well. The biofilter is placed at the beginning of the process to mimic the natural process of groundwater creation. Since this water contains high levels of NOM and possesses a stable pH, chemical flocculation with ferric chloride and aluminium sulphate is inserted as a second step. The chemical flocculation is divided into two steps in order to remove different types of NOM. The process of chemical flocculation also reduces the turbidity. The addition of ferric chloride is followed by a rapid sand filter, with the aim to collect the created flocs and biomass from the earlier biological process. The addition of aluminium sulphate is followed by a UF with the purpose to collect other types of NOM. In order to protect the UF from residuals such as grains of sand, a MF is placed after the rapid sand filter.

pH (-)	Alkalinity (mg/l HCO_3^-)	Turbidity (FNU)	Fe (mg/l)	Mn (mg/l)	TOC (mg/l)	Al (mg/l)	Ca (mg/l)
7.5 - 8	97 - 120	92	12	1.1	40	2	54

Table 4.4: Intervals of pH and alkalinity for water type 3, along with maximum values for the key foulants identified

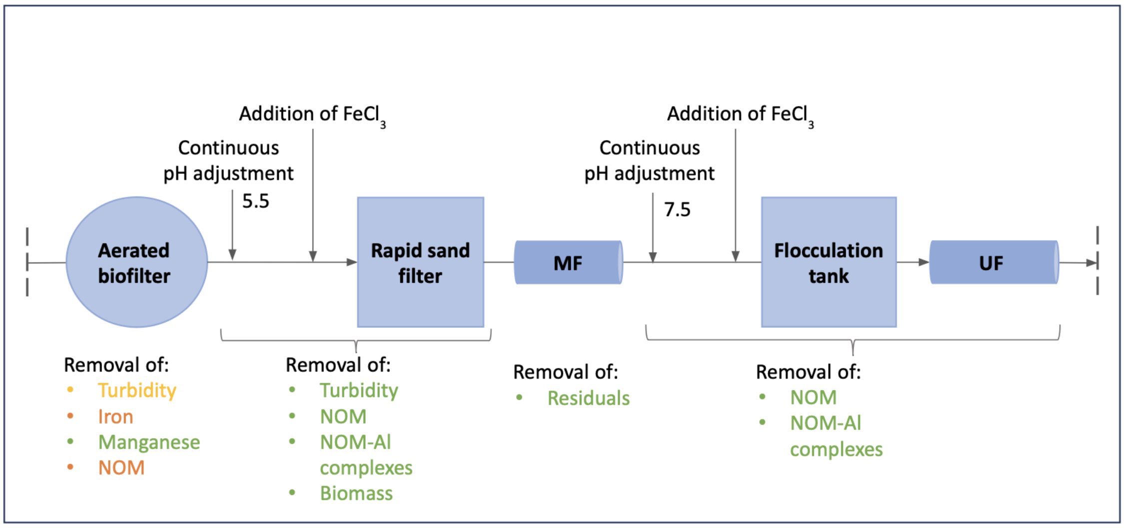


Figure 4.9: Pretreatment chain suggested for water type 3, including an aerated biofilter, chemical flocculation over a rapid sand filter, and lastly chemical flocculation over an UF

This water has extremely high values of especially NOM and turbidity, which leads to the discussion whether this water should be used as a raw water source for a MDWTS. In addition, waters with extreme values of multiple key foulants are not so common. These types of water require a multiple stage treatment chain, which become a problem when operation possibilities and space are limited in a MDWTS. In order to remove the high levels of NOM present in type water 3, chemical flocculation and ozonation in combination with a biofilter were considered. Processes like chemical flocculation and ozonation requires regularly monitoring, which is not optimal for a MDWTS placed in a rural area. However, for type water 3 chemical flocculation was chosen instead of ozonation due to the safety risks of ozonation. If not all ozone is removed in the earlier stages in the process, the possibility of destroying the membranes is severe.

In the treatment chain for water type 3, an UF was inserted instead of a NF. This decision was made since the pretreatment of the water resulted in a water which were considered as unnecessarily clean to treat in a NF. The chemical flocculation is a highly effective method to remove NOM, NOM-Al complexes and turbidity, and the aerated biofilter removes manganese and iron. However, if the calcium content/hardness had been higher, a NF is a better option since this step reduces the calcium content. In order to prevent calcium scaling in the NF, an antiscalant would have been added as well. In addition, by implementing chemical flocculation over an UF is more space beneficial compared to two additions of coagulant over two rapid filters followed by an UF.

The dense UF was also implemented in order to create an additional microbial barrier. Together with the chemical flocculation and the UV-treatment present in the posttreatment, three microbial barriers are generated. Even though the concentration of microorganisms are not considered in this project, three microbial barriers were desired. The barriers are considered as a necessary safety measure for surface waters in a MDWTS, where the process monitoring is not as extensive as in a centralised DWTP.

The addition of chemicals, results in the risk of residual coagulants causing fouling. In this project, ferric chloride and aluminium sulphate were considered as alternatives for the chemical flocculation. Since studies has conducted that aluminium residuals causes greater fouling problems, ferric chloride was considered as the favourable option. In addition, ferric chloride is beneficial for removing NOM. However, since NF is not implemented in this treatment chain, the coagulant residuals is not a severe problem. Chemical flocculation over a UF, requires a greater creation of flocs compared to flocculation over a sand filter, resulting in a longer retention time and the need for a flocculation tank. It is important to monitor the temperature of the water during the year, since this parameter highly affects the outcome of the treatment step.

The choice of adding coagulant twice during different pH conditions, is motivated by the different properties of NOM. A pH value of 5-5.5 is optimal for the flocculation of HMW hydrophobic NOM, whilst a higher pH enhances the removal of functional groups. In addition, the biological filtration process is favourable for the removal of LMW NOM. Biological processes also enhance the flocculation of the water. Compared to aluminium sulphate optimum pH between 5.5-6.5, ferric chloride's optimal pH is lower around 5-5.5, which could be seen as a disadvantage since it can result in an additional pH-adjustment. However, for type water 3, two pH-adjustment are already applied in order to remove different types of NOM. Therefore, there is no need of additional pH-adjustments.

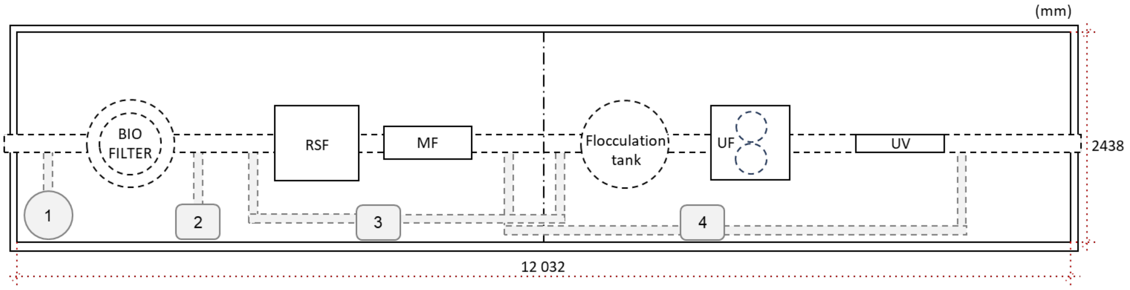


Figure 4.10: Drawing of the suggested pretreatment chain for water type 3 in a 40 ft container, with approximate dimensions given in mm. Tank 1, 2, 3 and 4 represents storage of oxygen/aeration, acid, $FeCl_3$ and caustic soda, with sizes corresponding to approximately 125 and 200 liters

4.2.4 Flowchart of key foulants with suggested pretreatment methods

Since the constituents of raw water varies a lot, it is hard to design a general solution which meets the requirements applicable for all raw waters. In many cases, a general solution implies over-dimensioning which is not defendable when it comes to economical means, operation and maintenance work, footprint, as well as the use of material, chemical and energy. This is of specific concern when designing a MDWTS, where most of these factors are even more limited. A possibility to reduce the risk of over-dimensioning, is to consider seasonal aspects by installing sensors measuring key foulants such as turbidity in the MDWTS. By doing this, it is possible to turn on and off different pretreatment steps depending on the concentrations of key foulants in the water.

Instead of creating one general solution, a flowchart containing the identified key foulants with suggested pretreatment methods is presented. This general solution is adapted for a MDWTS, which means that several pretreatment methods such as sedimentation, slow filtration and ozonation are excluded due to having a large footprint and/or involving risky procedures. The goal with this general solution is to create an easily operated and space effective process which can mitigate NF fouling and produce excellent drinking water. By following the flowchart for each key foulant, a pretreatment method is added to the treatment chain if the guideline is exceeded. At the end, when all the key foulants are reviewed, a pretreatment chain for a specific water is suggested. Before implementing pretreatment methods, the SDI should be tested and reviewed. If the SDI is exceeding 5 this could be used as an indication that pretreatment is needed. However, if the SDI is below 5, pretreatment should still be considered since dissolved substances also causes fouling. To develop the flowchart further, SDI can be incorporated in the procedure, either in connection to the turbidity parameter or on its own.

However, it is important to note that this is only a suggestion and all treatment chains should be tested in a pilot study before implementing it. In order to distinguish what effect each pretreatment method has on the NF fouling, it would be valuable to test them separately as well. Variations in the raw water composition during a year are also an argument to run a pilot study. Even if the pretreatment chain is designed based on the maximum values of the raw water constituents, it is preferable to test if the process can adapt and handle changes.

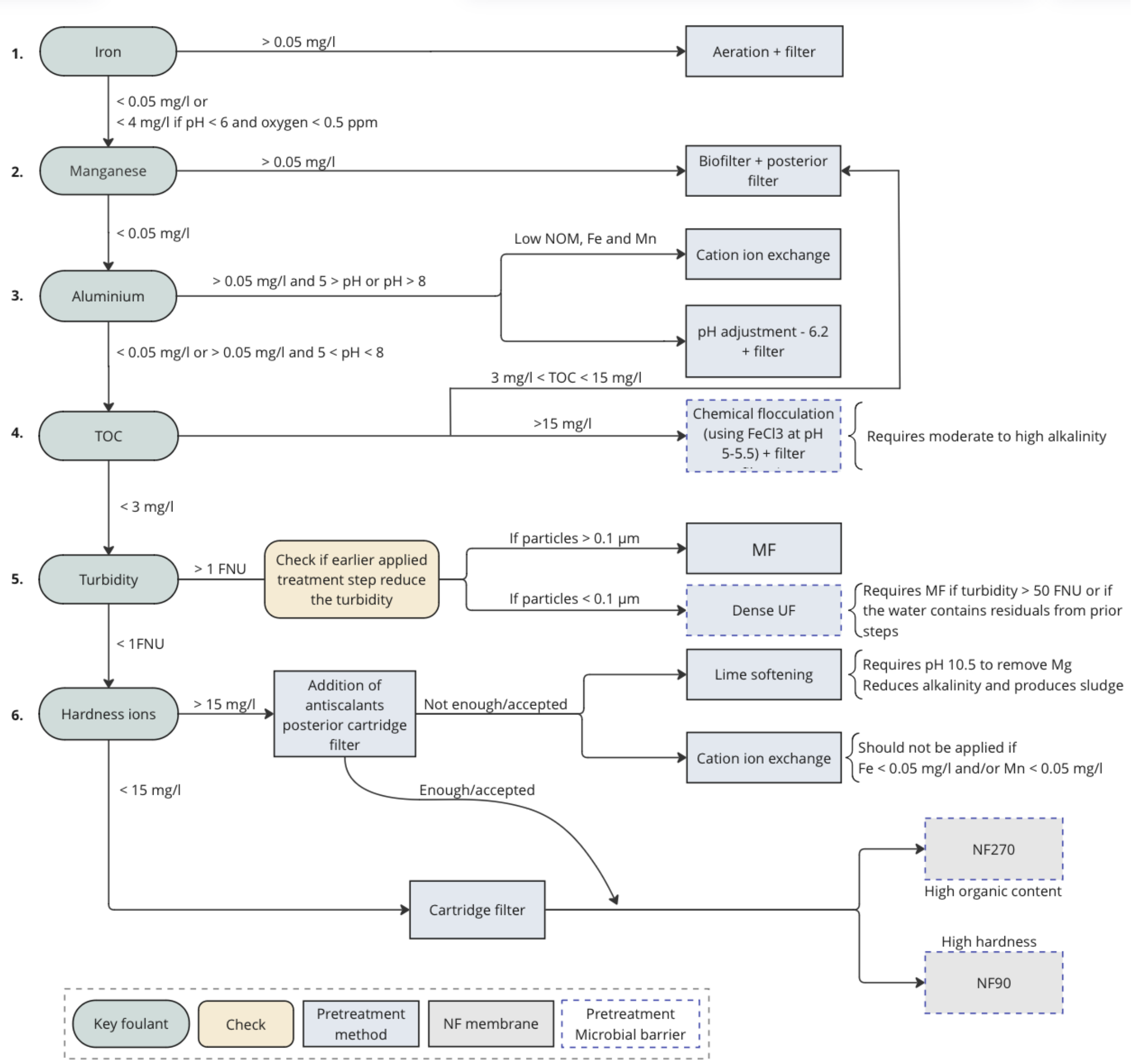


Figure 4.11: A general solution to select a suitable pretreatment method/chain, created as a flow chart

The guideline values for the key foulants are based on guidelines from a membrane manufacturer and discussions with experts within the field of drinking water processes. The guidelines for iron, manganese, aluminium and TOC are based on the membrane manufacturer's guidelines. As a result, the guidelines are strict since the manufacturer wants to protect the membrane and extend the lifetime, and therefore these guidelines are discussible. After all, NF membranes are made for removing organic and inorganic compounds and if the guidelines are too low, a NF is redundant. However, this discussion should be considered for each case. In some cases, a higher value of a key foulant may be acceptable.

The estimated guideline values include turbidity, calcium and the upper limit of 15 mg/l for TOC. Since these are estimated and not developed by a membrane manufacturer as the other guidelines, it is uncertain how these concentrations affect the fouling tendency. However, the measures to reduce the fouling potential of turbidity and calcium is relatively easy to implement. In comparison, higher concentrations of TOC require extensive treatment processes, such as chemical flocculation, which is not favourable in a MDWTS. In the flowchart, when TOC is between 3-15 mg/l, biofiltration followed by another filter is suggested as pretreatment. Biofiltration followed by filtration does not remove all the NOM, but it reduces the concentration. In combination with a NF270 membrane, which is made to treat water with organic content, this process was assessed to take care of the NOM. Still, if the TOC concentration approaches 15 mg/l, other measures may be relevant. As a result, if water contains high concentrations of NOM, it is crucial to characterize the types of NOM present in the water and to test different pretreatment methods in a pilot test. The pretreatment chain for type water 3 contains two steps of chemical flocculation, which is a possibility when the NOM concentrations are extremely high. However, this suggestion is not inserted in the flowchart since the synergy effect of the different steps is unknown.

In this solution, the focus is on removing the identified key foulants. The criteria with microbial barriers are also considered in the flowchart. Despite this, pollutants such as, PFAS, pesticides, pharmaceuticals, which can occur in raw water are not targeted. This is something that requires additional attention in order to make a complete process.

4.3 Bench study

In this section, results from the bench study are presented. The removal efficiency of key foulants for the investigated methods is further analysed, while identified sources of errors are discussed. All the chemical parameters measured during the bench study period are presented in tables in Appendix B.

4.3.1 Removal of key foulants

The removal of key foulants is presented in graphs, accounting for the concentrations of turbidity, TOC, aluminium, iron and manganese in the raw water and posterior the pretreatment methods. The methods' impact on calcium and magnesium was negligible for the charged sieve, while a fair reduction could be observed for the BAC filter. In contrast and in accordance with literature, the alkaline filter generated increased concentrations of calcium and magnesium. Silica was not included in the chemical analysis used in this bench study, and therefore the methods' impact on the concentration of silica remains uncertain.

The obtained turbidity in the raw water and in the effluent of the investigated methods are shown in Figure 4.12. The guidelines from Dupont are visualised as black lines in the graphs, and as can be seen, the turbidity in the raw water reaches values just above the guidelines. During the first sampling occasion, all the investigated methods reduced the turbidity. In contrast, the turbidity was increased for all the investigated methods during the second occasion, except for the uncharged sieve, which had no visible impact on the turbidity. During the third and final sampling occasion, the turbidity was decreased posterior the charged sieve, applied with 0 and 1 V. The BAC filter generates a moderate increase, while the alkaline filter increases the turbidity significantly. A possible explanation for this increase is that the BAC and alkaline filter drained at some point between the second and third sampling occasion. Without a continuous flow, components can easily accumulate in the filter, which can result in higher turbidity when the methods are put back into operation.

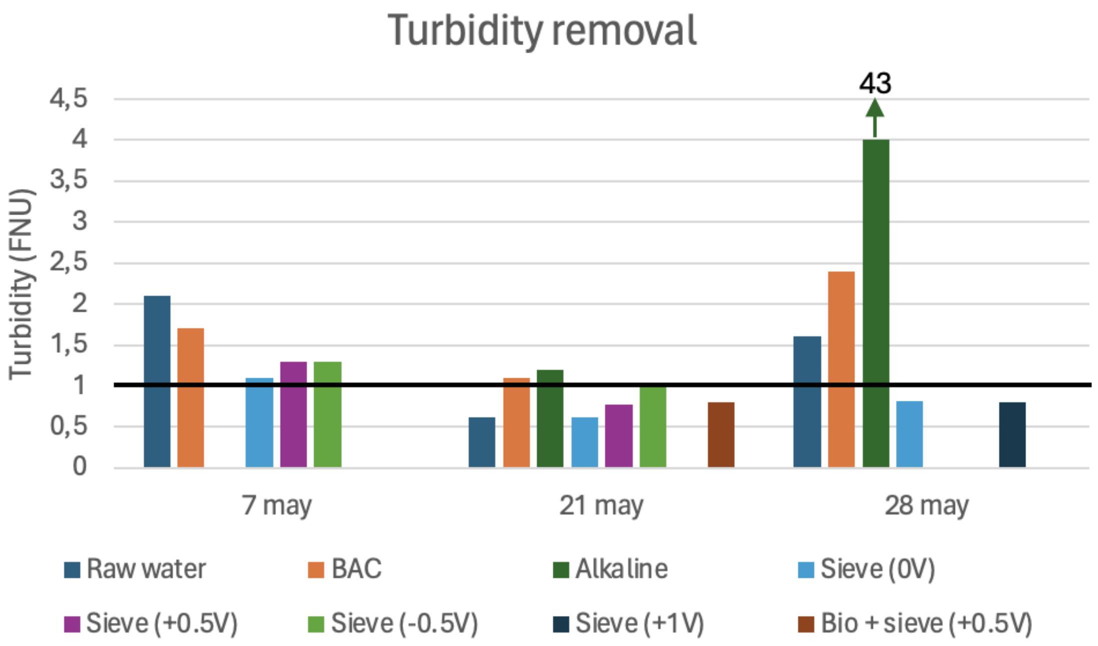


Figure 4.12: The turbidity obtained (in FNU) during the bench study period, for the raw water and effluent of the investigated methods. The results are grouped by the sampling occasions, and the black line corresponds to the guideline for feed water obtained from Dupont

The results for TOC from the water sampling of different pretreatment methods are presented in Figure 4.13. The first sample date did not result in any values for TOC due to sampling errors. For the second occasion, the alkaline filter and the BAC filter resulted in the greatest reduction. The uncharged sieve and the combination of BAC filter and charged sieve resulted in a minor reduction, whilst the sieve charged with -0.5V and +0.5V increased the level of TOC. During the third occasion, the BAC filter and the alkaline filter once again resulted in a reduction. However, this time the BAC filter showed a greater reduction of around 15% whilst the alkaline filter only reduced the concentration of TOC slightly. Both the uncharged sieve and the sieve charged with +1V increased the TOC concentration. Further, there is no significant difference between the different voltages. To conclude, none of the pretreatment methods reduced the TOC concentrations below the guideline of 3 mg/l.

The results for BAC are in accordance to the obtained values from the study of literature. However, the charged sieve was expected to reduce the organic content. The hypothesis for the charged sieve included removal of charged particles and according to the sampling results, most of the organic content in the water was dissolved. This raw water composition could explain the low reduction, and in some cases increase, of the organic content in the water. In addition, the tests which were performed at the end of the sessions resulted in a greater reduction compared to the first test. From the second occasion, the test with the uncharged sieve was performed last, resulting in the highest reduction. In comparison, sampling of effluent from the uncharged sieve was performed first during the third occasion, and resulted in the lowest reduction. A possible explanation to this is the use of filter papers, which led to different retention times. The first water sample was collected during half an hour, whilst the last took over an hour to collect. The reduction of the of TOC obtained from the combination of the BAC filter and the charged sieve are supposed to be completely dependent on the BAC filter.

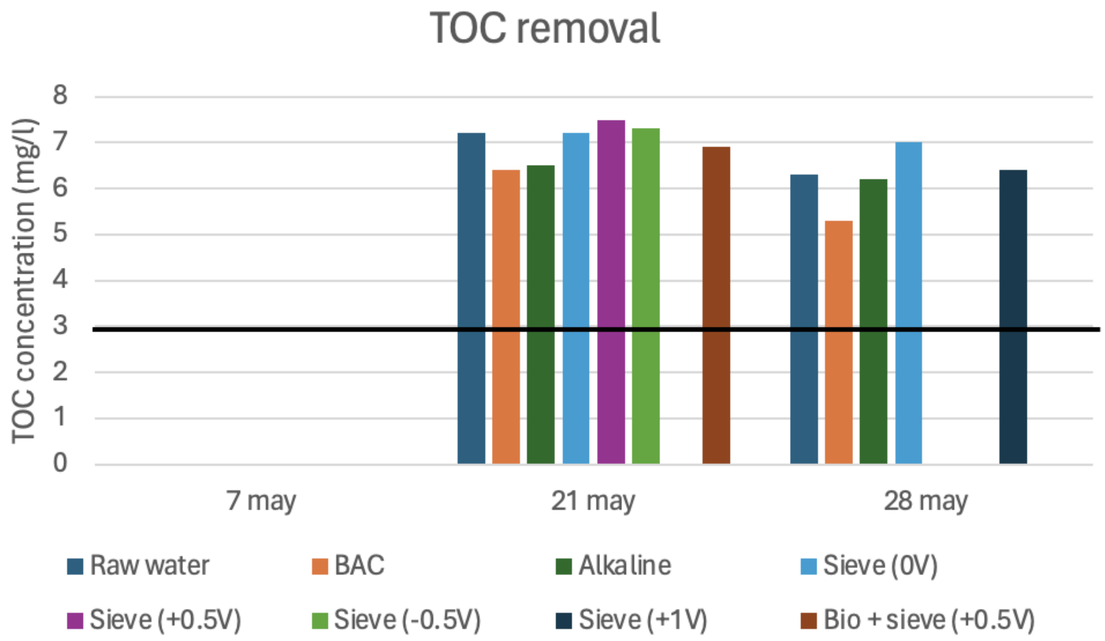


Figure 4.13: The TOC obtained (in mg/l) during the bench study period, for the raw water and effluent of the investigated methods. The results are grouped by the sampling occasions, and the black line corresponds to the guideline for feed water obtained from Dupont

For aluminium, the results from all three sampling occasions show an increased concentration of aluminium in the effluent of the BAC filter, which can be seen in Figure 4.14. In addition, the alkaline filter and the BAC filter combined with the charged sieve generated a slight increase of the aluminium concentration during the second occasion. There is no distinct explanation to the increased concentration of aluminium, but shifting flow conditions, allowing components to accumulate on the filters, may have influenced. The charged sieve show a negligible to moderate removal efficiency of aluminium, for all the applied voltages.

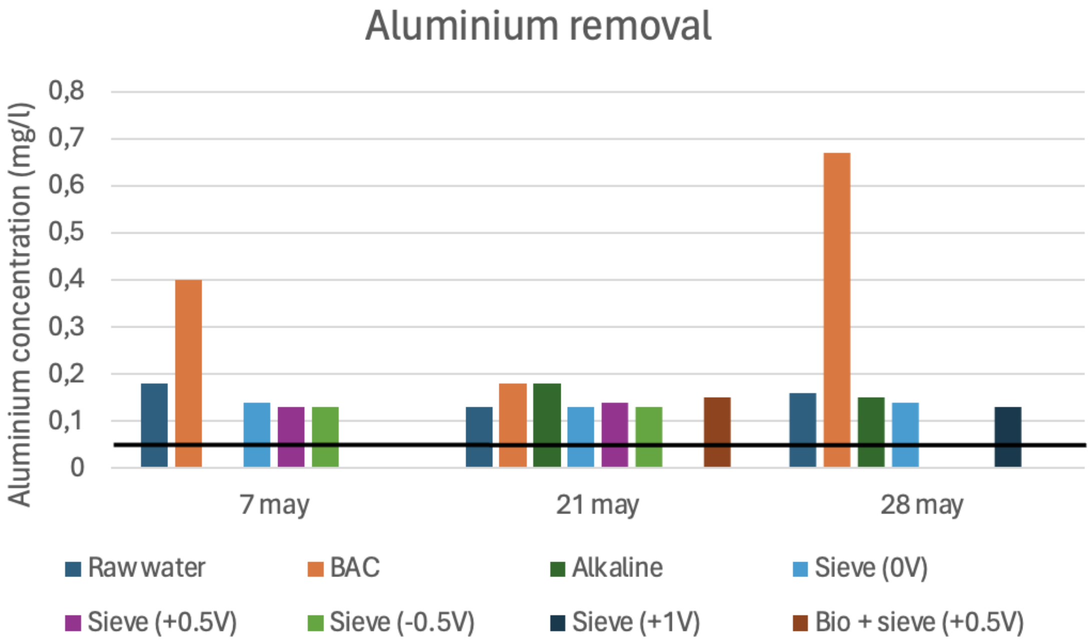


Figure 4.14: The aluminium obtained (in mg/l) during the bench study period, for the raw water and effluent of the investigated methods. The results are grouped by the sampling occasions, and the black line corresponds to the guideline for feed water obtained from Dupont

In Figure 4.15, the obtained reduction of iron for the pretreatment methods are visualized. All the investigated methods show a decrease of iron during the first and final sampling occasion, where BAC generated the largest removal, ranging around 35 to 65%. In contrast, the concentration of iron increases for the BAC and alkaline filter obtained from the second sampling occasion, while the other investigated methods have low influence on the iron concentration. The different voltages applied to the charged sieve generated a minor difference in the removal efficiency of iron, according to the observed results. Compared to when voltage is applied, the uncharged sieve has a slightly aggravated effect in the first sampling occasion, slightly improved effect in the second sampling occasion and no effect in the final sampling occasion. Although, most of the pretreatment methods resulted in reduction of iron, none of the concentrations are below the guideline for NF feed water.

Since the results vary widely, no distinct conclusions regarding the methods' performance are drawn. For the first and third occasion, BAC's removal of iron is in accordance with literature and the hypothesis for this bench study. The results from the second sampling session does not follow the same trend, but due to few data, it is unsure if the results are a one-time occurrence or if the methods possess a risk of increasing the iron concentration.

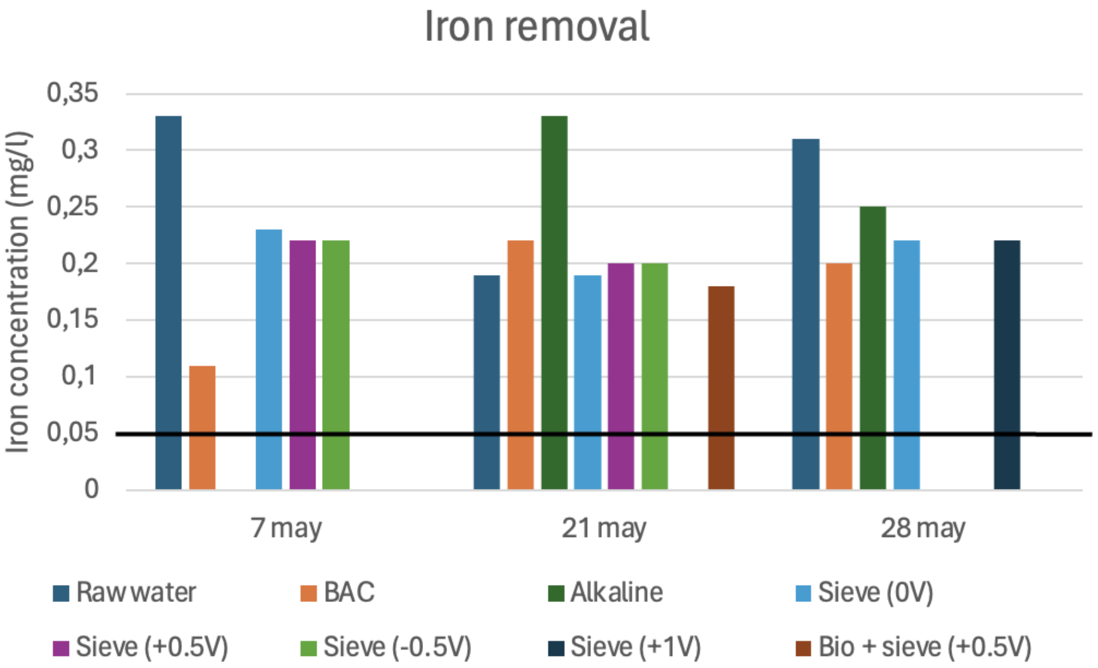


Figure 4.15: The iron obtained (in mg/l) during the bench study period, for the raw water and effluent of the investigated methods. The results are grouped by the sampling occasions, and the black line corresponds to the guideline for feed water obtained from Dupont

All the investigated methods show an ability to reduce the manganese concentration. During the first and final sampling occasion, BAC shows the greatest removal efficiency, while it generated an increase during the second sampling occasion. The voltage applied to the sieve made no difference for the removal efficiency of manganese, implying that the disc filter and filter papers had the greatest impact on the removal. In general, all the pretreatment methods reduced the manganese concentration to below the guidelines. On the other hand, the manganese was not highly exceeding the guideline value in the raw water.

The detection limit for the water sampling of manganese was 0.02, which makes it hard to compare the results correctly, since a majority of the results are around 0.02. In addition, the values are measured to the hundredths, resulting in less accurate results. For the BAC filter, the results from the first and third occasion match the hypothesis. This is the case for the last sampling for the alkaline filter as well. However, the second sampling occasion shows signs of error once again.

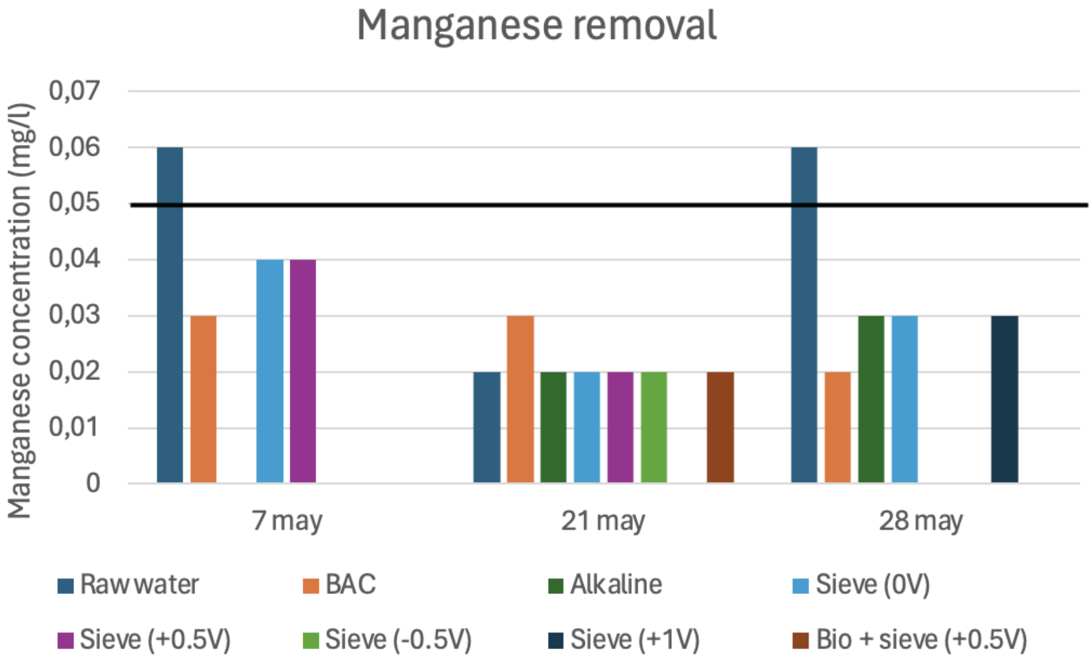


Figure 4.16: The manganese obtained (in mg/l) during the bench study period, for the raw water and effluent of the investigated methods. The results are grouped by the sampling occasions, and the black line corresponds to the guideline for feed water obtained from Dupont

4.3.2 Sources of error

To begin, water samples were only collected during three occasions. In addition, some of the methods were only tested during one or two occasions, resulting in few results. Preferably, tests should be performed during a year to catch the seasonal variations and get past the stabilization period. An additional identified source of error is connected to the container in the setup, which was filled with raw water before the water was distributed to the different pretreatment methods. As time went by, components in the raw water accumulated at the bottom and on the sides of the container. Further, the raw water samples were taken from the raw water inlet and not from the container where the concentrations of the key foulants possibly were higher. This could be the explanation for protruding values obtained during the second sampling occasion.

Another difficulty was to adjust the flows for the alkaline and BAC filter. Since the volumes of active material were small, the flows were low to achieve an EBCT of 30 minutes. The setups drained unintentionally at least two times during the bench study period, which can affect the removal efficiency of the filters depending on the time without water. For the charged sieve, three filter papers were used to prolong the retention time. The same filter papers were used for all tests with the charged sieve during one sampling occasion, which resulted in longer retention time for each collected water sample, due to accumulation of material and saturation of the filter.

To conclude, the alkaline filter and the BAC filter show good ability to remove some of the key foulants such as manganese and iron, whilst the charged sieve needs to be further developed. Suggestions of development for the charged sieve is to increase the retention time, try different voltages, or apply other alternatives to fulfil the aim of the filter papers. To validate all the results, tests should be performed during a longer period.

4.4 Further investigation

As mentioned before, pilot testing should always be performed before a treatment chain is implemented as a real water treatment system, providing people with clean water. Preferably, a study should be running for at least a year, in order to take account for a stabilisation period and seasonal variations. In this work, the time was limited and as a result pilot studies were not conducted. Therefore, in order to evaluate the suggested pretreatment chains for the water types, pilot studies should be performed. Although, the pretreatment chains are designed to handle the maximum values from the raw water source, unanticipated incidents may occur. In addition, a divided pilot test where each step is tested would be favourable to investigate the fouling mitigation tendency of each treatment step. The effect of silica on membrane fouling should also be further investigated in order to incorporate it into the flowchart.

The studies used different methods and measurements to investigate the NF fouling behaviour. The effect of fouling was measured in TMP drop, flux decline, total resistance and other calculations. Since the methods varied, it was hard to compare the results of obtained fouling mitigation. As a consequence, the results presented in Table 4.1 were not considered during the design of the pretreatment chains. Therefore, a proposal is to evaluate a universal method to measure fouling tendency of NF membranes.

Another approach to utilize the framework, is to create a pretreatment chain, adjusted to suit frequently/commonly occurring raw water qualities. This pretreatment chain could further be applied and produced in prefabricated modules, suitable for waters that obtain the quality within the dimensioned span. As mentioned earlier, to create a general solution will involve a trade-off between suiting as many raw water sources as possible and being over dimensioned, in the means of removal efficiency, footprint, material, operational and economical aspects. Regardless the pretreatment chain chosen, pilot studies for the specific raw waters are recommended to precede the implementation of prefabricated modules.

The potential pretreatment methods for NF fouling mitigation are not limited to the methods investigated within this work. Technologies such as electrocoagulation, charged membranes and the use of different adsorbents would be interesting to analyse and compare to the pretreatment methods investigated in this study. Although, the issue with complicated comparisons between the methods remains if not more studies on the removal of key foulants and fouling mitigation are conducted.

PFAS, pharmaceuticals, pesticides and other emerging contaminants are not considered in the pretreatment chains for the three water types nor in the general solution. Since the emerging contaminants can result in severe consequences for human health and the environment, an analysis of these emerging contaminants should be conducted for a MDWTS potential raw water. If the levels of emerging contaminants are problematic, the treatment process should be extended with the purpose to handle these contaminants. Another option is to exclude the raw waters containing emerging contaminants in a MDWTS.

5

Conclusion

The aim of this study was to suggest suitable pretreatment methods for raw water, implemented with the purpose to mitigate NF fouling in a MDWTS. To achieve this, a study of literature was performed, aiming to identify the components and factors contributing to NF fouling. Further, pretreatment methods applicable in a MDWTS were evaluated by their ability to remove key foulants and mitigate NF fouling. The findings were projected by designing pretreatment chains for three different water types occurring in Sweden. Given the varying pretreatment needs for these waters, it was not feasible to determine a single general pretreatment chain for the MDWTS. To address the challenge of over-dimensioning treatment processes in the limited space of a container, a general procedure to simplify the design of pretreatment chains was developed. In addition, bench scale tests were performed to evaluate two existing and one evolving pretreatment method.

Below, the main conclusions are listed:

- The key foulants impacting NF identified in this study are particles and colloids causing turbidity, NOM, iron, manganese, calcium, magnesium, aluminium and silica. To account for the key foulants is essential when determining suitable pretreatment.

- Several pretreatment methods applicable in a MDWTS possess the ability to remove key foulants and mitigate NF fouling, but the extent of the fouling mitigation is not well established in the literature. To understand the correlation between pretreatment and NF fouling, pilot studies are recommended, as well as a universal method to measure fouling.

- To design a MDWTS with NF as main treatment step, including pretreatment to enhance the NF performance, is spatially challenging but theoretical feasible. However, pilot studies are necessary to assure the performance of the pretreatment chain and the MDWTS.

- It is not feasible to treat all raw waters in a MDWTS due to spatial and operational limitations. Waters with high concentrations of organic matter may require chemical flocculation, which is not ideal for a MDWTS due to the operational complexity.

- Due to the risk of over-dimensioning the pretreatment chain, a general solution applicable for all raw water types can not be designed. However, a flowchart is developed to facilitate the procedure of designing a pretreatment chain for specific raw water types. Using this procedure, it is possible to design prefabricated modules tailored to commonly occurring water types.

- The results from the bench study determines the potential of removing manganese and iron for the alkaline and BAC filter. Although, the charged sieve should be further developed in order to evaluate its performance.

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A

Appendix 1

Pretreatment method	Specification	Turbidity	Organic matter	Hardness ions	Fe	Mn	Al	Si	Nanofiltration fouling mitigation	*Literature source*
MF Microfiltration	Surface water Off-line operation and hydrodynamic particle transport model	98%							Decrease of resistance to 17.4-27.5% Reduction of SDI to 28-50%	*Lee and Lee (2006)*
UF Ultrafiltration	Surface water Off-line operation and hydrodynamic particle transport model	98%							Decrease of total resistance to 31.5-45.7% Reduction of SDI to under detection limit	*Lee and Lee (2006)*
	Surface water from artificial lake Experiments on UF performance on removing NOM		22% (TOC)						Unknown	*Mijatovic et al (2004)*
	Synthetic and natural groundwater where UF was used as first treatment step (UF and NF used in combination)				92 - 99%	68 - 94%			Unknown	*Kasim et al (2016)*
	Synthetic groundwater cointaining humic acids where UF was used as first treatment step				95%	45%			Unknown	*Kasim et al (2016)*
	Synthetic surface water UF (0.02 mikron) before NF Removal efficiency of UF	62.2%	68.7% (DOC)		62.2%				Increase of NF permeate flux from 46% to 95% (49%-points)	*Sun et al (2012)*
RSF Rapid sand filtration	Seawater in SWRO facility Coagulant added to the feed water overlaying: A single medium RSF (Stage 1) A dual media RSF (Stage 2)		17-37% (TOC) 21-54% (TOC)						Unknown	*Bar-Zeev et al (2012)*
	Removal efficiency of SF Surface water from river Preozonation-coag-sedimentation-SF-O3-BAC-NF	94%	16% removal (DOC)						Fouling mitigation is estimated to not be adequately affected, due to residual NOM is low MW	*Chen et al (2021)*
AC Activated carbon	Tube well water and water from river GAC Batch test				83.20 - 99.48%	97.02 - 99.40%			Unknown	*Ismail et al (2017)*
	Bitumenic GAC before RO Wastewater Lab- and pilotscale test		80-90% (DOM) HPO and biodegrable						The major fraction of DOM which results in fouling is removed. Probably mitigate the organic fouling rather than biofouling.	*Gur-Reznik et al (2008)*
	Synthetic surface water Up-flow plastic column Hydraulic retention time=15 min GAC-NF, Removal efficiency of GAC	21.2%	17.8% (DOC)		21.2%				Increase of NF permeate flux from 46% to 48.8% (2.8 %-points)	*Sun et al (2012)*
	Sand filtration effluent Preliminary analysis at Buffalo Pound Water Treatment Plant						65 %			*Srinivasan et al (1999)*
	Groundwater pH 7.1-8.6 Jar test Permanganate oxidation followed by combined ion exchange		75% (DOC)	>55%					Calculated membrane fouling potential: Organic: 0.3 Inorganic: 0.1	*Apell et al (2010)*
	Groundwater pH 7.1-8.6 Jar test Permanganate oxidation followed by Anion exchange - Regenerated MIEX-Cl resin		>75% (DOC)	3%					Calculated membrane fouling potential: Organic: 0.25 Inorganic: 0.2	*Apell et al (2010)*

Pretreatment method	**Specification**	**Turbidity**	**Organic matter**	**Hardness ions**	**Fe**	**Mn**	**Al**	**Si**	**Nanofiltration fouling mitigation**	***Literature source***
IEX Ion exchange	Groundwater pH 7.1-8.6 Jar test Permanganate oxidation followed by Cation exchange - Regenerated MIEX-Na resin		20% (DOC)	54%					Calculated membrane fouling potential: Organic: 0.8 Inorganic: 0.4	*Apell et al (2010)*
	Unknown water source (obtained from compilation) Anion resin						0 - 20%			*Srinivasan et al (1999)*
	Unknown water source (obtained from compilation) Cation resin						90 - 100%			*Srinivasan et al (1999)*
BAC Biological activated carbon	BAC-UF-CEB Laboratory-scale UF, dead-end filtration GAC with coal-based broken carbon		60% (DOC)						Reduced TMP with 49%. Mitigate irreverible fouling with 78.7%.	*Yang et al (2023)*
	Synthetic water cointaining HA and sodium alginate BAC collected from a water plant BAC-NF, Removal efficiency of BAC	22.8%	29.2% (DOC)		22.8%				Decrease of permeate flux from 46% to 50.8% (4.8 %-points)	*Sun et al (2012)*
	Surface water Biofilter with sand	>50%	10% (TOC)		10 - 30%	60 - 80%			Unknown	*Unpublished*
COAG - FeCl3 Iron-based coagulants	Jar test Ferripol XL Dosage: 5-15 mg/l Natural water from river, pH=4-8 Optimum: 5-10-15 mg/l, pH=4,5-5	67%	80.5% (DOC)						Unknown	*Jarvis et al (2012)*
	Jar test FeCl3 Dosage: 15-90 mg/l Natural water from river, pH=5-10 Optimum: 65 mg/l and pH=7		57.9% (DOC)						Flux decrease: Al2(SO4)3 < FeCl3 < TiCl4	*Zhao et al (2014)*
	Jar test FeCl3 Dosage: 18-300 mg/l Source water, pH=7		51% (DOC), specifically hydrophobic and HA						Unknown	*Tubic et al (2013)*
	Fe III		80%							*Crittenden et al (2012)*
COAG - Al2(SO4)3 Al-based coagulant	Jar test Al2(SO4)3 Dosage: 5-50 mg/l Natural water from river, pH=4-9 Optimum: 30 mg/l and pH=7		43.2% (DOC)						Resulted in least membrane fouling potential compared to TiCl4 and FeCl3. Flux decrease: Al2(SO4)3 < FeCl3 < TiCl4	*Zhao et al (2014)*
	Jar test Al2(SO4)3 Dosage: 5-15 mg/l Natural water from river, pH=4-8 Optimum: 5-10-15 mg/l and pH=5-6	86%	77% (DOC)						Unknown	*Jarvis et al (2012)*
	Jar test Al2(SO4)3 Dosage: 0.2-10 mg/l Syntethic solution with DOC 8-13 mg/l Optiumum: 1.6 mg/l and pH=5.5-6.2		83% (DOC) 65% (HS)						Unknown	*Pivokonsky et al (2015)*
	Jar test-NF Al2(SO4)3 Dosage: 4 mg/g DOC Removal efficiency of coagulation	45.6%	59.6%		45.6%				Increase of permeate flux from 46% to 67.7% (21.7%-points)	*Sun et al (2012)*
	Alum III		70%							*Crittenden et al (2012)*

Pretreatment method	Specification	Turbidity	Organic matter	Hardness ions	Fe	Mn	Al	Si	Nanofiltration fouling mitigation	*Literature source*
OX Oxidation	Surface water from artificial lake O3 with PAC-filtration Ozone dose 0.5-2.1 mg/l				Under detection limit (~>94%)	Under detection limit (~>94%)				*Mijatovic et al (2004)*
	Surface water (moderate quality, subject to algal growth) and synthetic surface water Ozone dose: 0-1mg/l								Slight decrease of NF fouling for dense NF (5%) Slight increse of NF fouling for loose NF	*Yu et al (2018)*
	Unknown water source (obtained from compilation) Chemical oxidation						0 - 20%			*Srinivasan et al (1999)*
	Unknown water source (obtained from compilation) Aeration and stripping						0 - 20%			*Srinivasan et al (1999)*
LS Lime softening	Unknown water source (obtained from compilation)						40 - 70%			*Srinivasan et al (1999)*
	Uknown water source (statement from the US EPA)		40 - 80% (TOC)							*Crittenden et al (2012)*
Treatment combinations	In-line coagulation and UF pH=7 Pretreated water:		53% (DOC)						Positive effects on membrane fouling process (reduced fouling rate, more constant permeate flux, very slight increase in TMP)	*Wang and Wang (2006)*
	Coag + UF (prior to RO) brackish water G=2000s^-1, pH=7.1, Al2O3-dosage=30mg/L							65%		*Cheng et al (2009)*
	Surface water Treatment chain combining MF and UF	67.5 - 89%		None					Unknown	*Podaru et al (2008)*
	Coag+UF+NF Removal efficicency of Coag+UF	72.2%	78.8% (DOC)		72.2%				Increase of permeate flux from 46% to 95% (49%-points)	*Sun et al (2012)*
	UF+BAC+NF	79.5%	77.3% (DOC)		79.5%				Increase of permeate flux from 46% to 95% (49%-points)	*Sun et al (2012)*
	Surface water from river Removal efficiency of SF-O3-BAC Preozonation-coag-sedimentation-SF-O3-BAC-NF	95%	19% (DOC)						Fouling mitigation is estimated (due to residual NOM has high MW) but not proved	*Chen et al (2021)*
	Coag+BAC+NF Removal efficiency of Coag+BAC	76.7%	76.5% (DOC)		76.7%				Increase of permeate flux from 46% to 98% (52%-points)	*Sun et al (2012)*

B

Appendix 2

Table B.1: Water quality parameters for the untreated raw water at a DWTP

Parameter	**Unit**	**Result 30/4**	**Result 7/5**	**Result 21/5**	**Result 28/5**
Turbidity	FNU	1.9	2.1	1.2	1.6
Colour	mg/l Pt	60	50	60	60
Conductivity 25°C	mS/m	9.96	9.75	9.91	9.74
pH 20°C	-	6.9	6.9	6.8	6.7
Alkalinity	mg/l HCO3-	11	12	11	12
COD-Mn	mg/l	7.4	8.1	7.2	6.6
NH4-N	mg/l	0.028	0.032	0.025	0.024
NH4	mg/l	0.04	0.04	0.03	0.03
NO2-N	mg/l	<0.001	<0.001	<0.001	<0.001
NO2	mg/l	<0.004	<0.004	<0.004	<0.004
Aluminium	mg/l	0.19	0.18	0.15	0.16
Iron	mg/l	0.29	0.33	0.23	0.31
Calcium	mg/l	5.1	5.0	5.2	5.3
Copper	mg/l	<0.02	<0.02	<0.02	<0.02
Magnesium	mg/l	1.4	1.3	1.4	1.4
Manganese	mg/l	0.04	0.06	0.03	0.06
Hardness	dH	1	0.99	1	1.1
TOC	mg/l	6.8	7.1	7.0	6.3
DOC	mg/l	6.6	6.9	6.6	6.3

Table B.2: Results from the water sampling of the BAC filter

Parameter	**Unit**	**7/5**	**21/5**	**28/5**
Turbidity	FNU	1.7	1.1	2.4
Colour	mg/l Pt	15	50	40
Conductivity 25°C	mS/m	10.1	9.93	11.2
pH 20°C	-	6.6	6.7	6.8
Alkalinity	mg/l HCO3-	N.A	11	15
COD-Mn	mg/l	N.A	7.0	4.3
NH4-N	mg/l	0.30	0.040	1.6
NH4	mg/l	0.39	0.05	2.1
NO2-N	mg/l	<0.001	<0.001	<0.001
NO2	mg/l	<0.004	<0.004	<0.004
Aluminium	mg/l	0.40	0.18	0.67
Iron	mg/l	0.11	0.22	0.20
Calcium	mg/l	N.A	5.0	4.7
Copper	mg/l	N.A	<0.02	<0.02
Magnesium	mg/l	N.A	1.3	1.3
Manganese	mg/l	0.03	0.03	<0.02
Hardness	dH	N.A	0.99	0.95
TOC	mg/l	N.A	6.4	5.3
DOC	mg/l	N.A	6.2	5.0

Table B.3: Results from the water sampling of the uncharged sieve

Parameter	**Unit**	**7/5**	**21/5**	**28/5**
Turbidity	FNU	1.1	0.62	0.81
Colour	mg/l Pt	50	60	50
Conductivity 25°C	mS/m	9.85	10	10
pH 20°C	-	6.8	6.8	6.9
Alkalinity	mg/l HCO3-	N.A	12	12
COD-Mn	mg/l	N.A	7.3	7.4
NH4-N	mg/l	0.048	0.043	0.16
NH4	mg/l	0.06	0.06	0.21
NO2-N	mg/l	<0.001	<0.001	0.0013
NO2	mg/l	<0.004	<0.004	0.004
Aluminium	mg/l	0.14	0.13	0.14
Iron	mg/l	0.23	0.19	0.22
Calcium	mg/l	N.A	5.4	5.2
Copper	mg/l	N.A	<0.02	<0.02
Magnesium	mg/l	N.A	1.4	1.4
Manganese	mg/l	0.04	0.02	0.03
Hardness	dH	N.A	1.1	1.0
TOC	mg/l	N.A	7.2	7.0
DOC	mg/l	N.A	7.9	7.2

Table B.4: Results from the water sampling of the positively charged sieve

Parameter Specification	**Unit** V	**7/5** +0.5	**21/5** +0.5	**28/5** 1
Turbidity	FNU	1.3	0.77	0.8
Colour	mg/l Pt	50	60	60
Conductivity 25°C	mS/m	9.84	10.1	9.94
pH 20°C	-	6.7	6.8	6.9
Alkalinity	mg/l HCO3-	N.A	12	12
COD-Mn	mg/l	N.A	7.6	6.7
NH4-N	mg/l	0.045	0.056	0.023
NH4	mg/l	0.06	0.07	0.03
NO2-N	mg/l	N.A	<0.001	0.0012
NO2	mg/l	N.A	<0.004	<0.004
Aluminium	mg/l	0.13	0.14	0.13
Iron	mg/l	0.22	0.20	0.22
Calcium	mg/l	N.A	5.4	5.4
Copper	mg/l	N.A	<0.02	<0.02
Magnesium	mg/l	N.A	1.4	1.4
Manganese	mg/l	0.04	0.02	0.03
Hardness	dH	N.A	1.1	1.1
TOC	mg/l	N.A	7.5	6.4
DOC	mg/l	N.A	7.3	6.4

Table B.5: Results from the water sampling of the negatively charged sieve

Parameter Specification	**Unit** V	**7/5** -0.5	**21/5** -0.5
Turbidity	FNU	1.3	1.0
Colour	mg/l Pt	50	60
Conductivity 25°C	mS/m	9.84	9.94
pH 20°C	-	6.7	6.9
Alkalinity	mg/l HCO3-	N.A	13
COD-Mn	mg/l	N.A	7.6
NH4-N	mg/l	0.045	0.032
NH4	mg/l	0.06	0.04
NO2-N	mg/l	N.A	<0.001
NO2	mg/l	N.A	<0.004
Aluminium	mg/l	0.13	0.13
Iron	mg/l	0.22	0.20
Calcium	mg/l	N.A	5.4
Copper	mg/l	N.A	<0.02
Magnesium	mg/l	N.A	1.4
Manganese	mg/l	0.04	0.02
Hardness	dH	N.A	1.1
TOC	mg/l	N.A	7.3
DOC	mg/l	N.A	7.2

Table B.6: Results from the water sampling of the alkaline filter

Parameter	**Unit**	**21/5**	**28/5**
Turbidity	FNU	1.2	43
Colour	mg/l Pt	60	60
Conductivity 25°C	mS/m	12.4	14.5
pH 20°C	-	7.4	8.6
Alkalinity	mg/l HCO3-	27	69
COD-Mn	mg/l	7.1	6.2
NH4-N	mg/l	0.024	<0.01
NH4	mg/l	0.03	<0.02
NO2-N	mg/l	<0.001	<0.001
NO2	mg/l	<0.004	<0.004
Aluminium	mg/l	0.18	0.15
Iron	mg/l	0.33	0.25
Calcium	mg/l	10	29
Copper	mg/l	<0.02	<0.02
Magnesium	mg/l	1.5	2.2
Manganese	mg/l	0.02	0.03
Hardness	dH	1.7	4.5
TOC	mg/l	6.5	6.2
DOC	mg/l	6.6	6.0

Table B.7: Results from the water sampling of the treatment combination (BAC filter and charged sieve)

Parameter	**Unit**	**21/5**
Turbidity	FNU	0.80
Colour	mg/l Pt	50
Conductivity 25°C	mS/m	10.2
pH 20°C	-	6.7
Alkalinity	mg/l HCO3-	12
COD-Mn	mg/l	7.0
NH4-N	mg/l	0.054
NH4	mg/l	0.07
NO2-N	mg/l	<0.001
NO2	mg/l	<0.004
Aluminium	mg/l	0.15
Iron	mg/l	0.18
Calcium	mg/l	5.6
Copper	mg/l	<0.02
Magnesium	mg/l	1.4
Manganese	mg/l	<0.02
Hardness	dH	1.1
TOC	mg/l	6.9
DOC	mg/l	7.0

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