



Panu Laurell

**Retrofitting a water treatment plant with low pressure  
membranes – Preliminary study on retrofitting Pitkääkoski  
water treatment plant**

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for the degree of Master of Science in Technology.

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Supervisor: Professor Riku Vahala

Advisors: Dr.Eng Mehrdad Hesampour  
M.Sc.Tech. Heli Härkki

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**Author** Panu Laurell

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**Thesis supervisor** Professor Riku Vahala

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**Thesis advisors** Dr.Eng Mehrdad Hesampour, M.Sc.Tech Heli Härkki

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## Abstract

During the recent decades an increase in natural organic matter (NOM) has been noted in Nordic countries. At Pitkääkoski water treatment plant this has spurred on research about the effect of NOM increase to water treatment process. Simultaneously the water treatment plant is looking to increase its production capacity. Low pressure membranes (LPM), i.e. micro- (MF) and ultrafiltration (UF) membranes have emerged as a viable alternative to conventional treatment, as they separate particles effectively and can be combined with pretreatments such as coagulation to achieve high NOM removals. However, simultaneously NOM is a major component causing membrane fouling.

The diversity of ways to retrofit LPMs to the existing water treatment plants proves the robustness of MF and UF membranes. Several retrofits were studied and the findings show that most often LPMs are used as a final polishing step before water is delivered to customers or to replace sedimentation and sand filtration. If raw water is of good quality membranes can be used for direct filtration, but in over half of the cases studied LPMs were combined with pretreatments such as coagulation, adsorption, or prefiltration.

By studying existing retrofits two possibilities to retrofit LPMs to Pitkääkoski water treatment plant emerged. These two possibilities were studied intensely by using a MF membrane by Pall, and the performance, both operational data and water quality, were compared to conventional treatment currently in operation. When treating clarified water after sedimentation the MF membrane experienced a smaller initial drop in permeability compared to flocculated water, but the fouling rate after initial drop was faster. After two days the permeability with both feed waters was the same.

MF membrane was able to compete with conventional treatment, consisting of sedimentation and sand filtration, regarding water quality. Membranes are very effective in removing particles and microorganisms which were shown by low turbidity, low microbial growth, and residual iron in permeate. NOM removal was similar to conventional treatment. Membranes were shown to effectively remove aromatic and hydrophobic fraction of NOM, however, conventional treatment was more effective in removing hydrophilic fraction. Quality of permeate from MF was constant throughout the pilot and very little differences were found between two different feed waters.

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**Keywords** Low pressure membranes; membrane filtration; retrofitting; fouling.

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### Tiivistelmä

Viime vuosikymmeninä orgaanisen aineksen (NOM) määrän on todettu kasvavan Pohjoismaissa. Pitkääkosken vedenpuhdistamolla kasvu on johtanut tutkimuksiin NOM:n kasvun vaikutuksista veden puhdistukseen. Samalla puhdistamo pyrkii kasvattamaan kapasiteettiaan. Matalapaineiset kalvot (LPM), eli mikro- (MF) ja ultrasuodatuskalvot (UF) ovat nousseet varteenotettavaksi vaihtoehtoksi perinteisille käsittelyille, sillä ne poistavat partikkeleita tehokkaasti ja yhdistettynä esikäsittelyihin, kuten saostukseen, ne kykenevät poistamaan myös NOM:ta tehokkaasti. Samanaikaisesti kuitenkin NOM on suurin kalvojen tukkeutumista aiheuttava komponentti.

LPM:en jälkiasennusten monimuotoisuus vedenpuhdistamoilla osoittaa MF- ja UF-kalvojen toimivuuden. Tätä tutkimusta varten tutkittiin useita jälkiasennuksia ja tulokset osoittavat, että LPM:ja käytetään usein joko viimeistelevänä käsittelynä ennen vedenjakelua tai korvaamaan selkeytys ja hiekkasuodatus. Mikäli laitokselle saapuvan raakaveden laatu on hyvä, kalvoja voidaan käyttää ilman esikäsittelyjä, mutta yli puolessa tutkituista tapauksista LPM:ja käytettiin yhdistettynä esikäsittelyihin, joita ovat mm. saostus, adsorptio ja esisuodatus.

Toteutettuja jälkiasennuksia tarkastelemalla löytyi kaksi mahdollista tapaa jälkiasentaa LPM:t Pitkääkosken vedenpuhdistamolle. Näiden kahden vaihtoehdon toimivuutta testattiin tiiviisti käyttämällä Pallin valmistamaa MF-kalvoa. Suorituskykyä verrattiin niin toiminnan kuin veden laadun avulla perinteisten käsittelyiden suorituskykyyn. Selkeytyksen jälkeen otetulla selkeytetyllä syöttövedellä ominaisvuo MF-kalvon läpi heikkeni heti suodatuksen alussa vähemmän kuin saostetulla syöttövedellä mutta tukkeutumisen nopeus alkunotkahduksen jälkeen oli nopeampaa. Kahden päivän jälkeen läpäisevyys oli sama molemmilla syöttövesillä.

MF-kalvo osoittautui kilpailukykyiseksi perinteisiä käsittelyjä vastaan kun vertailtiin tuotetun veden laatua. Perinteiset käsittelyt tässä tarkoittavat sedimentaatiota ja hiekkasuodatusta. Kalvo oli erittäin tehokas poistamaan partikkeleita ja mikro-organismeja, tämän osoittivat alhainen sameus, mikrobien kasvu ja jäännösrauta permeaatissa. NOM:n poisto oli yhtä tehokas kuin perinteisillä menetelmillä. Kalvo poisti tehokkaasti aromaattisia ja hydrofobisia NOM-yhdisteitä, mutta perinteiset käsittelyt olivat tehokkaampia poistamaan hydrofiilistä fraktiota. Permeaatin laatu MF-kalvolla pysyi vakiona koko pilotin ajan ja eri syöttövesien vaikutus permeaatin laatuun vähäinen.

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**Avainsanat** Matalapaineikalvot; kalvosuodatus; jälkiasennus; tukkeutuminen.

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---

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## Sammandrag

Under de senaste decennierna ökning av organiskt material (NOM) har noterats i de nordiska länderna. På Långforsens vattenreningsverk detta har sporrat till forskning om effekten av NOM ökning i vattenrening process. Samtidigt reningsverket är ute efter att öka sin produktionskapacitet. Lågtryck membran (LPM), dvs. mikro- (MF) och ultrafiltrerings (UF) membran har dykt upp som ett alternativ till konventionell vattenbehandling, eftersom de separerar partiklar effektivt och när de kombineras med förbehandlingar som koagulering, effektiv avledning av NOM nås. Men samtidigt NOM är en viktig komponent som orsakar membran igensättning.

Mångfalden av olika sätt att utrusta LPMs till befintliga vattenreningsverken bevisar robustheten av MF och UF-membran. Flera ombyggnader studerades och resultaten visar att oftast LPMs används som en sista behandling steg innan vattnet levereras till kunden eller för att ersätta sedimentering och sandfiltrering. Om råvattnet är av god kvalitet membran kan användas för direkt filtrering, men i över hälften av de undersökta fallen LPMs kombinerades med förbehandlingar som koagulering, adsorption eller förfiltrering.

Genom att studera befintliga ombyggnader två möjligheter att ombygga LPMs till Långforsens vattenreningsverk uppstod. Dessa två möjligheter studerades intensivt med hjälp av en MF-membran från Pall och prestanda, både operativa data och vattenkvalitet jämfördes med konventionell behandling för närvarande används på vattenreningsverket. Vid behandling av sedimenterat vatten MF membranet upplevde en mindre inledande minskning i permeabilitet jämfört med flockat vatten, men hastigheten av igensättning efter initial nedgång var snabbare. Efter två dagar permeabiliteten med båda matningsvatten var densamma.

MF-membran kunde konkurrera med konventionell vattenbehandling, bestående av sedimentering och sandfiltrering, gäller vattenkvalitet. Membran är mycket effektivt för att avlägsna partiklar och mikroorganismer som visades genom låg grumlighet, låg mikrobiell tillväxt, och låg återstående järn i permeat. Effektivitet i NOM borttagning liknade konventionell vattenbehandling. Membran var effektivt att avlägsna aromatiska och hydrofoba fraktionen av NOM, emellertid konventionell vattenbehandling var mer effektiva i att avlägsna hydrofila fraktionen. Kvalitet av permeat från MF var konstant under hela piloten och små skillnader påträffades mellan två olika matningsvatten.

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**Nyckelord** Lågtryck membran; membranfiltrering; ombyggnad; igensättning.

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## Foreword

First and foremost I want to apologise to my friends. Three years ago I decided to graduate by the end of 2012, a decision I was able to keep. However, during this process I continuously neglected my friends in order to achieve the goal I had set. Now it is time to make amends.

This thesis was done as a part of ADWATECH-project, which is funded by Tekes, HSY, and Kemira. I'd like to express my gratitude to aforementioned participants for making it possible for me to work with such fascinating subject.

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# Nomenclature

## Abbreviations

AOC	Assimilable organic carbon
CA	Cellulose acetate
CAPEX	Capital expenses
CEB	Chemically enhanced backwash
CIP	Cleaning in place
COP	Cleaning out of place
DBP	Disinfection-by-product
DOC	Dissolved organic carbon
EPS	Extracellular polysaccharides
GAC	Granular activated carbon
GFD	Gallons per square foot per day
HPSEC	High performance size-exclusion chromatography
HSY	Helsinki Regional Environmental Services Authority
LC-OCD	Liquid chromatograph – organic carbon detector
LPM	Low pressure membrane
MF	Microfiltration
MIEX	Magnetic ion-exchange
MW	Molecular weight
MWCO	Molecular weight cut-off
NF	Nanofiltration
NOM	Natural organic matter
OCD	Organic carbon detector
OPEX	Operational expenses
PAC	Powdered activated carbon
PES	Polyethersulfone
PLC	Programmable logic controller
PP	Polypropylene
PS	Polysulfone
PVDF	Polyvinylidene fluoride
RO	Reverse osmosis
SF16	Sand filter 16
SOPH	Sum of all peak heights

SUVA	Specific UV absorbance
THM	Trihalomethane
TMP	Transmembrane pressure
TOC	Total organic carbon
UF	Ultrafiltration
WTP	Water treatment plant

#### **Greek alphabet**

$\Delta$	transmembrane
$\mu$	viscosity

#### **Roman alphabet**

A	surface area
C	concentration
J	flux
P	pressure
Q	flow
R	resistance
S	reduction
T	temperature
t	time
V	volume of water
Y	turbidity

#### **Subscripts**

a	average
b	backwashing
f	feed
i	irreversible
k	specific
M	measured
m	membrane
O	operation
o	outlet
P	produced
p	permeate
r	reversible
S	standard

# 1 Introduction

## 1.1 Background

Surface water is a common source of potable water in the world. Most of the population in Helsinki Region is supplied with potable water from surface water originated at Lake Päijänne some 100 kilometres north of Helsinki. Raw water from Päijänne can be characterised as having low alkalinity, low turbidity, low hardness, but elevated natural organic matter (NOM), represented by total organic carbon (TOC) values averaging over 7.5 mg/l and  $\text{KMnO}_4$ -values around 26.0 mg/l. A growing concern in Nordic countries in potable water treatment is the increase of NOM in surface waters, which has been noted also by Helsinki Regional Environmental Services Authority (HSY), authority in charge of potable water production. The changes in climate, especially in precipitation and drainage patterns, have been suggested as the reasons for this increase in NOM (Eikebrokk, 2012).

The removal of NOM requires an advanced water treatment. Fractions of NOM can cause colour, taste, and odour problems, react with chlorine to form carcinogenic disinfection-by-products (DBP), and facilitate bacterial growth in the supply system. NOM removal of the conventional treatment can be enhanced by adding more coagulant but this raises turbidity and increases the amount of residual iron causing worse feed water quality and higher operational expenses later in the treatment process as conventional treatment steps are not adaptable to sudden changes in raw water quality.

During recent two decades low pressure membrane (LPM) techniques, i.e. micro- and ultrafiltration, have emerged as an alternative for conventional water treatment. Compared to conventional treatment, comprised of coagulation, sedimentation, and sand filtration, membranes offer certain advantages. They have a highly automated operation and smaller footprint, and membranes are also robust, reliable, and effective. The combination of LPM with coagulation and other pretreatments has the potential of reducing many shortcomings of the conventional treatment. Membranes are very effective in removing particles from water, which is shown as excellent treated water quality regarding turbidity. With optimised coagulation conditions high NOM removals, similar to conventional treatment, can be reached. Simultaneously membranes offer a good microbiological barrier. To further enhance membrane performance other pretreatments, such as activated carbon and ozonisation, can be used.

The main challenge for membranes is fouling. The accumulation of small particles on and inside the membrane surface causes flux decline and thus reduces the amount of treated water, or raises operational expenses as membranes have to be cleaned. Membrane fouling is dependable on feed water quality, membrane characteristics, and a variety of other factors. A lot of information is available on the effect of membrane characteristics but very little information is available about the effect of Nordic type raw water on fouling. Feed water quality, especially parameters like turbidity, NOM, and UV absorbance, can be used to estimate the effect of raw water on fouling. However, it is very difficult to predict the fouling pattern and thus a study was conducted in order to find if membranes are an economical option for treating the raw water from Pääjärvi.

## **1.2 Scope of thesis**

This thesis was done as a part of ADWATECH project that focuses on issues related to the increase of NOM in Nordic waters. The task was to investigate the possibility to retrofit LPMs into the existing Pitkärkoski water treatment plant (WTP). Targets were to first analyse which treatment steps could be replaced by membranes and secondly suggest a solution that would be economically most interesting.

Membranes are examined as one possibility to increase the treatment capacity of Pitkärkoski WTP. Current conventional treatment is capable to a certain point to produce excellent quality drinking water, but as consumption increases, somewhere down the line physical limits of the conventional treatment, such as sedimentation tank size, will limit the treatment efficiency. Land area for the WTP is very limited and building new treatment lines is not possible. LPMs have a significantly smaller footprint and thus they might be a viable option for conventional treatment.

This thesis consists of a literature part on retrofitting LPMs to an existing WTP and an experimental part where two different feed waters for membrane filtration were tested. For the literature part of this master thesis, the existing literature on membrane technologies, membrane fouling, and retrofits were studied. Due to rapid development of membrane technology, very few books about membrane science are up to date, and thus focus has been on scientific articles. Additionally, two surveys were conducted in order to get a better understanding on the costs and advantages of retrofitting. One survey was sent to WTPs in the Northern Europe that have decided to retrofit LPMs and the goal was to understand reasons behind retrofitting and the costs related to

retrofitting. Second survey was sent to membrane manufacturers; the goal was to evaluate different membrane manufacturers based on existing WTPs employing membranes and the applicability of their membrane for this project. Supplemental information on existing retrofit projects in Nordic countries was kindly provided by researchers in Nordic countries.

The experimental part was conducted at Pitkääkoski WTP. The findings from literature study were used to estimate and select which treatment step membranes could replace and the operational parameters that should be used. To estimate the performance of the membrane, changes in permeability and water quality were followed.

### **1.3 Thesis outline**

This study focuses on low pressure membrane technologies (i.e. micro- and ultrafiltration). The applicability of nanofiltration for water treatment in Helsinki Region has been studied earlier at Aalto University. The experimental part of this thesis was done at Pitkääkoski WTP and using the existing treatment process brought some limitations. Ferric sulphate is used as coagulant at the WTP and thus, even though in theoretical part aluminium-based coagulants are discussed, in the experimental part iron-based chemical PIX-322 was used exclusively.

During the experimental part two types of treated water were used as feed water, namely clarified water after sedimentation and flocculated water taken from flocculation tank. Because of limitations with time, only these two feed waters are tested. Clarified water after sedimentation was hypothesised to be the most optimal feed water and thus a fixed conditions and optimisation periods were done. With flocculated water, only a fixed conditions test was performed.

This thesis consists of seven chapters. In chapter 2 low pressure membrane technologies and process fundamentals, such as membrane materials and separation potential of different membranes, are introduced. Chapter 3 is dedicated to membrane fouling. A basic understanding on process of fouling as well as causes for fouling is given; also methods to reduce to effect of fouling are presented. In chapter 4, information gathered about retrofitting is discussed. Reasons for retrofitting are reflected and different ways to retrofit are examined. Additionally costs and changes in water balance related to retrofitting are also discussed. Chapter 5 presents different pretreatments that can be used to alleviate membrane fouling and simultaneously increase the removal

efficiency of membranes. In chapter 6 the premise for experimental part is presented. The pilot equipment is introduced. Also testing arrangements and performance analyses are presented. Chapter 7 reveals the results regarding both membrane performance and water quality from the piloting phase. Chapter 8 summarises the thesis, conclusions from piloting are drawn and areas for future research are discussed.



## **2 Low pressure membrane technologies**

In this chapter an overview of low pressure membrane technologies is given. First low pressure membranes are introduced and filtration process is presented. This includes membrane configuration, flow regime, and flow direction. Then process fundamentals, such as membrane materials and separation potential, are discussed.

### **2.1 Membrane filtration process**

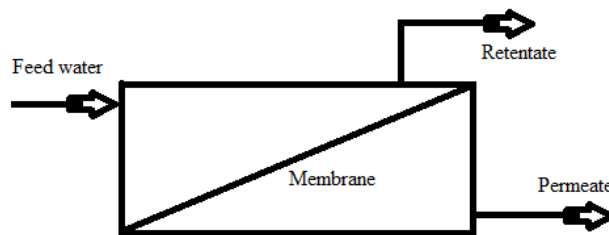
#### **2.1.1 General**

Industrial users have applied membranes for water treatment ever since the 1950s. Food processing industries have used filtration for clarifying, concentrating, and purifying fruit juices, dairy products, and beverages. In 1980s water treatment sector became increasingly concerned about microbiological contamination and it became interested about membrane filtration. In USA small utilities began to consider between granular filters and membrane filtration (Crittenden et al., 2005). In Europe membrane filtration pioneers were located in France where in the late 1980s Aquasource developed membranes for groundwater treatment and virus removal (Vickers, 2005).

In 1993 *Cryptosporidium* oocysts passed through the conventional treatment in Milwaukee, USA and caused more than 400 000 illnesses and 50 deaths (Crittenden et al., 2005). This marked a watershed year for membrane filtration as the weaknesses of conventional treatment were revealed. Conventional treatment may not able to produce safe potable water when rapid changes happen in feed water conditions and the treatment buffers are inadequate. The quality of water produced by membranes is constant regarding particles and the quality is independent on operating conditions and the raw water quality (Lerch et al. 2005a; Xia et al., 2008a; Ho et al., 2012). At the same time membranes have smaller footprint and lower energy consumption than most conventional treatments, and they have compact modules (Zularisam et al., 2006). The stricter water quality regulations are also affecting the popularity of membrane filtration as more and more water treatment plants choose to retrofit membranes in order to reach water regulations.

Membrane filtration occurs when water is forced through porous material. A variety of forces can be used such as concentration, temperature, and electric potential. Most common in water treatment is pressure (Meyn, 2011). Water passing through the

membrane is called permeate and water remaining on the feed side is called retentate, as shown in Figure 1. Membranes are semipermeable meaning that they are highly permeable to some components of feed stream whilst blocking other. In water treatment this means that the retentate side has high concentration of impurities, though not all impurities leave with retentate. Some impurities accumulate on the membrane surface and start to affect the flux through membrane. One way to maintain constant flux is to increase transmembrane pressure (TMP) but as some membranes are incapable to withstand very high TMP, cleaning the membrane by backwashing might be necessary.



**Figure 1. Schematic picture of membrane separation process. Impurities are removed by the membrane and concentrated as retentate, treated water is separated by the membrane to permeate.**

Four different pressure driven membrane systems are currently used in water treatment and they can be classified into two physiochemical processes. Low pressure membranes (LPM) are microfiltration (MF) and ultrafiltration (UF), whilst reverse osmosis (RO) and nanofiltration (NF) processes require a higher operating pressure. There are clear differences between these two processes. Differences can be found in materials used for the membranes, the configuration of membrane elements, operating modes, and flow regimes. Table 1 shows a comparison between LPMs and high pressure membranes.

**Table 1. Differences between low pressure and high pressure processes (Crittenden et al., 2005).**

Process characteristic	Low pressure membrane	High pressure membrane
Membrane types	Microfiltration Ultrafiltration	Nanofiltration Reverse osmosis
Objectives	Particle removal Microorganism removal	Salt removal (mono- and divalent ions), disinfection-by-product control
Typical source water	Fresh surface water	Seawater, brackish or coloured groundwater
Most common configuration	Hollow fibre	Spiral wound
Most common flow pattern	Dead-end	Crossflow
Typical TMP	0.2...1 bar	5...55 bar
Typical permeate flux	30...70 l/m <sup>2</sup> h with UF 130...170 l/m <sup>2</sup> h with MF	1...50 l/m <sup>2</sup> h
Typical recovery	> 95 %	40 % for seawater, 90 % for groundwater
Competing process	Granular filtration	Carbon adsorption, ion exchange, distillation

### **2.1.2 Membrane configuration**

Membranes used for water treatment are usually packed into modules. One module can hold thousands of membranes, thus 1 m<sup>2</sup> of floor space may contain 100 m<sup>2</sup> of membrane area. Most common configuration for LPMs is hollow fibre where membranes are cast as hollow tubes. The packing density is usually 1200...1700 m<sup>2</sup>/m<sup>3</sup> (Crittenden et al., 2005). The hollow fibre is particularly well suited for use as a separation media because it has a high surface to volume ratio and it exhibits bi-directional strength (Vickers, 2005). Other configurations are flat sheet, tubular, and spiral wound. Flat sheet membranes are widely used for membrane bioreactors in wastewater treatment. For potable water treatment flat sheet membranes require too much space. Spiral wound is used in RO, whereas tubular membranes are commonly used in small WTPs with high turbidity waters. Modifications of these four exist, like seven-bore membranes, but the working principles are the same as with previous.

LMPs have two configurations: pressure or suction configuration. In pressure driven configuration TMP, usually 0.4...1 bar, is created by a pump delivering water to a common manifold where the water is supplied to each module. Since each module must be piped individually, a substantial amount of piping has to be done on both feed and permeate side (Crittenden et al., 2005).

In suction configuration the feed side requires less piping. Modules operated by suction are suspended in basins containing feed water and are usually called immersed systems. A single pump on the permeate side creates TMP, usually 0.2...0.4 bar, by suction and permeate is sucked out of the basin while retentate remains in the tank. As water is constantly sucked out of the basin, concentration of substances in the basin increases. To remove the solids either feed-and-bleed or semi-batch strategy needs to be applied. In feed-and-bleed strategy a small, constant waste stream is taken out of the basin. In semi-batch strategy there is no constant waste stream out of the basin and a backwash is required. During backwash the volume of water in the basin increases and the excess water, and solids, exits through an overflow (Crittenden et al., 2005; Pikkarainen and Laine, 2011).

### **2.1.3 Flow regime and direction**

Flow regime of a membrane affects membrane flux and fouling. Two different flow regimes can be distinguished. These are crossflow regime and dead-end regime,

presented in Figure 2. In crossflow filtration the feed water runs parallel to the membrane surface. Particles are carried away by retentate whilst purified water flows through the membrane pores. Retentate is circulated back to the feed water. In crossflow mode the permeate flow is relatively low, only about 25 % of the feed water. However, crossflow stream creates shear forces that clean the membrane surface (Crittenden et al., 2005; Pikkarainen and Laine, 2011). When crossflow regime is used, TMP is calculated with the following equation:

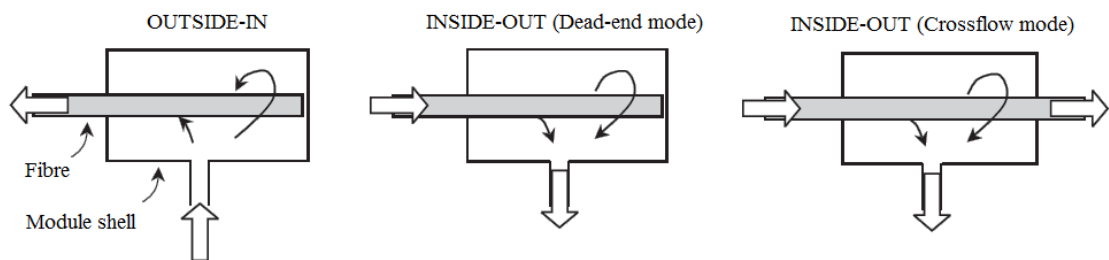
$$\Delta P = \frac{P_f + P_o}{2} - P_p \quad (1)$$

where  $\Delta P$  = transmembrane pressure [bar],  
 $P_f$  = feed pressure [bar],  
 $P_o$  = retentate pressure [bar], and  
 $P_p$  = permeate pressure [bar].

In dead-end mode feed water flow is perpendicular to and towards membrane surface. The rejected particles accumulate on the membrane surface and form a cake layer which must be removed with a backwash cycle. In dead-end mode no water exits the system without passing through the membrane. In feed-and-bleed mode, mentioned before during suction configuration, a portion of raw water exits the system without ever being in contact with the membrane which has led to some experts calling this system transverse filtration (Crittenden et al., 2005; Pikkarainen and Laine, 2011). In dead-end regime permeate is the only outlet, thus equation for TMP is simpler:

$$\Delta P = P_f - P_p \quad (2)$$

where  $\Delta P$  = transmembrane pressure [bar],  
 $P_f$  = feed pressure [bar], and  
 $P_p$  = permeate pressure [bar].



**Figure 2. Flow regimes and directions with hollow fibre membranes (Crittenden et al., 2005). With dead-end regime all water is treated whilst with crossflow some water leaves as retentate.**

Flow direction during filtration has two possibilities, also presented in Figure 2. First is flow from outside to inside, meaning that the feed water is outside the fibre and permeate inside. In inside-out system the direction is reversed: water flows from inside the membrane to outside. Outside-in configuration is used by all immersed systems and some pressure-driven systems. The advantage of outside-in systems is that they can treat more water using same flux because the outer surface of the membrane has more surface area.

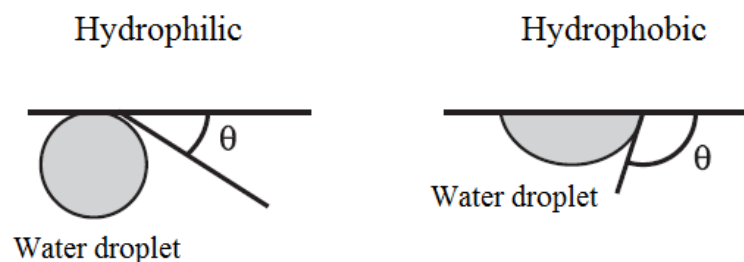
Crossflow modules can be operated at higher flux than dead-end because of shear forces cleaning the membrane surface. With this configuration, however, less water can be treated because most of the water entering the module goes to recirculation. This also raises operational expenses.

## 2.2 Process fundamentals

When retrofitting a WTP with membrane filtration is considered, understanding the fundamentals of membrane filtration must be known. Previously membrane configuration and flow regimes were presented but just as important as choosing the right configuration is choosing the correct membrane material and type. These will be discussed next.

### 2.2.1 Membrane materials

The physical and chemical properties of membrane material have a strong influence on membrane performance. Membrane porosity affects the head loss through membrane, higher porosity leads to lower head loss. Membrane durability and material cost affect operational costs of a WTP. Tolerance to different chemicals affects the ability to clean and disinfect the membrane. But one of the most important membrane characteristic is hydrophobicity which is measured as the contact angle between a water droplet and a membrane surface. Higher contact angle means more hydrophobic membrane (Figure 3).



**Figure 3. Measuring the hydrophobicity with a water droplet (Crittenden et al., 2005). Contact angle, indicated with  $\theta$ , is higher for hydrophobic membrane.**

In general hydrophilic materials have been found to have lower fouling tendencies (Nakatsuka et al., 1996; Crittenden et al., 2005; Zularisam et al., 2006). Hydrophobicity is, however, not directly related to membrane productivity. Pezeshk and Narbaitz (2012) compared a modified membrane and a commercial membrane and showed that with a modified membrane a slightly higher flux, higher cumulative permeate production, and lower flux reduction could be achieved despite it being more hydrophobic.

A variety of materials can be used for LPM filtration. Most common materials used for LPMs are polymeric membranes such as cellulose acetate (CA) and different synthetic polymers (Jacangelo and Noack, 2005). One inorganic application that has been used in LPM filtration is ceramic membranes with an aluminium, carbon, titanium, or zirconium structure (Hofs et al., 2011). Metal membranes have been used in pilot-scale conditions (Lieknes et al., 2004).

Polymeric membranes are the most common material for all membrane systems. Membranes are made from CA or synthetic polymers such as polyvinylidene fluoride (PVDF), polysulfone (PS), polyethersulfone (PES), and polypropylene (PP). Characteristics of these membrane materials are presented in Table 2. Number of WTPs opting for synthetic polymers is increasing due to better chemical and acidic durability of the membrane, yet CA membranes are said to be more fouling resistant due to their hydrophilic nature (Crittenden et al., 2005).

**Table 2. Characteristics of organic membranes (Crittenden et al., 2005). Synthetic membranes withstand worse chemical conditions but have a higher fouling tendency.**

Membrane material	Hydrophobicity	Oxidant tolerance	Operating pH range	Fouling resistance
Polyvinylidene fluoride (PVDF)	Slight hydrophobic	Very high	2...11	Good
Polypropylene (PP)	Hydrophobic	Low	2...13	Acceptable
Polyethersulfone (PES)	Slight hydrophobic	High	2...13	Good
Polysulfone (PS)	Slight hydrophobic	Moderate	2...13	Good
Cellulose acetate (CA)	Hydrophilic	Moderate	5...8	Very good

The smaller fouling tendency of CA membranes can be questioned as one WTP that answered the survey conducted for this study reported that they had changed their membrane material from CA to synthetic polymer due to extensive fouling problem. CA membranes are incapable to withstand high doses of chlorine and highly acidic or alkaline chemicals are unusable due to very narrow operating pH. CA membranes are also more susceptible to biological degradation (Crittenden et al., 2005). Thus, it is possible that organic substances accumulating on the membrane surface caused biological degradation because proper cleaning could not be performed.

Ceramic membranes made from  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$ ,  $\text{ZrO}_2$ , or a carbon composite like  $\text{SiC}$ , are thicker than organic membranes. Ceramic membranes have a very high tolerance towards chemicals, something which can relate to lower operational costs (Jacangelo and Noack, 2005). Downsides are that they require higher TMP and that the cost of the membrane is higher than the cost of polymeric membrane (Heijman and Bakker, 2007). Ceramic membranes are said to have better operating properties than polymeric membranes, but scientific evidence to back up these claims is hard to find. Research done with ceramic membranes is usually conducted for or with companies producing said membranes.

Operation of ceramic membranes is similar to polymeric membranes and they perform well under conditions optimized for polymeric membranes (Lerch et al., 2005b; Meyn, 2011). Hofs et al. (2011) compared ceramic and polymeric membranes on their permeability and fouling potential and found that ceramic membranes were subjected to less fouling than polymeric membranes. Researchers credited this to larger pore size of ceramic membranes. Simultaneously they, however, reported a better removal of NOM and turbidity for ceramic membranes meaning that the overall performance of ceramic membranes was better. This is backed up by studies by Heijman and Bakker (2007) who found that ceramic MF membranes operate better under extremely fouling conditions of Twente Canal water. Benefits that Heijman and Bakker presented for ceramic membranes were higher mechanical strength, operational pressure of up to 20 bar, higher backwash pressure which results in more efficient backwash, longer backwash interval and higher resistance to chemicals, higher clean water permeability of more than  $180 \text{ l/m}^2\text{h}$ , longer lifetime of membranes, and recyclability of membrane material.

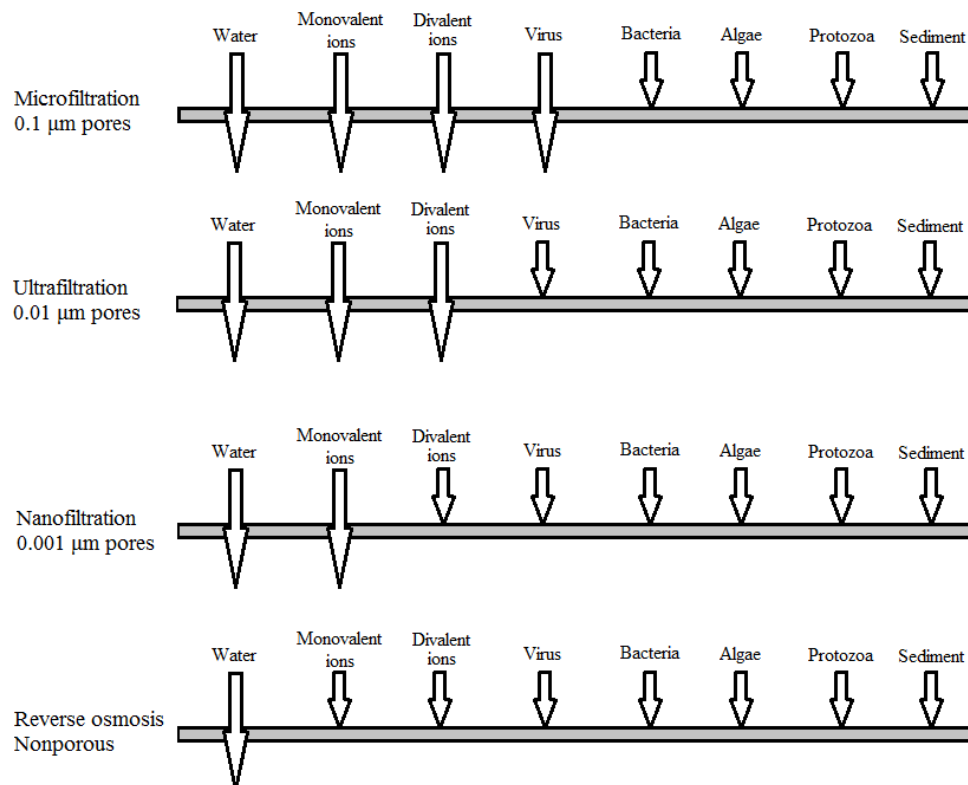
Leiknes et al. (2004) conducted a pilot-scale study on utilising metal MF membranes for NOM removal from artificial surface water. Operation of these metal membranes was similar to immersed polymeric membranes. Consistently high quality permeate was produced irrespective of the operational conditions. Leiknes et al. estimated that a flux of approximately  $200 \text{ l/m}^2\text{h}$  could be achieved with regular backwashes for duration of 2.5 months before membranes had to be taken out of production and extensively cleaned. Metal membranes, much like ceramic membranes, are however much more expensive than polymeric ones.

Environmental factors, however, favour the usage of metal or ceramic membranes. According to Liikanen et al. (2006) polymeric membranes go to landfills after they have

reached the end of their operation cycle. Ceramic membranes can be recycled (Heijman and Bakker, 2007). Same can be true for metallic membranes, depending on the metals used. Information about the energy usage of production is scarce and thus it is difficult to conclude anything about the environmental aspects of membrane manufacturing.

### 2.2.2 Separation potential

When it comes to particle separation membrane processes have a hierarchy, presented in Figure 4. Still, there exists some conflict between membrane types as no agreement or standards exist on how microfiltration (MF) and ultrafiltration (UF) membranes should be rated and the classification is often done as a part of manufacturers marketing strategy. Pore sizes might be stated as nominal or absolute pore size, but MF membranes are usually considered to have a pore size of  $0.1\text{--}0.2\text{ }\mu\text{m}$ , though MF membranes with pore size of  $10\text{ }\mu\text{m}$  exist. UF membranes have pore size between  $0.01\text{--}0.05\text{ }\mu\text{m}$  (US EPA, 2005). Due to historical reasons, however, UF membranes are usually rated according to molecular weight cut-off (MWCO); substances with a larger molecular weight than MWCO of the membrane are retained (Crittenden et al., 2005).



**Figure 4. Separation potential of different membranes (Modified from Crittenden et al., 2005). LPMs remove particles and microbial substances but removal of ions needs a tighter membrane.**

LPMs use physical sieving to reject substances and are widely used as a pretreatment in water treatment (Zularisam et al., 2010). LPMs are usually very successful in removing



turbidity and in many WTPs this is their primary function. Despite variations in raw water quality, permeate typically has values less than 0.1 NTU. With particles, the removal rate is usually between 2...5 log for both MF and UF (Speth and Reiss, 2005).

As can be seen from Figure 4, MF is effective against bacteria and protozoans such as *Giardia* and *Cryptosporidium*, but viruses are not completely separated. UF can be used to remove most viruses and the efficiency is enhanced by fouling (El-Hadidy, 2011). The removal rate of both LPMs with larger pathogens is usually around 6 logs, with viruses UF can reach a removal rate of 3 logs. Persson et al. (2005) have reported on a removal rate of 4.7...5.5 logs on bacteriophage with submerged UF. However, despite excellent removal efficiency of LPMs, WTPs require disinfection step in their water treatment process (Zularisam et al., 2006; Ho et al., 2012). Ho et al. showed that even after nanofiltration bacteria cells were present although they were 100...10 000 times bigger than the MWCO of the membrane. They did conclude that some contamination had happened in the outlet pipe after the membrane but they were unable to completely rule out bacteria passing through the membrane.

The rejection of NOM depends highly on the membrane characteristics and NOM aromaticity and hydrophobicity (Cho et al., 2000; Kennedy et al., 2005). MF membranes are generally too loose to reject NOM whereas some UF membranes can reject NOM. Cho et al. found that a negatively charged UF membrane treating surface water was effective in removing NOM, rejection rates as high as 80 % could be achieved. However, a non-charged hydrophilic CA UF membrane with a significantly smaller pore size was capable to reject only 50 % of NOM. Kennedy et al. found no dissolved organic carbon (DOC) removal when treating surface water with a hydrophilic PES UF membrane. Cho et al. suggested that the rejection of NOM depends on aromaticity of the NOM, and therefore the shape of NOM particle, instead of MWCO of the membrane. They also indicated that hydrophobic NOM was effectively removed, whilst transphilic and hydrophilic NOM either penetrated the membrane or stayed on the membrane surface.

As commercial membranes are designed to remove large particles, the removal of viruses, NOM and ions is low. Combining LPMs with pre-treatment steps like coagulation and adsorbents enhances the removal rate. Keucken et al. (2012) reported on a 46 % removal of NOM when in-line coagulation was applied, while Best et al. (1999) said that by using powdered activated carbon (PAC) a removal of 58 % could be

achieved. Heijman and Bakker (2007) reported of an increase of 300...400 % in virus removal rate with ceramic MF membrane when coagulation was used as pretreatment. Pretreatment options that existing WTPs have adopted will be further discussed in chapter 5.

### 2.2.3 Membrane performance

The performance of LPMs is usually presented as flux through the membrane. Typical unit for flux is litres of water per membrane area per hour (l/m<sup>2</sup>h). Americans usually measure flux in gallons per square foot per day (GFD). If GFD is to be presented in l/m<sup>2</sup>h, GFD is multiplied by a factor of 1.7. Pure water flux depends on TMP and viscosity of water at operating temperature. To calculate pure water flux a modified Darcy's equation can be used (Chellam and Zander, 2005):

$$J_M = \frac{Q}{A} = \frac{\Delta P}{\mu_M R_m} \quad (3)$$

where  $J_M$  = flux at measured temperature [l/m<sup>2</sup>h],  
 $Q$  = flow rate of water [l/h],  
 $A$  = area of membrane surface on the feed side [m<sup>2</sup>],  
 $\Delta P$  = transmembrane pressure [bar],  
 $\mu_M$  = dynamic viscosity of water at measured temperature [Pa·s], and  
 $R_m$  = membrane resistance coefficient [m<sup>-1</sup>].

Difference with equation 3 to Darcy's equation is the employment of TMP instead of pressure gradient. The membrane resistance coefficient is determined by measuring clean water flux and it is assumed to remain constant during filtration (Kim and DiGiano, 2009). As can be seen from equation 3, flux depends on viscosity of water which changes with varying temperatures. At lower temperature the viscosity is higher. Viscosity of water in the temperature range of 0 °C...35 °C can be accurately calculated with the following equation (Chellam and Zander, 2005):

$$\mu = \frac{1.777 - 0.052T + 6.25 \times \frac{T^2}{10^4}}{1000} \quad (4)$$

where  $\mu$  = dynamic viscosity of water [Pa·s] and  
 $T$  = temperature of water [°C].

In the northern hemisphere water has highest viscosity during winter, thus causing the lowest membrane flux. If the production of a WTP varies little over seasons, the

membrane system has to be designed to operate with coldest water temperatures. To be able to compare water flux at different temperatures standard temperature of 20 °C is used. Viscosity of water at 20 °C is approximately 0.001 Pa·s. To convert flux at measured temperature to flux at standard temperature the following equation is be used:

$$J_S = J_M \left( \frac{\mu_M}{\mu_S} \right) \quad (5)$$

where  $J_S$  = flux at standard temperature [l/m<sup>2</sup>h],  
 $J_M$  = flux at measured temperature [l/m<sup>2</sup>h],  
 $\mu_M$  = dynamic viscosity of water at measured temperature [Pa·s], and  
 $\mu_S$  = dynamic viscosity of water at standard temperature [Pa·s].

The most common way to present membrane performance is membrane permeability, or specific flux, calculated by dividing temperature normalized flux with TMP. The equation for specific flux is (Kim and DiGiano, 2009):

$$J_k = \frac{J_S}{\Delta P} = \frac{Q}{A \Delta P} \times \frac{\mu_M}{\mu_S} \quad (6)$$

where  $J_k$  = specific flux [l/m<sup>2</sup>·h·bar],  
 $J_S$  = flux at standard temperature [l/m<sup>2</sup>h],  
 $\Delta P$  = transmembrane pressure [bar],  
 $Q$  = flow rate of water [l/h],  
 $A$  = area of membrane surface on the feed side [m<sup>2</sup>],  
 $\mu_M$  = dynamic viscosity of water at measured temperature [Pa·s], and  
 $\mu_S$  = dynamic viscosity of water at standard temperature [Pa·s].

Specific flux, measured as l/m<sup>2</sup>·h·bar, normalises the response, namely permeate flow, to the stimulus, namely TMP and allows comparison of various membranes. As manufacturers have adopted different designs in their LPM systems, comparing membranes should be done carefully. For example it is important that the membrane surface area in contact with the feed water is used, as there is a notable difference in surface area between different flow directions (Chellam and Zander, 2005).

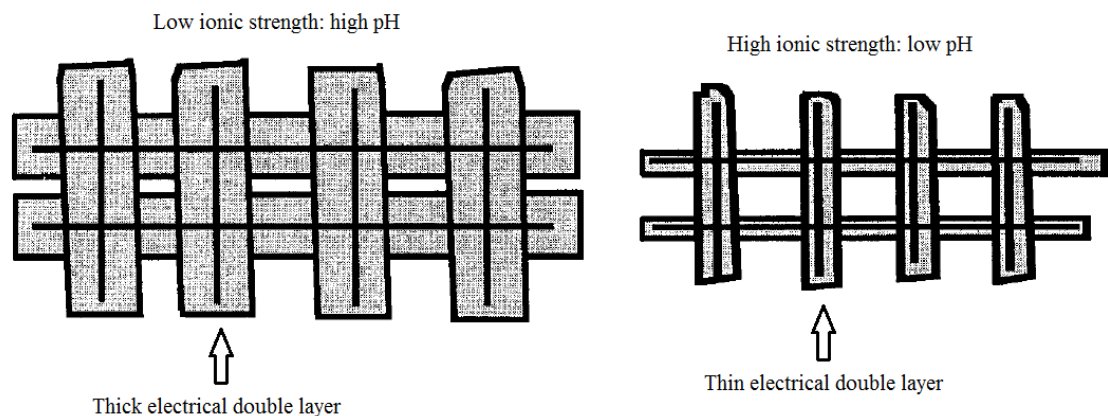
#### 2.2.4 Water quality

As mentioned before, permeate quality of LPMs is constant despite the quality of raw water. However, the productivity of a WTP is dependent on these parameters and they influence membrane flux (Xia et al., 2008a). Water quality, especially turbidity,

temperature, algae content, and taste and odour vary significantly throughout the year. These play a significant role when planning to use membrane systems for water treatment.

During summer the water demand is highest, but since viscosity of the water is lower at higher temperatures, the membrane flux is also highest. According to Chellam and Zander (2005), a 5 °C change, from 15 °C to 20 °C will cause an 11.7 % increase in membrane flux. Persson et al. (2005) say that the decrease in productivity is 2 % with each declining degree. This drop, if incorrectly interpreted, might be credited to fouling.

Braghetta et al. (1997) found that at higher pH the flow of organic-free water through a NF membrane was higher. This increase was credited to electrolyte-swollen nature of the membrane matrix: at high pH the charged functional groups of the membrane matrix force membrane polymers apart, thus increasing the membrane pore size (Figure 5).



**Figure 5. Effect of pH on membrane polymers (Braghetta et al., 1997). A higher feed water pH results in thick electrical layer and a tighter membrane, and thus a better substance rejection.**

Warmer water is advantageous for the membrane flux but it facilitates algae growth which might cause problems for membrane systems. If the retention time of raw water source is short, seasonal events might also an effect raw water quality. During autumn months the organic content of surface water increases due to the decay of fallen leaves. This will cause an increase in both turbidity and TOC. Another natural event happening during autumn is turnover of lakes which might create iron and manganese problems. Another turnover might happen during springtime, resulting again in deterioration of raw water quality (US EPA, 2005). As mentioned before, the changes will not affect the permeate quality, however all these characteristics cause membrane fouling.

### 3 Membrane fouling

In this chapter membrane fouling is discussed. Although significant efforts have been made to control membrane fouling, it is still the main drawback of membrane systems. First the mechanisms related to fouling are presented, then the effect of different components of raw water on fouling are discussed. Lastly both physical and chemical methods to eradicate foulants are discussed.

#### 3.1 Fouling mechanisms

When water is filtrated through a low pressure membrane (LPM), the membrane is prone to losing permeability due to accumulation of impurities on or inside the membrane. Fouling causes high operational and maintenance costs, lowers productivity, and increases the membrane regeneration. It is usually defined as either decline in permeate flux or increase in transmembrane pressure (TMP). Positive note is that the deposit of foulants on membrane can act as a rejecting layer and might improve permeate quality regarding some characteristics (Nakatsuka et al. 1996; Zularisam et al., 2010). Although fouling has been intensively researched during the past three decades, it still remains as one of the biggest drawbacks of membrane systems.

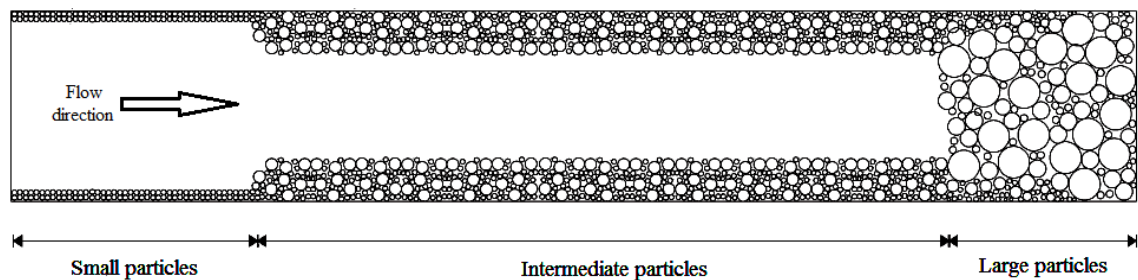
The principle fouling types are pore blocking and adsorption, cake layer formation, and combinations of these which are caused by organic substances. Mechanisms of the principle fouling types are listed in Table 3. Concentration polarization, accumulation of rejected particles near the membrane surface, increases the effect of these mechanisms. At steady-state situation the polarization of particles to and from the membrane surface are at equilibrium (Chellam and Zander, 2005). However, concentration polarization is not as severe with LPMs as with RO and NF membranes (Crittenden et al., 2005).

**Table 3. Typical fouling mechanisms (Baker and Dudley, 1998; Gao et al., 2011). MF membranes are more subjected to pore blocking and adsorption whereas UF membranes are most affected by cake layer formation.**

<b>Fouling type</b>	<b>Type of foulant</b>	<b>Mechanism</b>
Pore blocking	Large particles	Large particles accumulate on the surface blocking the membrane pores
Inner pore adsorption	Small particles	Small particles are adsorbed into the membrane pores where they create a blockage
Cake layer	Particles	More and more particles precipitate on the initial blockage caused by large or small particles.
Biofouling	Aquatic organisms	EPS formation, deposition and growth of cells on the membrane.
Organic fouling	NOM	Partially unknown

Membrane characteristics and operational mode affect fouling rate of membrane. For microfiltration membranes pore blockage and inner pore adsorption have been found to be responsible for fouling whilst with ultrafiltration cake layer formation was found to cause most fouling (Zularisam et al., 2006). Membranes with larger pores have shown a greater flux decline compared to smaller ones. One suggested reason for this is internal fouling (Zularisam et al., 2010). Pikkarainen and Laine (2011) claim that fouling of suction system membrane is less severe than pressurised system due to lower flux applied.

Lerch et al. (2005a) studied fouling of a membrane with inside-out flow direction and dead-end configuration, and found out that large particles are transported to the end of the capillary where they cause pore blocking whereas the amount of inner pore adsorption caused by small particles was uniform throughout the membrane capillary. An illustration of the fouling is in Figure 6.



**Figure 6. Coating layer build-up in dead-end inside-out filtration, where large particles accumulate at the end on membrane fibre but small particles foul the membrane evenly (Lerch et al., 2005a).**

Fouling is usually divided into two categories, reversible and irreversible fouling. During the initial operating period a decline in membrane flux occurs but part of the flux can be recovered with physical cleaning methods. This part is known as reversible fouling. The fouling that is not recovered with physical methods is called physically irreversible fouling and is usually caused by inner pore blocking. Part of the physically irreversible fouling can be recovered by adding chemicals to the backwash sequence. If this is ineffective, a longer chemical treatment is necessary (Crittenden et al., 2005; Yamamura et al., 2007).

### 3.2 Causes for fouling

Different fractions of feed water have been found to cause fouling of the membrane. In this chapter the mechanisms and severity of fouling caused by different foulants is looked at in more detail.

### **3.2.1 Fouling by colloids and inorganic compounds**

Surface waters contain a wide range of colloids such as silt and clay, precipitated iron and aluminium from incomplete treatment, and corrosion products. Colloids cause fouling mainly by pore blocking and inner pore adsorption. Additionally, particles deposited on the initial foulants can form a cake layer with a microstructure of numerous irregular pores. In high pressures this layer is subjected to cake compression, which will further increase the decline in flux (Guo et al., 2012).

Inorganic compounds cause fouling by precipitation on the membrane surface due to hydrolysis or oxidation during filtration. LPMs are incapable of removing manganese and calcium from water (Yamamura et al., 2007; Panglisch et al., 2008). They are, however, effective in removing iron and aluminium. Panglisch et al. (2008) found that iron is usually present in particular or colloidal forms which are retained by the membrane. However, iron colloids smaller than 0.45  $\mu\text{m}$  were found to strongly influence the membrane permeability. Peiris et al. (2010) found colloidal and particular components to cause reversible fouling when LPMs are treating natural waters. However, according to Peiris et al. only 10...15 % of the principle components of natural water were found to be colloidal or particular. Similar percentages were measured from Pitkääkoski raw water.

### **3.2.2 Biofouling**

Biofouling is caused by microorganisms in the water. Formation of biofouling is a two-step process. First the microorganisms attach irreversibly to the membrane surface. Once a microorganism has attached its cells to the membrane it starts to use nutrients in the feed water and to grow and reproduce, i.e. a biofilm starts to form. Microorganisms use carbon as one of their main nutrient. This usable carbon is present as assimilable organic carbon (AOC). The amount of AOC can therefore be used as a measurement of the biofilm formation potential (Chellam and Zander, 2005). Vrouwenvelder et al. (1998) found an amount of 27  $\mu\text{g}$  AOC/l in surface water to cause biofouling on a NF membrane.

Drews et al. (2006) studied biofilm formation closer and concluded that microorganisms such as algae can exude extracellular polysaccharides (EPS) which forms a gel-like biofilm on the membrane. Fluctuating levels of dissolved oxygen and nitrate have a profound

effect on EPS concentration. Unsteady operation has also been found to be responsible for EPS formation.

Biofilm inside the feed channels and spacers causes friction loss. Biofilm also causes channelling where hollow fibre bundles become bound together by foulant. Channelling is very difficult to clean since cleaning solution more often fails to reach the membrane leaves (Baker and Dudley, 1998).

Biofilm formation is very problematic when wastewater is treated with membranes, but it can cause problems in potable water production as well. Even if the amount of microorganisms is regularly very low, a spike in microorganism quantity may cause the formation of biofilm. When the biofilm has been formed, it can thrive under very scarce nutrient conditions due to concentration polarization of AOC to close proximity of the membrane where AOC is consumed by microorganisms (Chellam and Zander, 2005).

### **3.2.3 Fouling by natural organic matter**

Surface waters contain natural organic matter (NOM), which is a heterogeneous mixture of organic compounds with wide range of molecular weight and functional groups (Zularisam et al., 2006). The main source of NOM is terrestrial and vegetative debris. As previously mentioned, LPMs are not designed to remove NOM with the exception of UF membranes with smallest MWCO. However, NOM causes severe fouling of the membrane. The mechanism of how NOM causes fouling is at least partially unknown, but Guo et al. (2012) presented three fouling mechanisms due to NOM. First is adsorption of NOM into the membrane pores, second is forming a separate gel layer on the membrane surface and third mechanism is forming a low permeability layer on the membrane together with particles. Li et al. (2012) found that the presence of calcium increases NOM fouling on the membrane surface due to calcium bridging.

Specific UV absorbance (SUVA) is the easiest method to study the rejection of NOM from surface water (Cho et al., 2000). It is calculated by dividing UV absorbance with DOC. SUVA can be used to study the fouling potential of raw water as it correlates with hydrophobicity and aromatic characteristics of the raw water. SUVA-values above four refer to high hydrophobicity and aromatic characteristics (Edzwald and Tobiasson, 1999; Cho et al., 2000; Zularisam et al., 2006). High SUVA-value for permeate confirms that most of the rejected compounds are non-humic substances (Mozia and Tomaszewska, 2004;



Zularisam et al., 2006). Waters with high SUVA-value have been found to cause more severe fouling (Pezeshk and Narbaitz, 2012).

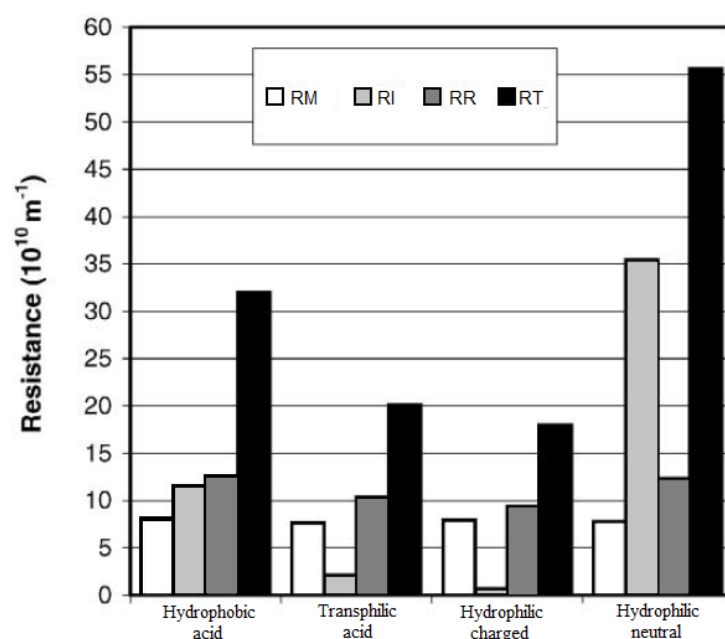
Zularisam et al. (2006) have reviewed studies related to NOM and conclude that NOM can be divided into three segments based on their hydrophilicity: hydrophilic, transphilic, and hydrophobic fractions. Cho et al. (2000) claim that hydrophobic fraction has a higher molar mass than transphilic and hydrophilic fractions. Gray et al. (2007) state that hydrophobic compounds, such as humic substances, foul the membrane by blocking the pores, whereas the hydrophilic compounds, such as carbohydrates and proteins, form a cake layer on the membrane surface.

Fan et al. (2001) have studied the influence of different NOM components on the fouling potential of a hydrophobic PVDF microfiltration membrane. They actually divided NOM into four fractions: hydrophilic acids, transphilic acids, hydrophilic charged compounds, and hydrophilic neutral compounds. Each fraction caused fouling but hydrophilic neutral compounds had the greatest fouling potential. Second was hydrophobic acids, then transphilic acids, and hydrophilic charged compounds were found to be the fraction causing least fouling. Similar results were presented by Carroll et al. (2000) and Kennedy et al. (2005) who conducted studies with PES/PVP membrane. Kennedy et al. hypothesised that during the separation of different fractions colloidal matter remained in the hydrophilic fraction and caused significant fouling on its own or when NOM was bind with  $\text{Ca}^{2+}$ . However, Carroll et al. added calcium and sodium to the other fractions which neglected the effect of said components.

Gray et al. (2007) also found hydrophilic neutral fraction to have the highest fouling potential. They, however, showed that the second most fouling fraction depends on the membrane material. With hydrophobic PP MF membrane and PES UF membrane, hydrophilic charged fraction was found to be second most fouling fraction and hydrophobic acids the least fouling fraction. With hydrophobic PVDF MF membrane hydrophobic charged had the lowest fouling potential, similar to the results presented by Fan et al (2001). With hydrophilic PVDF MF membrane no difference on fouling potential between hydrophilic charged and hydrophobic fractions was found.

Fan et al. (2001) and Kennedy et al. (2005) also studied the resistance of filtration of these NOM fractions. Hydrophilic fraction was found to be the main cause for irreversible fouling resistance. Fan et al. suggested the order of difficulty of removal by

backwashing to be hydrophilic neutral > hydrophobic > transphilic > hydrophilic charged. Kennedy et al. had a slightly different order: hydrophilic > transphilic > hydrophobic. Resistance observed by Fan et al. is also presented in Figure 7.



**Figure 7. Resistance of filtration of the four NOM fractions (modified from Fan et al., 2001). Hydrophilic neutral fraction has the highest irreversible and total resistance; hydrophobic acids cause highest reversible fouling. RM = membrane resistance, RI = irreversible fouling resistance, RR = reversible fouling resistance, and RT = total resistance.**

Fan et al. (2001) studied the relationship of membrane hydrophobicity and NOM. They used two PVDF membranes, one hydrophobic and one hydrophilic and found out that hydrophilic membrane performed much better, and that after 30 minutes of testing the relative flux through hydrophobic membrane was only one sixth of the flux through hydrophilic membrane. Pre-filtration was found to reduce the fouling potential of hydrophilic membrane but did not affect the hydrophobic membrane. The poor performance of hydrophobic membrane was credited to the interactions between the organic matter and the membrane. Similar argument was presented by Gray et al. (2007) on using coagulation as pretreatment, they claimed that coagulation mainly reduces hydrophobic compounds and have very little effect on hydrophilic neutral compounds.

Lin et al. (2000) and Zheng et al. (2011) claim that hydrophobicity and molecular weight should not be considered separately. In their studies, Zheng et al. found waters containing high proportion of large molecules (molecular weight > 10 kDa) with hydrophilic neutral characteristics to be the most fouling substances. Zheng et al. also looked into the fouling potential of different water sources and concluded that in waters where foulants are of microbial origin, the fouling is highest and foulants from terrestrial substances were found to be less fouling.

The feed water pH has been found to have an effect on membrane fouling by NOM but the effect, whether it is positive or negative, is unclear. Braghetta et al. (1997) found that the removal rate of negatively charged NOM at higher pH increased due to charge repulsion. Kabsch-Korbutowicz (2005a) noticed similar trends: at higher pH NOM removal increased. She used two UF membranes in her studies, one made of cellulose and the other of PES in different pH conditions varying from pH 5 to pH 10. Kabsch-Korbutowicz credited the decrease at low pH to smaller macromolecular configuration of NOM. Therefore NOM could easily penetrate through the membrane. As pH increases the macromolecules expand and the efficiency of NOM separation is increased.

Dong et al. (2006) agree that pH has an effect, but present results that are totally opposite: NOM removal increases as pH decreases. Dong et al. also hypothesise that at low pH the adsorption of NOM onto the membrane is increased. This is in conflict with what Braghetta et al. and Kabsch-Korbutowicz claimed. Ruohomäki et al. (1998) say that hydrophilic PES UF membrane experiences worst fouling at pH 6.5. The flux reduction was 21 % and at pH 3 and pH 9 the reduction was only 13 %. They also found irreversible fouling at pH 6.5 which did not occur at other pH-levels.

### 3.2.4 Effect of fouling on membrane performance

Previously equation 3 was presented for calculating water flux through membrane. This equation, however, assumed that only the temperature of water affects the flux. However, as previously has been pointed out, surface waters have substances that cause fouling of the membrane and decrease of flux. This means that instead of using just membrane resistance in equation 3, reversible and irreversible fouling resistance should be added to the equation. A common way to model LPM systems is by a resistance-in-series approach. As such, equation 3 is changed to (Chellam and Zander, 2005):

$$J_M = \frac{\Delta P}{\mu_M(R_m + R_r + R_i)} \quad (7)$$

where  $J_M$  = flux at measured temperature [ $\text{l/m}^2\text{h}$ ],

$\Delta P$  = transmembrane pressure [bar],

$\mu_M$  = dynamic viscosity of water at measured temperature [ $\text{Pa}\cdot\text{s}$ ],

$R_m$  = membrane resistance coefficient [ $\text{m}^{-1}$ ],

$R_r$  = reversible fouling resistance [ $\text{m}^{-1}$ ], and

$R_i$  = irreversible fouling resistance [ $\text{m}^{-1}$ ].

Kim and DiGiano (2009) suggest that in a resistance-in-series model the membrane resistance term is negligible compared to the fouling resistance. Xia et al. (2008a) have presented a mathematical model to predict membrane flux for UF membrane when turbidity of the feed water is known. Parameters in the model are TMP and water quality, i.e. raw water turbidity and temperature. The model is designed to be used when turbidity of the feed water is high. The model is following:

$$J(\Delta P, Y, T) = -0.252 \times \frac{\Delta P \times \ln Y}{\mu} + 10.5 \times \ln Y + 1.45 \times \frac{\Delta P}{\mu} - 17.4 \quad (8)$$

where  $J$  = flux through membrane [ $\text{l/m}^2\text{h}$ ],  
 $\Delta P$  = transmembrane pressure [bar],  
 $Y$  = raw water turbidity [FTU], and  
 $\mu$  = dynamic viscosity of water [ $\text{Pa}\cdot\text{s}$ ].

For the effect of turbidity on membrane to be visible, turbidity needs to be about 10 FTU. This model was tested on small pilot equipment and proven to be fairly accurate. The model is, however, not capable of predicting membrane flux on a long-term basis because Xia et al. have ignored irreversible fouling resistance from their equation. Membrane resistance was also ignored as suggested by Kim and DiGiano.

To gain deeper understanding into the mathematical models related to fouling of ceramic and polymeric membranes, the reader is advised to familiarise oneself with the works of Konieczny (2002) and Kim and DiGiano (2009), respectively.

### 3.3 Membrane cleaning

To control the flux decline because of fouling, membranes should be cleaned. Cleaning techniques, both physical and chemical will be presented in this chapter. The effect of different chemicals on different foulants is also presented.

#### 3.3.1 Physical methods

To control the level of reversible membrane fouling, membranes need to be backwashed. Backwashing may appear to be a simple process: however, many factors of backwashing affect the overall operation of LPM system. Mass balance of the system is affected by frequency and duration of backwash sequence whilst energy input during backwash raises operational costs.

Backwashing can be done with either liquid or air or a combination of both. Backwashing with permeate is most common with inside-out systems, whereas a combination of air scour and permeate is used with outside-in configuration. Air scour alone can be used in submerged systems, but also permeate backwashing is used to enhance the backwash efficiency. Usually the trigger that starts the backwash sequence is TMP but operational period and produced volume can be used as well. Backwashing at the operational WTPs is usually performed every 30 to 120 minutes of operation. Shorter interval is disadvantageous for filtration performance (Leiknes et al., 2004). The concentration of feed contaminants can be 10 to 20 times higher in backwash water than in feed water and this water might require further treatment before it can be discharged (US EPA, 2005).

Easiest way to conduct a backwash sequence is a physical cleaning phase of predetermined duration. During the backwash water and/or air are applied with constant pressure to clean the membrane. However, much more complex backwash sequences have been developed in order to maximize cleaning effect and minimize consumed energy, water, and air. Xia et al. (2008b) used two consecutive pulses to backwash a hollow fibre polyacrylonitrile ultrafiltration membrane fouled by reservoir water. With an optimized backwashing conducted by first backwashing with permeate for 25 seconds, cross-flow filtration for 10 seconds, and backwashing for 15 seconds, a flux enhancement of 63 % compared to normal backwashing was achieved. When fouling is caused by algae Liang et al. (2008) recommend a longer backwash sequence. In laboratory experiment they found out that with 1 to 9 minute long backwash only 20...38 % recovery was achieved. A combination of backwashing and forward flushing for 20 minutes reached 80 % efficiency.

At St. Saviours WTP submerged membranes are cleaned with three different physical backwashes. In the first process, which happens every 10 seconds, air is injected to the membrane module to remove cake layer. During this time the filtration continues normally. Second cleaning process happens every 15 minutes and then water is forced through the lumen side of the membrane. Third cleaning is similar to the previous, but hypochlorite is added to assist the cleaning process (Redhead, 2008).

Usually permeate water is used for backwashing but as some substances pass through the membrane the efficiency of permeate as backwash water can be questioned. Monovalent and divalent cations, namely sodium and calcium, respectively, were found

to weaken the backwash efficiency. NOM in permeate, however, has no effect on permeate backwashing efficiency (Li et al., 2009). Li et al. (2012) studied the removal of different NOM foulant fractions with demineralized water and concluded that whilst permeate backwashing was capable to remove only 16.5 % of humic substances demineralized water was able to remove almost 72 %. It was especially effective in removing irreversible fouling (Li et al. 2009). The reason for this is that demineralized water reduces the charge screening effect caused by cations diffused on the membrane surface and helps to maintain the original negative charge of the membrane. Demineralized water was, however, still not effective for all foulants and chemical cleaning is required (Li et al. 2012).

### 3.3.2 Chemical cleaning

Backwashing is a routine method but when foulants can no longer be removed with backwashing, chemically enhanced backwashing (CEB) is required. In a CEB chemicals are added to the backwash. The duration of cleaning sequence might also be increased to ensure enough time for chemical reactions to take place and foulants to be removed effectively. As CEB only removes chemically reversible foulants, a longer and more intensive cleaning must be performed to retain the original permeability of a membrane. The intensive cleaning is usually done *in situ* and called cleaning in place (CIP) but especially in smaller treatment plants the modules can be detached from their place and intensively cleaned separately. This method is called cleaning out of place (COP). Chemical cleanings should, however, be kept to minimum as repeated chemical cleanings affects membrane life (Kimura et al., 2005). Table 4 presents the chemicals usually used for chemical cleaning.

**Table 4. Chemical cleaning agents (Zondervan and Roffel, 2007). Different chemicals work for different foulants. Simple caustic, acidic, and oxidizing chemicals are regularly used at WTPs; more advanced chemicals are targeted towards certain foulants.**

Category	Examples of chemicals
Caustic	NaOH, KOH, NH <sub>4</sub> OH
Acidic	HCl, HNO <sub>3</sub> , H <sub>2</sub> SO <sub>4</sub> , H <sub>3</sub> PO <sub>4</sub> , Oxalic
Sequestering/complexing	EDTA
Detergent/surfactant	Alkyl sulphate, SDS, CTAB
Enzymatic	Aplha-CT, CP-T, Peroxidase
Oxidizing/disinfectant	NaClO, H <sub>2</sub> O <sub>2</sub> , KMnO <sub>4</sub>
Blend	4AquaClean, Divos, Triclean, Ultrasil/AquaClean

Kimura et al. (2005) and Gao et al. (2011) present the following relationship with chemical solution and fouling: acid solutions work best to control inorganic fouling,

caustic solutions are relatively effective against organic fouling, and biocide solutions are most effective against biofouling. Lipp et al. (2003) and Panglisch et al. (2008) suggest that PES membranes can only be cleaned with caustic or acidic chemicals when natural waters are used; they found oxidising chemicals such as hypochlorite and hydrogen peroxide to be ineffective. These results are only partially backed up by Zondervan and Roffel (2007). They found that NaOH had both high instant cleaning rate and high overall cleaning efficiency on PES UF membrane fouled by surface water. But they also achieved very similar results with NaClO which is an oxidising cleaning agent. Zondervan and Roffel claim that acidic chemicals perform weakly when membrane is fouled by organic foulants. Poor performance of acid chemicals on organic fouling is supported by Lerch et al. (2005b) and Yamamura et al. (2007). One reason for the differences in experimental results might come from the characteristics of the water used for filtration as in Germany, where Lipp et al. and Panglisch et al. have done their studies, the water usually has lower pH and higher content of inorganic components.

For biofouling Baker and Dudley (1998) suggest a three stage cleaning. At first an alkaline cleaning agent should be used to break down organic fouling. Secondly microbiological growth is eliminated with non-oxidising biocide. Lastly alkaline cleaning agent is applied again to remove micro-organisms and organic debris. Acidic cleaning could also be considered as the last cleaning but not before, as certain humic acids might be difficult to remove during acid conditions. Oxidising chemicals can be used as well, if membrane can withstand them. Liang et al. (2008) recommended a combination of NaClO and NaOH when membrane has been subjected to biofouling.

Finding the correct chemicals to regain original permeability might be difficult as the membrane material might affect the fouling. Almgren (2008) found that during similar fouling conditions, for a hydrophilic in-out PES membrane sulphuric acid and sodium hydroxide were capable to regain membrane permeability but for a neutral immersed PVDF membrane sodium hypochlorite, phosphoric acid, sulphuric acid, citric acid, and sodium hydroxide each failed to regain the original permeability. One reason for this difference might be that the PVDF membrane was found to be 80 % fouled by organic substances mainly originated from microorganisms and 20 % by inorganic substances meaning that two different foulants were present. However, no clear reason for the irreversibility of the fouling was found.

Although Lipp et al. (2003) found chlorine very effective in recovering membrane permeability using chlorine-based chemicals may cause problems when CEB is performed. Lipp and Baldauf (2002) point out that dissolved organic substances which have accumulated at the membrane surface may formulate disinfection-by-products (DBPs) when chlorine is introduced during CEB. DBPs formed during CEB cause problems when washwater is discharged into wastewater system. Moreover DBPs have been found from permeate right after backwash (Lipp et al. 1998). Zondervan and Roffel (2007) point out that overdosing of chemicals leads to an increase of TMP, instead of decline. The pores will become covered with cleaning agents but the loss is not permanent and can be eradicated with extensive rinsing.



## **4 Retrofitting with membranes**

Retrofitting existing water treatment processes with membranes has become increasingly interesting. In this chapter the reasons for retrofitting will be discussed, followed by statistical information on how existing WTPs have adopted membranes into their treatment process. After this issues related to cost of retrofitting and changes in water balance of the treatment process will be discussed. It should be noted at this point, that some of the information gathered for this chapter is from presentations and reports released by authorities, membrane manufacturers, or consultant companies and thus should not be taken as absolute truth but rather as suggestive information to support the decision making. Very few scientific and biased reports have been released and this is why the aforementioned sources are used.

### ***4.1 Reasons for retrofitting***

The cost of LPM systems might be slightly less than for a conventional treatment at the moment (Furukawa, 2008). This, however, is highly dependable on what is the limiting factor in treatment process. When turbidity is removed, membranes are a low-priced solution. When NOM removal is targeted, membranes are prone to fouling and require pretreatment. Both problems result in higher costs. However, the affordability of membranes has resulted in a rapid growth of membrane applications in drinking water treatment. One area of growth is changing existing treatment plants conventional treatment to LPMs, which require far less land area. LPMs are also used for debottlenecking (Howell, 2004).

One other big reason for the increasing interest in retrofitting membranes is problems with the treated water quality of the conventional treatment processes. With seasonally fluctuating conditions in raw water quality comes changing effluent quality of the treatment process. Problems with high effluent turbidity and residual chemicals are common at WTPs. In Scandinavian countries, where water resources are copious, trend is the adoption of multi-barrier systems to ensure hygienically safe water (Ødegaard, 2006). Water quality problems in Scandinavian countries are related to turbidity, iron and manganese, pathogens, and humic substances.

In Gothenburg, Sweden, Lackarebäck WTP was proven to be inefficient towards removing microorganisms from the water (Almqvist et al., 2012). In a worst case scenario an outbreak of microorganisms would have ceased regular water supply for a

long time. Currently the WTP is retrofitting UF membranes to their process to guarantee the safe and continuous supply of potable water (Egle, 2012). Problems with increasing amounts of NOM have also forced WTPs to look for additional processes. LPMs in combination with coagulation can remove NOM (Pikkarainen and Laine, 2011).

Some WTPs have experienced permanent deterioration in raw water quality. Reasons for this might be overpopulation, overuse of raw water source, or long droughts. This deterioration brings about similar problems as the fluctuating conditions. Such problems have been increasing in the United States (US EPA, 2005). Howell (2004) and Ødegaard (2006) noticed similar trends in Europe. Multi-barrier systems are increasing in numbers but the focus is on microorganisms like *Cryptosporidium* and *Giardia*. Due to increasing water stress especially in Southern Europe, the quality of raw water sources is deteriorating. Man-made pollutants, such as pesticides, pharmaceuticals, and genes in raw water sources are causing problems in addition to naturally occurring matter.

Howell (2004) recognises removal of parasites, bacteria, and toxic metals as the main water concerns in developing countries. In developing countries desalination is also important step in treatment process. In Middle Eastern countries the use of reverse osmosis for brackish groundwater treatment and seawater desalination is increasing and so is the use of LPMs as pre-treatment.

Simultaneously as the raw water quality deteriorates the quality requirements set by authorities for potable water have become stricter. Increasing amount of research has shown the harmful effects of by-products created by conventional treatments on the health of the public. Chlorine added for disinfection has been shown to react with NOM and create disinfection-by-products such as trihalomethanes (THMs) and haloacetic acids (US EPA, 2005). Similarly authorities have started to regulate the amounts of residual metals in the potable water as these might also cause severe health problems. Iron and aluminium are usually results of ineffective coagulation, whereas manganese and other inorganic compounds are already present in the raw water.

Most aforementioned problems could be solved by using conventional processes instead of membranes. However, the reason for the increasing numbers in WTPs choosing to retrofit membranes stems from three factors: membranes have a very high packing density meaning that they require very little space, the permeate quality regarding

turbidity is constantly excellent independent of the feed water quality, and the total costs of membrane process might be less than conventional treatment. Removal of NOM is still, however, dependent on coagulation process. Additionally, once membranes have been installed, increasing the treatment capacity is simple with membranes, as modules are fairly easy to install next to old modules. These factors have contributed to the exponential growth that membrane retrofits have experienced in the recent years (Furukawa, 2008).

## 4.2 Retrofitting membranes to the existing process

To find out how WTPs have retrofitted membranes into their treatment process, information from several sources was gathered. This gathered information is presented in appendix 1. WTPs are mostly from Europe and USA. Table 5 has been assembled based on this data. It presents the different ways WTPs have adopted membranes, whether they have opted to use pre- or post-treatments and also the popularity of these pretreatments. These percentages are compared to percentages presented by Lipp (2006) who studied the WTPs with membranes in Germany.

**Table 5. Information on how existing 47 examined WTPs have retrofitted membranes. 60 % of studied WTPs have used pretreatment before membrane filtration.**

Process Description	% of total	Lipp (2006) %
LPM alone or with disinfection	33	19
LPM + Post-treatment	5	5
Pretreatment + LPM	38	14
Pretreatment + LPM + Post-treatment	21	14
N.N.	-	48
Backwash treatment	13	No information
Number of plants	47	91

As can be seen from Table 5, LPMs are most often used with pretreatments at the end of the treatment process. According to Pikkarainen and Laine (2011) this is the most obvious place to install membranes, as the quality of feed water is good and thus the operation of LPM filtration is stable and the operational costs are low. LPMs alone or with disinfection are popular at small WTPs where the raw water quality is already good. Third choice is to use pretreatment before membrane filtration and subsequently post-treatment to further improve the water quality. However, as can be seen from the percentages of different process descriptions, LPMs are used in many ways which shows their adaptability and robustness.

An interesting addition to the usability of membranes is instalment of membranes for backwash treatment line. Altogether 14 WTPs examined used membrane to treat backwash, eight of them used membrane exclusively for backwash treatment. In some WTPs membranes are installed to treat the backwash water from conventional treatment steps, such as sand filtration, whilst some WTPs have chosen to treat the backwash from the primary membrane filtration line. Membranes are usually retrofitted to treat the backwash water of conventional treatment because of the strict regulations on the quality of wastewater effluent from the WTP. Reissmann and Uhl (2006) have proven that backwash water from conventional treatment can be filtered through an UF membrane and reused without any safety concerns. Alternatively, membranes are chosen to treat the backwash from the primary membrane line to increase the recovery of the treatment process.

Another way to interpret the data presented in appendix 1 is to study the functions for which LPMs are retrofitted for. The different functions and percentages are presented in Table 6. Direct filtration means that the water is treated only by a strainer. Final polishing means using membranes at the end of the water treatment process to ensure microbial safety and to remove chemicals used in the treatment process.

**Table 6. Different functions to utilise membranes in existing WTPs and the popularity of these alternatives. Percentages prove that membranes can be used for various purposes.**

Function	%
Direct filtration	38
Replace sedimentation and/or sand filter	36
Final polishing	26

It can be seen, again, that membranes can be utilised for different purposes. The use of LPM for direct filtration of raw water is most common. Mainly MF membranes are used for direct filtration as UF membranes require a better feed water quality. When this data is compared to the capacity of WTP (appendix 1), it can be concluded that small WTPs (capacity < 500 m<sup>3</sup>/h), use LPMs for direct filtration. Larger treatment plants are using LPMs to replace sedimentation and/or sand filtration and as a final polishing step before water is delivered to consumers. The treatment complexity is, however, dependent on the quality of raw water. Good quality raw water only needs membrane filtration, such as in Keldgate WTP, where 3750 m<sup>3</sup>/h is treated with an UF membrane as direct filtration.

In the following paragraphs few retrofits are studied in more detail. The only WTP currently operating with LPMs in Scandinavia is Lello WTP in Levanger commune in

Norway. Because the WTP uses good quality groundwater as raw water, the treatment process is fairly simple. Pre-filtration with a strainer with a pore size of 50 µm is applied and PAX-coagulant is added to reduce membrane fouling before the water is led to the ultrafiltration modules. After ultrafiltration the water is treated with NaClO and Na<sub>2</sub>SiO<sub>3</sub> to disinfect the water and to raise pH, respectively. Ultrafiltration was retrofitted to Lello WTP because of the reduced capacity and high OPEX on nanofiltration (Skrøvseth, 2009; Sandvik and Ødegaard, 2010).

In Gothenburg, Sweden, the treatment process at Lackarebäck WTP was found to be vulnerable against microorganisms in raw water. To reduce the risk of an outbreak of water borne diseases ultrafiltration will be retrofitted. The retrofit is expected to be finished in 2016. The treatment process after the improvement will be following: raw water from River Göta is precipitated with Al<sub>2</sub>SO<sub>4</sub> and the floc is removed in sedimentation basins. After sedimentation the remaining particles are removed with granular activated carbon (GAC) filters. Then water will go through ultrafiltration membrane and finally the treated water is adjusted with limewater, NaOH, and chlorine (Almqvist et al., 2012).

In Roetgen, Germany, the old treatment process suffered from inefficient removal of humic substances, particles, and inorganic substances i.e. iron, manganese, and ammonium. After eight years of intensive research the WTP was retrofitted with UF membranes in 2006. The new treatment process consists of five steps. First step is pre-filtration, which can be enhanced by adding PAC to the raw water. Then aluminium-based coagulant is added. After flocculation the water is filtered through an UF membrane followed by limestone filtration. Finally the water is disinfected and piped to storage. At Roetgen WTP the backwash water from the UF membranes and limestone filters is also treated, however, the CEB water is led directly to waste treatment. The capacity of the backwash treatment line is approximately 10 % of the primary treatment line (Holy et al., 2004; Panglisch et al., 2005; Panglisch et al., 2008).

Ratnayaka et al. (2008) reported on the retrofit of Chua Chu Kang Water Works in Singapore with immersed ultrafiltration membranes. The goal was to improve the quality of treated water, particularly total removal of algae, microorganisms and the reduction of THMs. They retrofitted ultrafiltration membranes in the existing rapid gravity sand filter tanks thus keeping the plants environmental footprint the same. Similar retrofit

was also done in the U.K., where the clarification tanks of St. Saviours WTP were used to house the new submerged LPMs (Redhead, 2008).

Membranes have proven to be robust and flexible technology that can be retrofitted to an existing WTP to enhance the treatment process and final treatment quality in many ways. Measures such as using existing infrastructure have been taken to reduce the cost of retrofit. However, still the cost is a concern as many factors have to be considered to realise the final costs of retrofitting. These factors will be discussed next.

### **4.3 Cost of retrofitting**

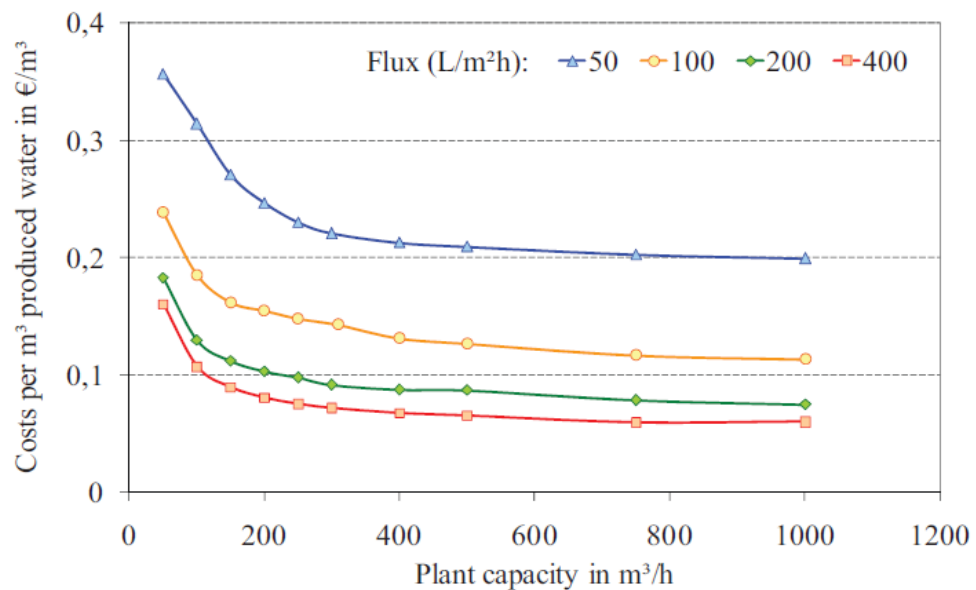
One reason for the rising interest in retrofitting LPMs to existing processes has been the increasing number of researchers claiming that the operating expenses (OPEX) for LPM system are lower than for conventional treatment (Furukawa, 2008). Still, very few cases of OPEX or capital expenses (CAPEX) were found and thus some numbers which are presented here, especially numbers in Scandinavian countries, are calculated by consultancies and a certain caution towards these numbers should be expressed.

#### **4.3.1 Specific cost of retrofitting**

Specific cost is the cost that is added to the total costs of water treatment process when membranes are retrofitted. It consists of both CAPEX and OPEX. For example Panglish et al. (2008) have reported that the specific cost (CAPEX and OPEX) for a m<sup>3</sup> of produced water is less than 0.1 € when depreciation rate of 5 % was used. This 0.1 € was added to the costs of Roetgen WTP. The specific cost of the treatment step replaced by membrane is reduced. At Varberg WTP in Sweden, UF was compared with ROS - contact filter. The estimated specific cost for an UF system was 0.05 €/m<sup>3</sup> whilst for the contact filter the total costs were estimated to be approximately 0.03 €/m<sup>3</sup> (Heinicke and Zagerholm, 2012). The depreciation time used was 25 and 50 years for process equipment and infrastructure, respectively. Higher specific costs for retrofit have been given by Lipp et al. (2005). They summarised LPM filtration costs in Germany and concluded that for LPM treatment the specific cost is 0.1...0.3 €/m<sup>3</sup>.

Heijman and Bakker (2007) reported on a pilot experiment treating Twente Canal surface water with ceramic membranes. Despite the fact that ceramic membranes require a higher operating TMP, they found that the specific cost of ceramic membrane system for a WTP with a capacity of 200 m<sup>3</sup>/h would be 0.12 €/m<sup>3</sup> when depreciation

times of 10, 15, and 40 years were used for membranes, automation, and building, respectively. Similar specific costs have been calculated by Meyn (2011). Numbers by Meyn are presented in Figure 8. Of the specific cost presented by Heijman and Bakker, 50 % comes from membrane modules while the relationship between OPEX/CAPEX in specific cost is 30/70, similar to polymeric membranes. Heijman and Bakker have not counted the price of module replacement which was a significant cost factor with polymeric membranes.

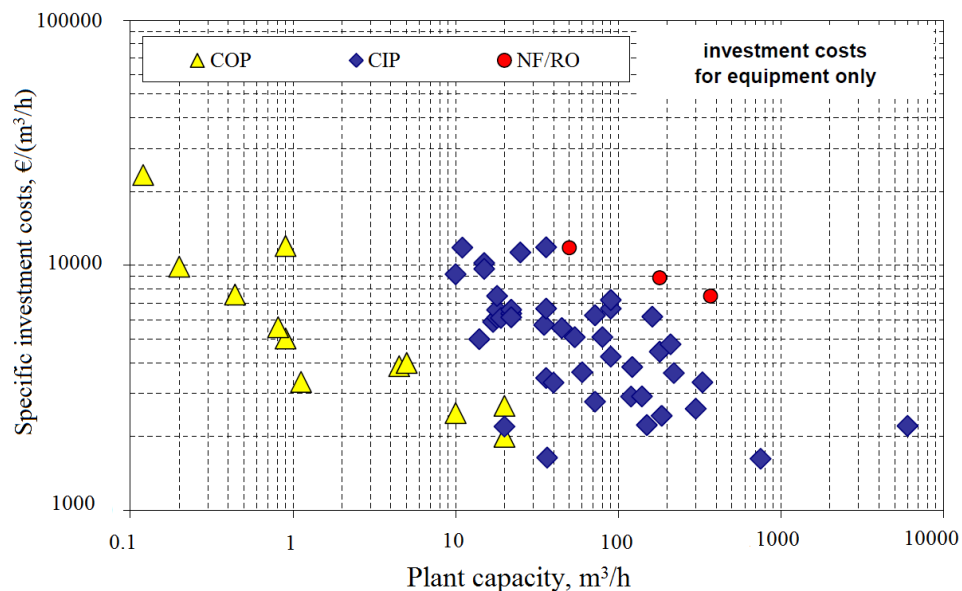


**Figure 8. Treatment cost in dependence of membrane flux (Meyn, 2011). Higher flux results in lower specific cost but the decline is not linear. (Assumed maximum capacity; specific membrane cost 250 €/m²; interest loan 5 %; price increase for on-going costs 0 %.)**

When looking at the specific costs for a retrofit, it can be concluded that the specific cost, both CAPEX and OPEX, is 0.05...0.12 €/m³, depending on the complexity of the retrofit and on membrane material. Biggest investments are membrane modules and process technique around the membranes, on condition that constructing a new building for the membrane modules is unnecessary.

Sorgini (2003) has suggested that the CAPEX and OPEX of submerged systems are significantly smaller than pressurized system. She studied costs of projects from same manufacturer and similar membrane materials and concluded that capital and energy costs for submerged systems were 37...38 % and 20...29 % smaller, respectively. No such observations were made by Holy et al. (2004) when comparing different configurations for Roetgen WTP but reasons to support the propositions by Sorgini regarding energy costs, chemical consumption, and water balance are presented in latter chapters.

Lipp (2006) has studied the specific cost of equipment from WTPs in Germany; these costs are presented in Figure 9. From this figure it can be seen that small treatment plants use COP and larger plants CIP. CIP decreases downtime and thus lowers operational costs; initial costs however might be higher. As plant capacity increases there is a decreasing trend in specific investment cost. It can also be observed that the specific investment costs are lower for LPMs than for RO and NF systems. According to Pikkarainen and Laine (2012) the specific cost given by manufacturer Inge AG for membrane installations are similar to what Lipp has presented, 2500...4000 €/m<sup>3</sup>/h for small plants, 1500...3000 €/m<sup>3</sup>/h for medium sized plants, and 1000...1200 €/m<sup>3</sup>/h for large plants treating above 800 m<sup>3</sup>/h.



**Figure 9. Specific investment costs of equipment for different WTPs (Lipp, 2006). The increase in WTP size results in lower specific cost. NF/RO systems have a higher specific cost than LPMs.**

To conclude, it can be said that the specific cost (CAPEX and OPEX) for retrofitting LPMs to an existing WTP is 0.05...0.12 €/m<sup>3</sup> meaning that the total treatment costs for a WTP is raised by this amount if membranes are retrofitted. However, higher costs have also been reported. Specific cost is highly dependable on the complexity of the retrofit.

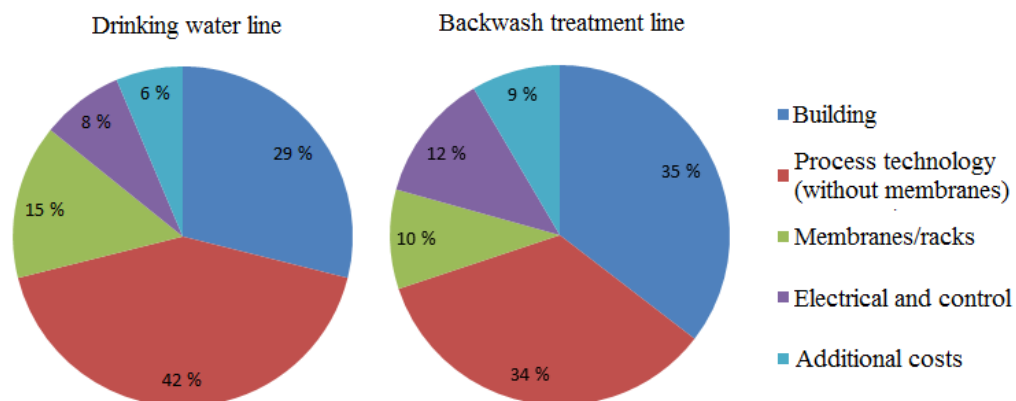
#### 4.3.2 Capital expenses

CAPEX of retrofitting depends mainly on how much infrastructure is required and on the possibility of utilizing the existing infrastructure. Ratnayaka et al. (2008) and Redhead (2008) have shown that existing sand filtration tanks can be used when retrofitting an existing plant and this has led to smaller CAPEX.



The CAPEX for retrofitting Roetgen WTP in Germany was approximately 25.5 M€ for a treatment capacity of 7000 m<sup>3</sup>/h (Panglisch et al., 2008). Panglisch et al. told that CAPEX for the drinking water treatment line was 20.5 M€ and for the backwash water line the CAPEX was 5 M€. Despite the backwash water treatment line only treating about 10 % of the amount of drinking water treatment line, the CAPEX of backwash treatment was 20 % of the total CAPEX. One reason for this might be the complexity of the backwash treatment line. In Germany the regulations for effluent from a WTP are very strict and the backwash treatment line includes sedimentation tanks, chemical treatments, and centrifuges for sludge treatment.

The cost breakdown of CAPEX of retrofitting Roetgen WTP is presented in Figure 10. From this figure evidence for the above mentioned statements can be found. The cost for the building is much higher, probably due to sedimentation tanks. The costs for electrical and control and for additional costs are higher as well, which might be because of the complexity of the treatment line.



**Figure 10. CAPEX breakdown at Roetgen WTP (adapted from Panglisch et al., 2008). Process technology and constructing a new building have highest costs, membranes are only 10...15 % of total costs. This implies that simple retrofits have much lower CAPEX.**

In Gothenburg, Sweden, the retrofitting of Lackarebäck WTP with a treatment capacity of 7750 m<sup>3</sup>/h is estimated to cost 80 M€ (Egle, 2012). This number is vastly different to CAPEX in Roetgen. In Gothenburg eight new active carbon filters are built as well, but this is not enough to explain the huge difference. The costs given for Roetgen include the secondary treatment line treating of backwash water, which costs 5 M€. In Gothenburg no secondary treatment line is going to be installed. Heinicke and Zagerholm (2012) estimated that retrofitting Varberg WTP in Sweden needed an investment of 4.3 M€ in a WTP with a capacity of 1080 m<sup>3</sup>/h.

All these projects included building a completely new building for the membranes. Thus it can be concluded that the total cost compared to capacity for a retrofit with complete infrastructure is between 3600...10300 €/m<sup>3</sup>/h) and that the specific cost for equipment is roughly 10...33 % of the total CAPEX. In all treatment plants UF membranes were selected. The cost for MF filtration might be little less, as MF membranes have generally a higher flux and thus higher productivity. These numbers are, however, very rough and the cost breakdown and estimation of total cost is very difficult.

### 4.3.3 Operational expenses

Operational expenses in a membrane system consist of energy and chemical consumption, cost of maintenance and module replacement, and the earnings lost because of membrane backwashing. When studying the numbers presented by WTPs it should be remembered that the performance of a membrane system is highly dependable on climate conditions. Liikanen et al. (2006) have reported that one stumbling block for membrane filtration in Nordic countries is that the seasonal variation causes up to 20 % higher total costs comparing to warmer climate. The operational cost of both treatment lines combined at Roetgen WTP was 0.37 M€ annually (Panglisch et al., 2008).

The OPEX of Lello WTP in Norway was presented by Sandvik and Ødegaard (2010). This data plus OPEX of five other WTP in Europe are presented in Table 7. WTPs presented use polymeric membranes. Operational costs of LPM system are approximately 0.04...0.05 €/m<sup>3</sup> of treated water. This is, however, highly depending on the complexity of the system. At Lello the costs for chemicals and energy are much higher than in the five other cases presented. One reason for the high chemical consumption is that unlike all other WTP, in Lello coagulant is used before the membrane filtration.

**Table 7. OPEX of UF at six WTP using UF (Hagen and Theis (as cited in Meier et al., 2006); Klahre and Robert (as cited in Meier et al., 2006); Sandvik and Ødegaard, 2010). The costs for chemicals and energy at Lello are higher partially due to coagulation pretreatment and higher energy cost.**

WTP	Chemicals (c/m <sup>3</sup> )	Energy (c/m <sup>3</sup> )	Maintenance (c/m <sup>3</sup> )	Module replacement (c/m <sup>3</sup> )	Backwash (c/m <sup>3</sup> )	Total (c/m <sup>3</sup> )
Hermeskiel	0.4	0.4	1.2	2.5	0.4	4.9
Torgon	0.07	0.5	2.5	1.7	-	4.8
Lello	1.4	1.9	1	-	-	4.3
Vionnaz	0.07	0.7	1.7	1.6	-	4.1
Thyon	0.005	0.5	1.5	1.5	-	3.5
Muotathal	0.07	0.4	0.9	1.6	-	3.0

Module replacement is usually calculated by using information given by manufacturers, who say that the operational period of a polymeric membrane is seven years. After three years of experience from Roetgen WTP Panglisch et al. (2008) observed that membrane lifetime might be even longer, as only two broken capillaries had been identified, which resulted in a defect rate of  $3.6 \times 10^{-8}$ . Meyn (2011) has estimated that the expected lifecycle for ceramic membrane is 12 years which lowers the operational expenses for a ceramic system.

Energy requirement for LPMs is generally higher than requirements for conventional treatment. The process step that is usually replaced by LPMs is sand filtration. Energy requirement for gravity filter is said to be in the range of 0.005...0.014 kWh/m<sup>3</sup> (WEF, as cited in Plappally and Lienhard, 2012). Crozes et al. (2003, p. 25) say that energy consumption of UF membrane is approximately 0.026 kWh/m<sup>3</sup>. Manufacturers give an average consumption in the range of 0.02...0.05 kWh/m<sup>3</sup>. However, WTPs that have retrofitted MF membrane treatment into their treatment process have reported on an increase of 0.0...0.22 kWh/m<sup>3</sup> in energy consumption after retrofit, which is notably higher than the average given by manufacturers (Elliott et al., 2003, p. 38; Redhead, 2008).

Ratnayaka et al. (2008) reported that power consumption of ultrafiltration system was not directly proportional to the water production. The most energy efficient production in Singapore at Choa Chu Kang WTP was achieved with production of 91 000 m<sup>3</sup>/day. Though a lower pumping energy was required for a lower production, a similar amount of power was needed for air blower and pumps for recirculation and backwashing. They reported an energy consumption of 0.007...0.008 kWh/m<sup>3</sup> treated water.

Chemical consumption required for coagulation prior to LPM filtration is said to be lower than what is required for conventional. Xia et al. (2008b) compared ultrafiltration with coagulation to conventional treatment by coagulation, sedimentation, and sand filtration at Binxian WTP. They found out that membrane filtration required less chemical usage and produced better quality water regardless of the raw water quality. The results gotten by Xia et al. are presented in Table 8.

**Table 8. Added coagulant and treated water quality (Xia et al., 2008b). UF treatment has resulted in less residual turbidity although lower amount of coagulant was used.**

Raw water	Coagulant added (mg/l)		Treated water (NTU)	
	WTP	Direct UF	WTP	Direct UF
21 NTU	10	3	0.8	0.1
107 NTU	23	8	0.9	0.1
423 NTU	40	15	1.3	0.1

Best et al. (1999) also claimed that immersed ultrafiltration enhanced coagulation would require less added chemicals than a conventional treatment. The explanation for the lower chemical consumption is the membrane pore size. Since membrane pore size in ultrafiltration is usually 0.01...0.02  $\mu\text{m}$ , the size of the micro-flocs only needs to exceed 0.1  $\mu\text{m}$  for the membrane to effectively separate them.

Chemicals are also used for CEB and CIP/COP. The cost of using chemicals can be estimated from Table 7 as all other WTPs except Lello are not using coagulants. It can be seen that the cost of chemical for chemical cleanings is between 0.005...0.4  $\text{c}/\text{m}^3$ . One manufacturer has estimated that the chemical cost for LPM treatment is 0.05...0.3  $\text{c}/\text{m}^3$  (Inge AG, 2012).

Operational expenses depend on a number of factors. OPEX of a membrane filtration system is approximately 0.04  $\text{€}/\text{m}^3$  but this number is very rough. If coagulation is used to enhance the permeate quality the cost is considerably higher. A significant portion of OPEX, 40...50 %, comes from membrane replacement. However, costs given for membrane replacement are not based on actual operational experiences but they are estimated from expected lifetimes given by membrane manufacturers. Energy consumption of water treatment process can be estimated to raise 0.0...0.22  $\text{kWh}/\text{m}^3$ , depending on whether LPMs are attached to the existing process or retrofitted to replace treatment steps. If LPM filtration is retrofitted to replace sedimentation and sand filtration, chemical consumption might be less, resulting in a smaller total chemical costs.

#### **4.4 Effect on water balance**

As water is a scarce resource in many corners of the world, significant efforts have been made to enhance the recovery of LPM filtration and thus the total water balance of WTP. As previously in chapter 2.1.1 was mentioned, LPMs are capable of recoveries above 95 %. These recoveries are similar to sand filtration (Crittenden et al., 2005). The recovery percentage depends on flow regime, flux through membrane, and backwash frequency. Using crossflow regime the recoveries are usually below 25 %, but for a dead-end the recovery is theoretically 100 %. However, membranes treating surface waters tend to foul due to inorganic and organic substances present in the water. Thus water is lost because of backwashing. At Roetgen WTP the recovery percentage of the primary membrane treatment line is 90 %, ten per cent is used to conduct membrane

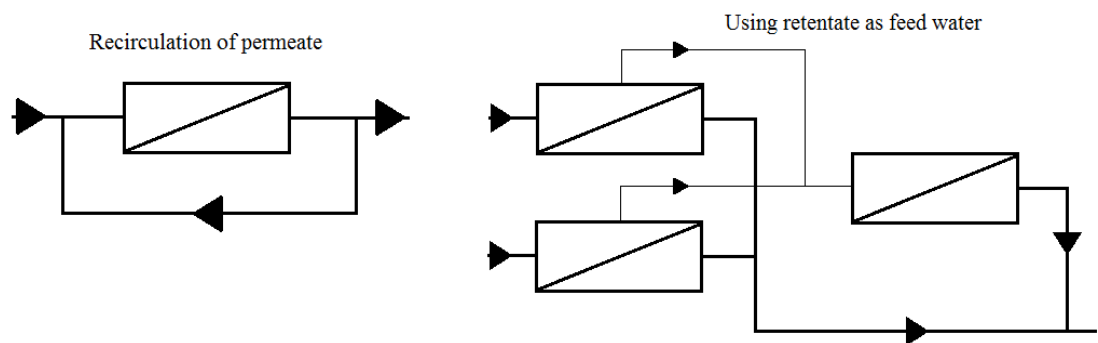
backwashes (Panglisch et al., 2005). Persson et al. (2005) reported that the recovery percentage of an immersed system pilot operating with water from Göta River was approximately 98 %, thus confirming the allegations of Pikkarainen and Laine (2012) that were presented in chapter 3.1; immersed system has a better recovery due to lower operational pressure.

To further increase the recovery percentage backwash water can be treated. Heijman and Bakker (2007) reported that with ceramic membranes treating Twente Canal surface water the recovery was 96.7 %, with optimized conditions a recovery rate of 98.9 % might be reached periodically. But if backwash was treated with a thickener and the supernatant was recycled back to the filtration process recovery rate of 99.9 % could be achieved. Instead of using a thickener the backwash water can also be treated with membranes. Many WTPs have decided to use UF membranes in their backwash treatment line. Permeate of the secondary line can be returned back to the primary treatment line, before the membrane filtration step. In addition to increasing the recovery also better quality effluent can be reached. In Roetgen the recovery of a two-stage process is 99.5 % (Panglisch et al., 2005; Gimbel et al., 2007).

The backwash interval should also be optimized to increase recovery. Lipp et al. (1998) optimised the performance of an UF membrane to minimise waste water demand. For a filtration cycle of one hour, five hour, and ten hour, the waste water demand was 5...7.5 %, 1...1.5 %, and 0.5...0.75 %, respectively. Filtration times as long as ten or five hours are not recommended for a LPM filtration of clarified water, and the membrane had problems even with 45 minute cycle thus decreasing the interval of chemical cleanings which in turn shortens the membrane life. Therefore, optimisation of backwash interval has to be done carefully and long term effects should be estimated.

Adding air to the backwash process can increase the interval and shorten the duration of backwash. Lipp and Baldauf (2002) reported on three different studies where backwash with permeate was supported with air scour. Air scour adds mechanical scrubbing to backwashing. This method was found to be very effective in keeping TMP low and minimize TMP increase. The wastewater demands for backwash with permeate and air scour in the three reported studies were 1...2, 4, and 5 %. The backwash interval for all studies was 30...60 minutes. Wastewater demand for CEB ranged from 0 to 0.3 % related to the permeate production. Lipp and Baldauf also report that the chemical demand of an air scour supported backwashing is less than backwash without air scour.

The recovery of a simple LPM filtration process is low but with an enhanced system recovery close to 100 % can be reached. Common method with LPMs is treatment of backwash water by using a secondary LPM filtration line, from which permeate can be recycled back to the primary filtration line. Another way to increase capacity is recirculation of permeate. These techniques are presented in Figure 11. When recirculation is used, the required treatment capacity of membranes is increased, e.g. if 10 % of permeate is recycled, the capacity of treatment line has to 1.1 times the amount of the raw water to the WTP.



**Figure 11. Methods to increase recovery of membrane filtration. The required membrane surface area has to be calculated based on the total amount of treated water, which makes designing complex systems challenging.**

## 5 Pretreatments

The integration of pretreatments with LPM filtration is widely used in full scale, as pointed out in Table 5. They are used to reduce membrane fouling and increase the removal of certain aquatic contaminants and NOM. These pretreatment strategies will be discussed in this chapter.

### 5.1 Principles of pretreatment

Processes conducted prior to membrane filtration, known as pretreatments, have become more common in water treatment processes for two reasons. Firstly pretreatment enhances the removal of aquatic contaminants and secondly it reduces membrane fouling. Both of these factors influence the operational and capital cost of a LPM system (Huang et al., 2009).

As the conventional and membrane-based systems have many disadvantages in the process of NOM removal, a combination of the processes must be applied to reach high quality potable water (Glucina et al., 2000; Kabsch-Korbutowicz, 2005a). Glucina et al. studied the applicability of conventional treatment and UF as pre-treatment for reverse osmosis. The study used water from River Seine upstream of Paris as raw water. The conventional treatment in the study consisted of coagulation, flocculation, sedimentation, and GAC filtration. Pre-filter with a pore size of 200  $\mu\text{m}$  was used to treat the raw water before ultrafiltration. As the raw water had a high fouling potential Glucina et al. concluded that a pre-treatment step would be economically interesting in order to improve ultrafiltration performance. Alternatively, ultrafiltration was able to completely remove microorganisms from the water, which the conventional treatment was not able to do. It should be noted that an ultrafiltration membrane with relatively high molecular weight cut-off was used in this study which resulted in a minimal NOM removal. A membrane with smaller pore size might have achieved a better NOM removal, but this might have led to a more severe fouling problem.

Pretreatment can affect the feed water by altering chemical, physical, or biological properties. Physical methods increase the size of aquatic contaminants and shifts membrane fouling from pore blocking and inner pore adsorption to cake layer formation. Chemical mechanisms alter the water chemistry and reduce the affinity of foulants to membrane surface. Biological methods can remove biodegradable contaminants relevant to membrane fouling or biofilm formation (Huang et al., 2009).

The most common pretreatment options chosen for LPM filtration are coagulation, adsorption, oxidation, and filtration. Various other methods have been created to pretreat water before membranes, e.g. biological pretreatment, dissolved air flotation, and photocatalytic oxidation, but since these are marginal technologies mostly used at pilot-scale studies they will not be further discussed in this text. One interesting pretreatment that might have future potential is membrane precoating. Membranes can be precoated to reduce fouling and increase flux but this might affect permeate quality (Maartens et al., 2000; Pezeshk and Narbaitz, 2012).

## **5.2 Coagulation**

Coagulation is the most widely used pretreatment for low pressure membranes as membranes are incapable to remove NOM. When combined with coagulation the removal efficiency of NOM is significantly enhanced (Howe and Clark, 2006; Chen et al., 2007). The variations on experimental conditions and reported values makes it difficult to draw conclusions on the specific effect of coagulation on membrane performance but no literature was found that claimed coagulation to have no or negative effect on NOM removal. However, membrane material and type are significantly affecting the efficiency of coagulation. Howe and Clark (2006) found that coagulation was more advantageous for MF membrane than UF membrane, while Tran et al. (2006) found aluminium coagulant at optimized dose to reduce fouling of a PVDF MF membrane whilst fouling of PP MF membrane was worsened.

Coagulation is targeted against viruses, humic and fulvic acids, proteins, polysaccharides with acidic groups, and colloids smaller than membrane pores (Huang et al., 2009). Coagulants are either organic macromolecules or inorganic metal salts. Metal salts are more used than organic macromolecules, most popular salts are iron and aluminium salts (Huang et al., 2009; Gao et al., 2011). Polyaluminium chloride and other polymeric inorganic coagulants can also be used. The required dosage for coagulation is usually derived from the amount of NOM in raw water. Literature reporting about the use of organic molecules as pretreatment for membranes is scarce and thus organic molecules will not be discussed.

Coagulation takes place either as standard coagulation or as in-line coagulation, where flocculation and sedimentation processes are absent and the coagulated water is directly introduced to the membranes. According to Huang et al. (2009) and Meyn (2011)



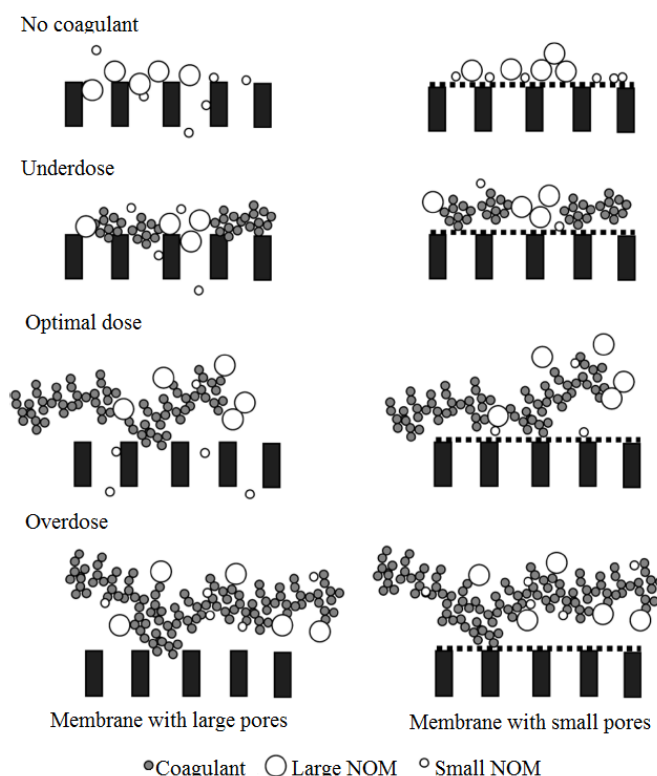
coagulation combined with flocculation with or without sedimentation causes less fouling but still do not completely remove foulants. To reduce fouling flocculation time should be no less than 45 seconds (Panglisch et al., 2005; Leiknes, 2009, Meyn, 2011). Longer time might give a bigger reduction of reversible fouling but irreversible fouling is not affected by longer flocculation time. Also DOC removal is only slightly better with longer flocculation (Leiknes, 2009). In-line coagulation is used when the footprint of the process is minimised but it suffers from more extensive fouling (Howe and Clark, 2006; Meyn, 2011). During in-line coagulation the coagulants may accumulate on the membrane surface and cause fouling (Huang et al., 2009).

Yu et al. (2013) studied the differences in floc formation in traditional coagulation and submerged coagulation which takes place in a submerged membrane filtration tank. Coagulation was found to be more effective in a submerged system. It resulted in a thinner cake layer and less chemical usage whilst creating larger and lower density flocs. Yu et al. claim that a cake layer formed by flocs with low density has higher porosity than that formed with higher density flocs and thus increases flux through the cake layer.

The efficiency of NOM removal when coagulation is integrated with membranes has been studied intensely and it is clear that coagulation is needed for NOM removal. Keucken et al. (2012) have studied the effect of coagulation as a pre-treatment for ultrafiltration on NOM removal from Swedish surface water. The efficiency of humic substance removal was 46 %, as comparison ultrafiltration without coagulation was unable to remove any humic substances. Similar results were presented regarding lower molecular weight (MW) fractions, ultrafiltration without coagulation showed no effect whilst ultrafiltration with coagulation performed better, building blocks (300-500 MW) and low molecular weight fractions (<350 MW) were reduced by about 10 %. However, as mentioned before, coagulation is incapable of removing all foulants from the water. Kimura et al. (2005) found that coagulation pre-treatment was inefficient in removing polysaccharides and proteins which caused irreversible fouling while Chen et al. (2007) found coagulation to be ineffective in removing hydrophilic lower MW fraction of NOM.

Measures used for conventional coagulation process, followed by sedimentation and media filtration may not be efficient when coagulation is followed by membrane filtration. Proper dosage of chemicals, type of coagulant, and mixing conditions need to be reassessed. Vickers et al. (1995) and Gao et al. (2011) say that a good settling ability,

something which in conventional processes is targeted, is irrelevant for membrane filtration and instead there should be a critical size for the flocs. Chemicals should not be overdosed or underdosed (Figure 12). Kabsch-Korbutowicz (2006) studied different aluminium based coagulants and found different NOM removal rates for each chemical, proving that coagulant has an effect as well.



**Figure 12. Effect of dosage amount during coagulation (Huang et al., 2009). In optimal conditions coagulant is removing substances it is targeted towards and membrane covers smaller substances.**

Another factor related to coagulation that has gathered much attention is pH during coagulation. Kabsch-Korbutowicz (2005a; 2005b) showed that pH plays a role in the effect of coagulation process in removing NOM. With aluminium-based coagulants pH 5 is advantageous when coagulation alone is used as flocs had largest surface area and good settling ability. When UF membrane was added, the removal efficiency of the coagulation/UF systems was optimal at pH 6. Lerch et al. (2005a) studied coagulation with different pH and found that at higher pH coagulation resulted in bigger flocs which in turn resulted in less fouling. This finding is supported by Dong et al. (2006). When iron is used as coagulant, pH of 4.5 was found most advantageous by Meyn (2011). However, at this pH the amount of residual iron was very high.

Precipitation at pH 6 is most advantageous for NOM removal when aluminium or polyferric chloride is used as coagulant (Kabsch-Korbutowicz, 2005a; Wang et al., 2012).

At pH 6 the flocs were most compact and resulted in cake layer formation which in turn resulted in smaller pore size and better organic matter exclusion. At Chua Chu Kang Water Works in Singapore, filtration pH in the range of 5.8 to 6.2 was found to have a lower fouling rate (Ratnayaka et al., 2008). At pH outside this range fouling caused by TOC and aluminium increased. The increased fouling was credited to the coagulation process not performing as well outside the mentioned optimal pH range. Panglisch et al. (2008) say that in Roetgen WTP the optimal pH for coagulation/ultrafiltration process is 6.2 to 6.5.

Chemical consumption is also affected by pH. At acidic conditions the negatively charged NOM particles are neutralized by the positively charged products of the coagulant hydrolysis. At higher pH the sweep flocculation is dominant mechanism and this mechanism needs 50...100 % more chemicals for the same removal effect (Soffer et al., 2000; Kabsch-Korbutowicz, 2005b).

However, all focus should not be on adjusting coagulation pH as membrane might have more effect on the success of coagulation. Soffer et al. (2000) argue that the effect of pH on membrane fouling is less severe than MWCO of the membrane and that with membranes with higher MWCO the rate of fouling is faster no matter what pH the coagulant is used. Lerch et al. (2005a) also pointed out that temperature effects coagulation, with decreasing temperatures a decline in membrane permeability was observed. One reason is water viscosity but floc formation in coagulation was also found to be responsible.

The cost-effective coagulation for LPM filtration differs significantly from the optimal for conventional treatment. Coagulant type, dosage, and precipitation conditions have to be reconsidered in order to reach the optimal process that also results in the best possible water quality possible. Therefore, bench-scale or pilot-scale tests are required to find the optimal conditions for a particular source water and membrane.

### **5.3 Adsorption**

Adsorbents can be used as pretreatment before LPM filtration or to further enhance the effectiveness of coagulation as pre-treatment. Adsorbents have a relatively large surface area due to their porosity and dispersity. Adsorbents uptake otherwise poorly removed humic substances and pollutants from water with their thermodynamically unstable surfaces and thus improve permeate quality and results in less fouling.

They also offer an alternative surface for substances to accumulate on. Adsorbents might also adsorb substances relevant to biofouling. Increase in the removal of disinfection-by-products (DBP) has also been observed (Huang et al., 2009).

Powdered activated carbon (PAC) is the most popular adsorbent when adsorption and ultrafiltration are combined (Gao et al., 2011). Granular activated carbon (GAC) and magnetic ion-exchange (MIEX) resin have also been used. PAC can be added directly into the membrane module or it can be used in an in-line PAC pretreatment system, similar to in-line coagulation. Lin et al. (2000) compared direct system and PAC pretreatment in removing NOM fractions and found that PAC pre-treated samples exhibited bigger flux decline but also better permeate quality than direct system. The fouling effect of different NOM fractions was different for these systems. Direct system was mostly affected by hydrophilic fractions whilst unfractionated fraction caused the biggest flux decline for PAC pre-treated samples.

Although adsorbents are widely used as pretreatment, their NOM removal effect is conditional. When adsorbents are combined with coagulation, the effect is clearly positive. Kim et al. (2009) have studied immersed MF membranes with a direct PAC system. The system had operational period twice as long as microfiltration alone. Kim et al. also reported that the removal rate of hydrophobic NOM was especially high, 50...60 %. This resulted in decrease of THM formation. After about 80 days the removal efficiency of the hybrid membrane system became almost the same as a system without PAC. Similar results were presented by Best et al. (1999) who studied immersed ultrafiltration membranes for colour and TOC removal. They used cold, low turbidity lake water with low alkalinity. Coagulation followed by ultrafiltration showed a reduction of 58 % in TOC, which was 19 % higher than what conventional coagulation process was able to achieve with same coagulant dosage. When PAC was added, THM-content was reduced significantly and a clear reduction in the decline of specific flux was observed. Fabris et al. (2007) showed that MIEX combined with coagulation was able to significantly reduce PVDF MF membrane fouling and improve permeate quality, especially the removal of DOC was good, approximately 76 %. If PAC was also added, the water quality and fouling tendency where further improved.

Adsorbents alone, however, might not be an efficient pretreatment for membrane processes. Fabris et al. (2007) showed that MIEX-treatment without any additional treatment steps before membrane filtration actually worsened the fouling of the membrane.

No improvement in the amount of reversible fouling was discovered either. Permeate quality did, however, improve. Researchers found that MIEEX was unable to effectively remove high MW colloidal NOM. When coagulation was added, all colloidal NOM was removed.

Mozia et al. (2006) found similar problems with PAC. With or without ozonisation pretreatment, when PAC was added to the treatment process of humic acid-rich feed water there was a significant drop in permeate flux. Lin et al. (2000) experienced clear reductions in filterability of different NOM fractions when the water was pre-treated with PAC. They investigated the effect of PAC pretreatment in removing different NOM fractions and concluded that PAC pretreatment always resulted in flux decline. Filterability of a direct PAC-UF-system was not affected by the addition of PAC. Carroll et al. (2000) and Mozia and Tomaszewska (2004) showed that when filtering natural water the membrane permeability was not affected by the addition of PAC, but the removal of organic substances was enhanced. Xia et al. (2007) noticed that while PAC addition enhanced organic matter removal, with long backwash intervals the initial flux could not be recovered when high PAC dose was added. With PAC dose of 10 mg/l and 60 minutes backwash interval the flux after backwashing was 89 l/m<sup>2</sup>h, but with PAC dose of 50 mg/l the flux after backwashing was only 81 l/m<sup>2</sup>h when similar interval was used. Pure water flux was 90 l/m<sup>2</sup>h, which means that a higher dosage resulted in a 10 % decline in permeability.

Different types of adsorbents have been synthesised for fouling control, e.g. heated aluminium oxide particles, heated iron oxide particles, PS colloids, surfactant modified powdered activated carbon, and carbon black, but these are all still used exclusively at pilot studies (Huang et al., 2006; Gao et al., 2011). Thus it can be concluded that whilst traditional adsorbents are incapable to significantly enhance filtration process alone, when combined together and/or with coagulation they offer a good option for treating waters with high NOM. When these previously mentioned synthesised adsorbents are commercially available, adsorbents alone might be feasible as a LPM filtration pretreatment.

## **5.4 Preoxidation**

Oxidants can suppress microbial growth, change NOM composition by increasing the amount of AOC, and provide disinfection for the water. Typical oxidants that are used

in water treatment processes are ozone, permanganate, and chlorine (Huang et al., 2009; Gao et al., 2011).

Ozone either reacts directly with dissolved substances, or it may decompose into radical species, which themselves react with NOM substances. Ozonisation does not change the amount of DOC under economically acceptable conditions (van Geluwe et al., 2011). However, it breaks down the organic matter and increases the amount of AOC. AOC can easily be removed by downstream biological filters. When ozonisation is used as a pretreatment for membrane filtration,  $UV_{254}$  and TOC reduction increases while the drop in permeate flux decreases (Mozia et al., 2006). Byun et al. (2011) studied the effect of ozone dosage prior to membrane filtration and found that for UF membrane the DOC rejection lowered during filtration, on the contrary NF membrane performed better in the final stages of filtration. At higher ozone dosage the DOC rejection was higher; this was credited to the higher polarity of ozonated NOM.

Ozonisation is effective in decreasing the amount of hydrophobic NOM in the water. But when ozonisation is used with natural waters some problems might occur. The problems related to divalent inorganic cations, such as calcium and magnesium, forming fouling layers have been found to be more severe when ozonisation is used as pretreatment (van Geluwe et al., 2011). Furthermore, highly alkaline waters contain bromine (Br) which reacts to free ozone in water after ozonisation. If the residence time inside the membrane reactor is long the dissolved ozone has time to produce carcinogens like bromate ( $BrO_3$ ) (Schlichter et al., 2004; Moslemi et al., 2011; van Geluwe et al., 2011). Schlichter et al., however, proved that for  $BrO_3$  to be clearly observed after ozonisation the initial concentration of Br has to be above 100  $\mu\text{g/l}$ .

van Geluwe et al. (2011) summarized the effects that ozonisation has on NOM and the benefits that it has on membrane filtration. The most important changes were the removal of aromatic rings, which decreases the adsorption of NOM on membrane surfaces, increase in the number of carboxylic functions repelled by negatively charged membrane surfaces, and decomposition of NOM molecules into smaller fragments. To enhance the performance of ozonisation  $H_2O_2$  has been added to the process. This should increase the contribution of OH radicals to DOC removal, but van Geluwe et al. (2011) found this increase to be very small.

The biggest drawback that ozonisation has as a pretreatment is incompatibility with most polymeric membranes. Most of the studies where ozonisation has been used have thus been conducted with ceramic membranes (Huang et al., 2009; Gao et al., 2011; van Geluwe et al., 2011). Schlichter et al. (2004) studied a hybrid ozonisation – membrane filtration – active carbon filtration using ceramic MF and UF membranes. When the residual ozone after ozonisation for UF and MF was above 0.05 mg/l and 0.1 mg/l, respectively, no drop in the permeate flux was observed, and thus no regular backwashing or chemical cleaning was necessary. Furthermore, the quality of permeate was constant and below German Drinking Water Guidelines. If the ozonisation was stopped the permeate flux started declining, but was recoverable after the ozone was reintroduced. Recoveries of 99.5 % were easily achieved.

Permanganate and chlorine are widely used as pretreatment for conventional treatment. Despite this, very little research has been reported on the usage of said pre-oxidants as a pretreatment for LPM filtration. Chlorine has been used in conventional treatment to prevent biological growth in the feed water but Baker and Dudley (1998) found chlorine as a pretreatment for LPMs problematic as it might have an adverse effect and actually worsen the biofouling potential. Chae et al. (2008) found that chlorine enhanced the fouling through the precipitation of iron and manganese on the membrane.

## **5.5 Prefiltration**

Prefiltration by using bed filters or other membranes could be used to remove particles detrimental to the performance of the primary LPM. Primary mechanism for removal is physical sieving but chemical adsorption and deposition might also occur (Huang et al., 2009). As can be seen from appendix 1, strainers with a pore size much bigger than membrane can be used to reduce solid loads on the filtration process. Other prefiltration treatments that have been added before membrane filtration include media filters, like sand and limestone filters, and membranes with a coarser pore size.

As previously has been stated, hydrophobic membranes are effective in chemically adsorbing organic foulants and can thus be used as prefiltration step before primary membrane. MF and UF membranes have been used as pretreatment for reverse osmosis process. If membranes are used as pretreatment cost-effective measures need be developed to restore the removal capacity of these pretreatment membranes (Huang et al., 2009).

Granular media filters have been proven to effectively remove particles from water. Removal of large and medium-sized particles might reduce internal fouling (Huang et al., 2009). Xia et al. (2004), however, found sand filtration prior to UF filtration to increase the drop in permeability due to removal of large particles. Without sand filtration the large particles formed a permeable cake layer on the membrane surface which could be easily removed. The cake layer also adsorbed smaller particles thus preventing them from blocking the membrane pores.

At Nonnweiler WTP in Germany UF has limestone filtration without coagulation as pretreatment (Lipp et al., 1997). Limestone is used to remove manganese from the water and also to raise the pH. The usage of limestone filter has not been found to affect the performance of the UF, positively or negatively, especially when the raise in pH is controlled.



## **6 Pilot study at HSY Pitkääkoski water treatment plant**

This chapter presents the premise for the pilot conducted at Pitkääkoski WTP. First reasons why retrofitting was of interest are discussed. Then the pilot equipment is presented, along with methods that were used to evaluate the performance of membrane filtration.

### **6.1 Introduction**

One part of this master thesis was to study the possibility of retrofitting low pressure membranes into Helsinki Region Environmental Services Authority's (HSY) Pitkääkoski WTP treatment process. There are two main reasons why the performance of membrane filtration was of interest. First reason is the increasing amount of NOM in raw water. Despite the fact that considerable measures have been done to improve the water quality at Lake Päijänne both TOC and  $\text{KMnO}_4$  have risen after 2008, from 6.0...7.5 mg/l and 20...26 mg/l, respectively.

Increase in NOM has been noted around the Nordic countries and has been credited to climate change (Dahlberg, 2012; Eikebrokk, 2012; Keucken et al., 2012). According to Dahlberg this trend has been continuous for the last 35 years. Eikebrokk has explained the complex influence that climate change has on NOM-levels in surface water. On one hand NOM production has increased due to the increased photosynthesis and primary production, which in turn is due to the increase in temperature. On the other hand NOM flux has increased due to increased amount of rainfall. These trends are estimated to continue in the future. The improvement of analytical methods might have affected the measured NOM in surface water, and thus climate change is not the only reason.

NOM is not of direct concern in potable water. However, it increases the demand of disinfectant in post-treatment and coagulant during coagulation, forms complexes with heavy metals and organic micro-pollutants, reacts with chlorine and forms THMs, enhances microorganism growth in distribution system, and gives the water an unpleasant colour (Kabsch-Korbutowicz, 2005a; Ødegaard, 2006). Furthermore, LPMs are vulnerable against high NOM loads, which cause fouling of the membrane.

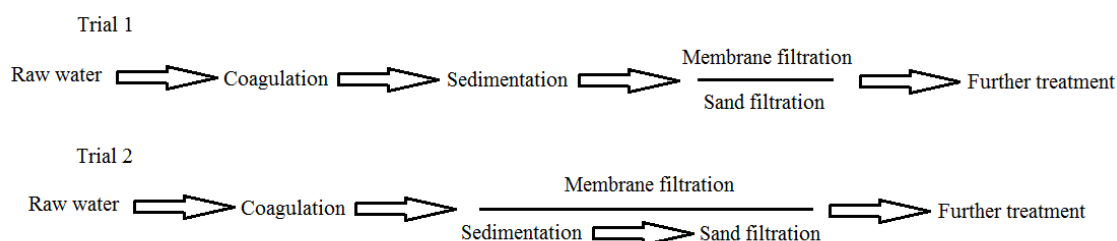
Another reason for retrofitting is directly related to the increase of NOM. Increased amounts of NOM causes the need to increase treatment efficiency. Increasing the efficiency with current conventional treatment is possible to a certain point; however, physical barriers such as sedimentation tank size will be limiting the increase in the

future. Increasing the surface load past the physical barrier of conventional treatment will result in poor water quality. Increasing the capacity by building new treatment lines is expensive and requires large areas of land which no longer are available for Pitkääkoski WTP. LPM filtration is a compact alternative for conventional treatment, it produces good quality water despite raw water conditions, and increasing capacity is easy because of modular structure and low footprint of membranes.

To evaluate if LPMs could be used at Pitkääkoski, and which treatment steps could be replaced, literature on existing retrofits was studied. In appendix 3 is data collected about raw water quality in WTPs that have retrofitted LPMs. This data was compared to raw water quality at Pitkääkoski WTP which can be described as low alkalinity (0.35 mmol), low hardness (1.2 °dH), and low turbidity (0.4 FTU) but high NOM.

Raw water at Pitkääkoski is treated by conventional treatment followed by further treatment. First treatment step is coagulation with ferric sulphate. Directly after coagulant is added, a rapid mixing with a mixer is done to enhance the even mixing of coagulant. Then water flows to flocculation chambers where the floc is formed. The chambers are in series, and the rotation speed slows down from one chamber to another moving towards sedimentation tanks. Sedimentation happens in two-layer horizontal sedimentation tanks. After sedimentation the remaining floc is removed with rapid sand filtration and then water is led to further treatment.

In appendix 1 is data collected about treatment processes in different WTPs using LPM filtration. Based on this data, two possibilities to retrofit membranes were found. These are presented in Figure 13. Replacing sand filtration was tested in trial 1 as the feed water for the membrane after sedimentation was estimated to be optimal. Sedimentation has lowered the amount of TOC to 60 % of the raw water TOC and thus fouling caused by NOM was estimated to be smaller. Another possibility was replacing both sedimentation and sand filtration with membrane filtration; this was tested in trial 2.



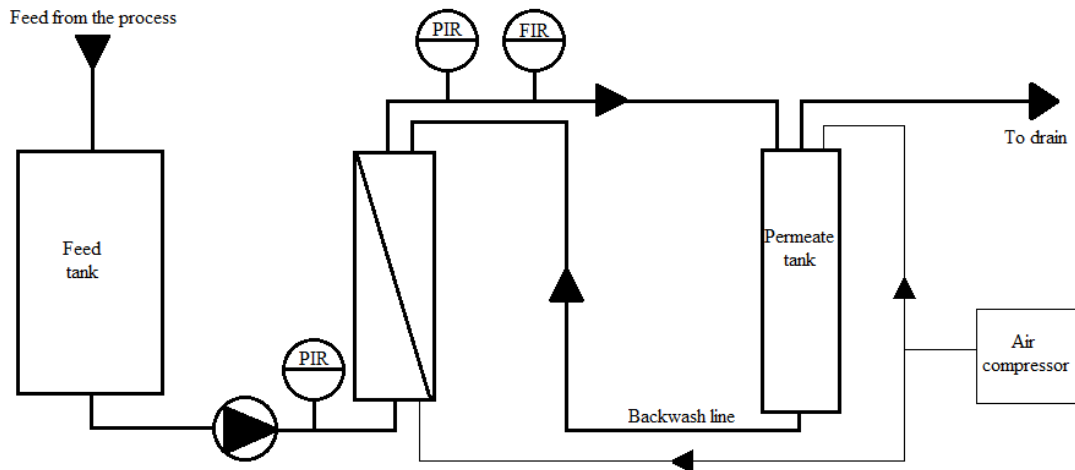
**Figure 13. Two identified possibilities for retrofitting parts of conventional treatment at Pitkääkoski WTP. Further treatment consists of ozonisation, GAC-filtering and UV treatment.**

## 6.2 Material and methods for the pilot

### 6.2.1 Pilot equipment

Hollow fibre MF membrane from Pall/Asahi in a UNP-1003 module by Microza was used. Membrane was a hydrophobic PVDF membrane with modified hydrophilic surface characters and the module housed 140 fibres, altogether the membrane surface area was 0.26 m<sup>2</sup>. Membrane pore size was 0.1 µm. The inner and outer diameters of one fibre were 0.7 mm and 1.4 mm, respectively. The filtration process was a dead-end mode filtration with an outside-in flow direction.

Experiments were conducted in a constant pressure configuration in a LPM system designed by Pall. The system had a Grunfos pump which provided the constant pressure. Pressures were measured from the feed and permeate side with a pressure transducer by Endress+Hauser. Flow through the membrane was measured from the permeate side with a magnetic flow meter by Kobold. Backwashing was performed with permeate by changing the direction of flow with compressed air; simultaneously an air scour to the feed side of the membrane was performed. Air was compressed with an air compressor Pioneer 402 by Prebena. A simplified MF system is presented in Figure 14. The backwash sequence, duration and interval, was controlled through the programmable logic controller (PLC). Feed tank was automatically filled at predetermined intervals and controlled through the PLC.



**Figure 14. Schematic of the pilot. Pressure was created with a pump, while backwashing was conducted using compressed air both for air scour and to push permeate as reverse flow.**

Backwashing was supported with a CEB every other day. The chemicals that were used for cleaning were 0.5 % citric acid and a mixture of 0.5 % NaOH and 1000 ppm NaClO. These chemicals were recommended by the equipment supplier. Based on results by

Zondervan and Roffel (2007) also NaClO alone was tested. For CEB the normal operation was stopped, the module was drained and potable water with cleaning chemicals was added. Then chemically enhanced washwater was filtered through the membrane for 30 minutes and during this time three backwash sequences were executed. As hypochlorite reacts with acids and forms chlorine gas, acid cleaning was always performed separately from the mixture cleaning. After each chemical cleaning the module was thoroughly rinsed with potable water to eliminate residual chemicals before normal filtration was continued.

### 6.2.2 Membrane performance analysis

The membrane filtration was evaluated by following both membrane performance and water quality. To estimate the filtration performance permeability, as presented in equation 6, was used. Temperature of the feed water, filtration flow, and TMP of the membrane were automatically measured and recorded to a memory card in the PLC. Viscosity of water was calculated with equation 4. The performance of backwash was estimated visually. The water consumed for the backwash process was reduced as the membrane pores became blocked and the nature of reduction was estimated to correlate with the reduction of permeability.

From the filtration flow recorded by PLC the total volume of filtered water can be calculated. Total volume of filtered water, however, included also the water that was used for backwashing. Thus the total volume of produced water,  $V_P$ , was calculated by subtracting water lost because of downtime from total volume of filtered water. Equation for calculating  $V_P$  is

$$V_P = Q_a t_o - \left( V_b t_o + Q_a \frac{t_b}{60} \right) \quad (9)$$

where  $V_P$  = volume of produced water [l],  
 $Q_a$  = average filtration flow [l/h],  
 $t_o$  = time of operation [h],  
 $V_b$  = volume of water consumed by backwashing [l], and  
 $t_b$  = time used to conduct backwashes [h].

To calculate the recovery percentage of the filtration process,  $V_P$  was divided with the average feed flow to the membrane,  $Q_a$ . Downtime of the LMP system means time consumed either to conduct backwashing or to produce water required for backwashing.

The time consumed to produce water for chemical cleanings was calculated by dividing average backwash flow with average filtration flow and multiplying the division with operational hours.

Performance of the membrane was also estimated economically. Energy consumptions of the module, including pump and PLC, and air compressor supplying the backwash pressure were measured with an off the shelf electric supply meter by KML. The energy consumed during each filtration test was divided by  $V_p$  and thus the energy consumption of each test could be compared in kWh/m<sup>3</sup>. The energy calculations should not be considered as the absolute value, but to show the differences between different operating conditions.

Water quality of membrane filtration was compared to sand filtered water. Sand filter 16 (SF16) at the end of treatment line 8 was chosen because it has been used in other pilots as a reference and thus the performance of the filter was known and changes in water quality could easily be explained. Also the treatment line from which feed water for the membrane was taken does not have a sampling point after sand filtration. Thus samples were taken from the clarified or flocculated water from treatment line 1 which were used as feed waters for the membrane, from permeate, and from sand filtered water. The retention time after the coagulant is added is different between treatment lines used, which might affect the treatment results.

Several water quality analyses were conducted. The feed water quality was assumed to be the same for both membrane filtration and conventional filtration, although different treatment lines were used for these processes. This means that feed water for conventional treatment was not analysed, but the feed water for membrane also represented feed water for conventional treatment. The reduction of substances in membrane filtration and conventional treatment was calculated with the equation

$$S = 1 - \frac{C_p}{C_f} \quad (10)$$

where  $S$  = reduction [%],

$C_p$  = concentration of substances in permeate [g/mole], and

$C_f$  = concentration of substances in feed water [g/mole].

The pH-value of the water is crucial in the process as it consumes concrete structures of the WTP. Alkalinity and pH were hypothesised not be affected by the membrane

treatment, but these were analysed for safekeeping. Membranes have a very good removal efficiency regarding particles and to conceive how the permeate quality would compare to conventionally treated water turbidity and residual iron were measured. The removal of NOM was especially interesting and it was studied extensively. TOC, DOC, and AOC were measured to understand the removal efficiency of the membrane while UV<sub>254</sub> absorbance, LC-OCD (liquid chromatography – organic carbon detector), and HPSEC (high performance size-exclusion chromatography) analyses were conducted to deepen the understanding of which fractions of NOM were effected by membrane treatment. As NOM removal in MF filtration is dependent on coagulation conditions, the quality of permeate and sand filtered water was expected to be the same. To study the microbiological quality of the permeate R2A (Reasoner's 2A) agar and ATP (adenosine triphosphate) analyses were conducted. Microbiological quality of permeate was expected to be excellent.

pH was measured according to standard SFS 3021:1979. Depart from standard pH was measured by using an electronic pH meter equipped with a glass electrode by Metrohm having a filling solution of 3 mol/l calcium chloride. The results are reported to two decimal places.

Turbidity measurement was done according to standard SFS-EN 27027:2000 by using a turbidity meter HACH 2100AN. The meter measures either scattered or adsorbed light ranging between 0...1000 FNU (formazin nephelometric unit). FNU value is equivalent to FTU (formazin turbidity unit). The results are reported by two decimal places when the turbidity is below 1 FTU and by one decimal place when turbidity is between 1.0...9.9 FTU. The uncertainty of measurement is 15 %.

UV absorbance was measured according to a guideline presented in Standard Methods for Examination of Water and Wastewater (Greenberg et al., 1995). UV absorbance is measured with a spectrophotometer at a wavelength of 254 nm. The result indicated the amount of substances absorbing UV light at fixed wavelength 254 nm (UV<sub>254</sub>). Centrifuge was used to pre-treat the flocculated water before measuring UV absorbance, as the floc might have damaged the spectrophotometer. This has resulted in lower UV<sub>254</sub> for flocculated samples compared to clarified samples.

TOC analysis was done according to standard SFS-EN 1484:1997 by using TOC analyser VCPH-TOC by Shimadzu. The organic carbon is oxidised into carbon dioxide

by burning the sample in 680 °C. The results are presented by one decimal place at concentrations between 0...10 mg/l. The uncertainty of measurement at concentrations between 0.4...6.0 mg/l and 6.0...10.0 mg/l is 20% and 11%, respectively.

DOC analysis was also done according to standard SFS-EN 1484:1997 by using TOC analyser VCPH-TOC by Schimadzu. First the sample was filtered to remove particular organic carbon. Filtration happened through a 0.45 µm PES-filter which had been flushed with ultrapure water. Thus DOC in this experiment is the fraction of TOC smaller than 0.45 µm. After filtration the course of analysis was similar to TOC analysis.

AOC analysis was done by the National Institute for Health and Welfare according to a modification of standard presented in Standard Methods for Examination of Water and Wastewater (Greenberg et al., 1995). Samples were taken at Pitkääkoski WTP and sent to Kuopio where they were analysed one day after the sample had been taken except the one which was analysed two days after sample was taken. 100 µl of nutrient solution was added to the samples to maintain nutrient balance. The samples were first sterilized at 60 °C for 30 minutes. Two bacterial strains were added to two parallel samples. First was *Pseudomonas fluorescens* P17 and the second was *Aquaspirillum* NOX. The growth was observed with R2A-agar culture. The amount of AOC was determined by the number of colonies produced by the sample water. Flocculated water was not analysed as high content of iron would have disturbed the results significantly.

Alkalinity measurement was done according to standard ISO 9963-1:1996. The results present the total alkalinity of the sample, including hydrocarbonate-, carbonate-, and hydroxide concentration. Measurement is done with a titrator Ttruno 794 by Metrohm by potentiometric titration of 100 ml sample with 0.02 mol/l hydrochloric acid to pH 4.5. The results are reported with two decimal places. The uncertainty of measurement is 28 % when alkalinity is less than 0.3 mmol/l.

Total amount of residual iron was measured according to standard SFS 3028:1976. First 0.248...0.254 g of potassium peroxodisulphate was added to 100 ml of sample. Then 25 ml of the mixture was oxidised in an autoclave for 30 minutes. After autoclave 2 ml of hydroxylammonium chloride, TPTZ-solution, and sodium acetate were added, in this order, to the samples. Then the residual iron was measured with a PerkinElmer

spectral photometer at wavelength of 593 nm. The uncertainty of measurement is 20 % when iron is in the range of 20...200 mg/l and 6 % within the range of 200...1500 mg/l.

HPSEC samples were taken at Pitkääkoski WTP and analysed at MetropoliLab. Samples were filtered through a 0.45 µm PES-filter which had been flushed with ultrapure water, and frozen before analysis. A high performance liquid chromatograph (HPLC-1100) by HP was used together with BioSep-columns (BioSep-SEC-s3000 7.8 × 300 mm). DAD-detector was used at wavelength 254 nm. The uncertainty of measurement is 20 %.

LC-OCD samples were taken at Pitkääkoski and analyses were done at Kemira's Espoo Research Centre according to method presented by Huber et al. (2011). First samples were filtered through a 0.45 µm PES-filter and separated in a chromatograph column (250mm × 20 mm, TSK HW 50S). Separation was followed by a UV-detector at a fixed wavelength of 254 nm and an organic carbon detector (OCD). At the inlet of the OCD the samples is acidified to convert carbonates into carbonic acid. Data from UV-detector and OCD is processed with a computer. DOC is divided into hydrophobic and hydrophilic fractions and hydrophilic fraction is further fractionated to biopolymers, humics, building blocks, and low molecule weight acids and neutrals. SUVA value of different samples is also calculated.

Microbiological analyses were done with R2A agar cultivation according to Greenberg et al. (1995) during two weeks of testing, one week for each phase. Sample volume of 0.1 ml was cultivated on a Petri dish and incubated for 7 days in  $22 \pm 2$  °C. After 7 days of cultivation the number of colonies was calculated. The results are presented in colonies per ml [pmy/ml]. If small colonies form in a tight formation the amount of total colonies might be difficult to estimate.

ATP was analysed at Kemira's Espoo Research Centre. Test was a 2<sup>nd</sup> generation ATP measurements and it was done with LumiKem water test kit by Kemira. First the microorganisms were concentrated by pushing sample through a filter. Then ATP was extracted from the filter with KemiLyze extraction reagent and measured. ATP is a molecule found in and around living cells which reacts with luciferase and produces light. This light is directly proportional to the amount of living organisms in the sample.



### 6.2.3 Filtration tests

The pilot experiment was carried out at Pitkääkoski WTP facilities and two different feed waters were used. For the first trial period the feed water to the membrane was taken from after the sedimentation process, at that point the TOC-level of the water has been significantly reduced and the water quality was expected to be better for LPM filtration. First a steady-state filtration phase for three weeks took place followed by another three weeks of optimization phase, where the driving parameters of the filtration were changed in order to find the most optimal conditions. During the second trial flocculated water was used as feed water. Schedule for the pilot is presented in Table 9. Membrane characterization was performed in the beginning of each trial and phase to ensure the integrity of the membrane.

**Table 9. Piloting schedule. Clarified water after sedimentation was hypothesised to work better and thus an optimisation phase was conducted with clarified water.**

Schedule	Duration	Date
Trial 1, Phase 1: Clarified water	3 weeks	3.12.2012-23.12.2012
Trial 1, Phase 2: Optimisation	3 weeks	24.12.2012-13.1.2013
Trial 2: Flocculated water	2 weeks	14.1.2013-27.1.2013

During the first trial the operating conditions remained constant. The feed water was filtrated through the membrane with a constant 0.9 bar pressure at an initial flux of 100 l/m<sup>2</sup>h. Backwash interval was 20 minutes and the backwash flux was approximately 1000 l/m<sup>2</sup>h for 20 seconds. Clarified water during trial 1 was taken from the collection channel after sedimentation lines 1 and 2, and piped to the module by gravitation. It can be assumed that no sedimentation happened in the collection chamber after the sedimentation pond or in the pipe leading from the chamber to the feed tank of the module; however, sedimentation took place in the feed tank.

After phase 1 of trial 1, optimisation phase was conducted in order to find optimised conditions to run the module. Eight different operating conditions were used, each test lasted for two days (41.5...48 hours). The conditions that were changed were flux through the membrane, backwash interval, and backwash flux (Table 10).

**Table 10. Optimisation tests. Each test lasted for two days. After two days the membrane was chemically cleaned and rinsed before the next test.**

Condition	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6	Test 7	Test 8
Flux (l/m <sup>2</sup> h)	90	90	90	90	125	125	125	125
BW interval (min)	20	30	20	30	20	30	20	30
BW flux (l/m <sup>2</sup> h)	1000	1000	1250	1250	1000	1000	1250	1250

Second trial period with flocculated water was also a fixed conditions phase. Operating conditions were similar to the fixed conditions phase in the first trial: initial flux at 0.9 bar constant pressure was 100 l/m<sup>2</sup>h, backwash interval was 20 minutes and backwash flux was 1000 l/m<sup>2</sup>h. During the second trial period the feed water was taken from the last flocculation chamber before sedimentation in treatment line 1. Again the water was piped to the module by gravitation. No sedimentation took place in the flocculation pond or in the pipe leading to the module but due to the weak structure of the floc it can be assumed that some of the biggest flocs came apart and resulted in smaller flocs than flocs in sedimentation. Sedimentation did take place in the feed tank of the module but as the feed water from the tank was taken from the bottom, it can be assumed that this sedimentation was not a severe factor.

## 7 Results from the pilot

In this chapter the results of the pilot experiment conducted during this thesis are presented. First the performance of the pilot is discussed and then findings related to capacity increase are presented. Lastly results from water quality analyses are presented. The discussion of the results takes place in chapter 8.

### 7.1 Pilot performance

#### 7.1.1 Membrane permeability

The experiment started with a fixed conditions phase where clarified water after sedimentation was used as feed water. During the first two weeks membrane filtration faced severe problems related to operational conditions. Accumulation of iron flocs on membrane surface was visually observed. The reason for this was inefficient air scour during backwashing. After air scour was fixed, the accumulation of iron flocs was eradicated. However, this meant that during trial 1 only few days of operation with fixed conditions could be conducted.

Permeability and TMP of two periods with fixed conditions from trial 1 are presented in Figure 15. The initial permeability in both experiments was above  $1300 \text{ l/m}^2 \cdot \text{h} \cdot \text{bar}$  but within the first hour of operation it dropped rapidly to approximately  $1000 \text{ l/m}^2 \cdot \text{h} \cdot \text{bar}$  and thereafter followed the progress shown in Figure 15. Duration of both filtration tests was 46 hours and during that time permeability dropped to 70 % of the original permeability.

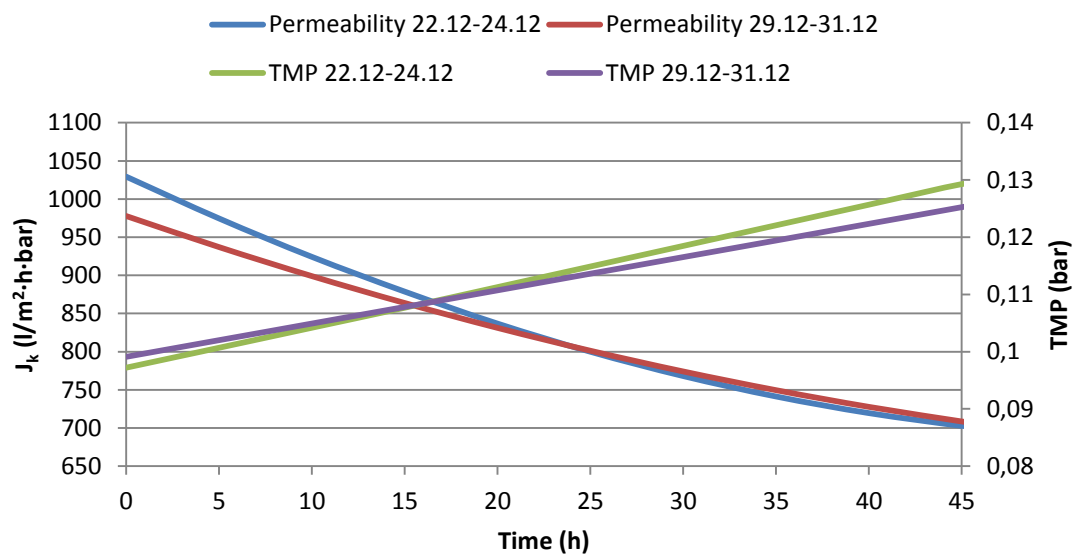
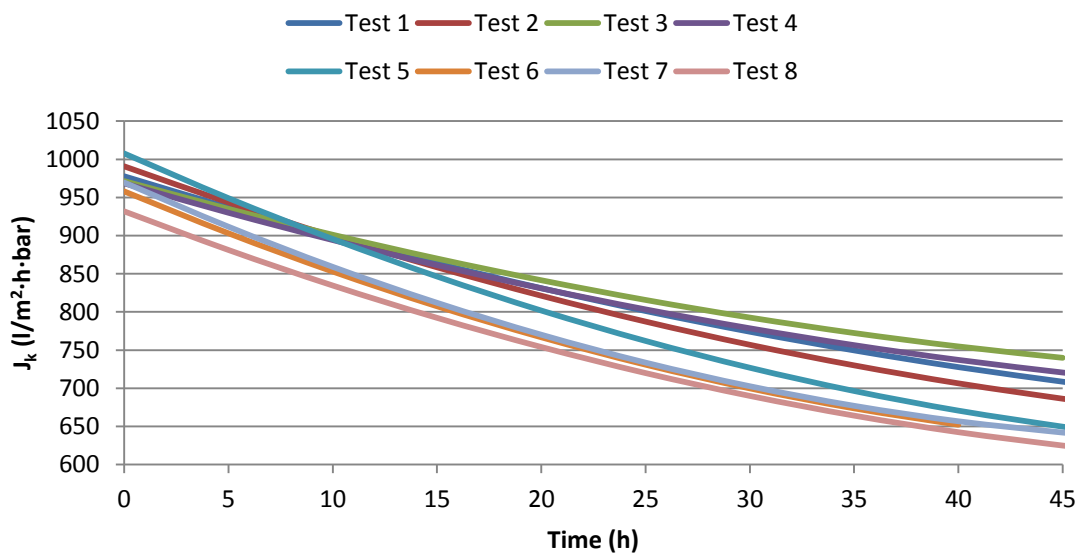


Figure 15. Permeability and TMP from fixed conditions phase in trial 1. After 46 hours the permeability in both periods is the same.

Increase in TMP during filtration tests is also shown in Figure 15. In both tests the increase in TMP was approximately 0.03 bar in 46 hours. During the experiment TMP increased and flux decreased which is different to usual operation of LPMs. Operational WTPs using membranes operate with either constant pressure or constant flux, and thus the operation here is a bit different.

When the operational performance with fixed conditions was acquired, the optimisation phase started. Operating conditions are shown in Table 10. Eight different operating conditions were tested according to Table 10 and the effect on permeability of different conditions is shown in Figure 16. Several observations can be made from Figure 16.



**Figure 16. Permeability with different operating conditions.**

First observation is related to flux through the membrane. In tests 5...8, when the average flux was higher, the drop in permeability was faster. After two days the permeability was only 66.6...69.0 % of the original permeability, whereas during tests 1...4 permeability dropped only to 71.0...77.0 % of the original. This is intuitive, higher amount of treated water means higher amount of treated impurities and thus more impurities can block membrane pores.

Second observation is that with a higher backwash flux, a better permeability after 46 hours of operation is achieved. When the flux through the membrane was 90 l/m<sup>2</sup>h, with a higher backwash pressure the final permeability was 3...3.5 % higher compared to lower backwash flux. When the flux through membrane was 125 l/m<sup>2</sup>h, however, no similar trend was observed as the final permeability was almost identical in each test.

This would indicate that small particles blocking the membrane pores require more efficient backwashing to be effectively removed.

As a third observation it can be concluded that with a longer backwash interval, a lower final permeability is achieved. The difference, however, is small, only 1.0...3.5 %. This is intuitive as well; longer backwash interval allows more impurities to accumulate in/on the membrane surface between backwashes.

The drop in produced water, however, is not similar to the drop in permeability. Tests 1 and 5 experienced highest declines in flow through membrane. The flow through membrane after 46 hours was 79.0 % and 76.6 % of original flow, respectively. During other tests the flow was 81.1...85.9 % of the original. This observation is in line with Leiknes et al. (2004); backwash interval shorter than 30 minutes is unfavourable regarding water production. However, shorter backwash interval kept TMP increase lower, thus permeability remained similar compared to other tests with similar flux.

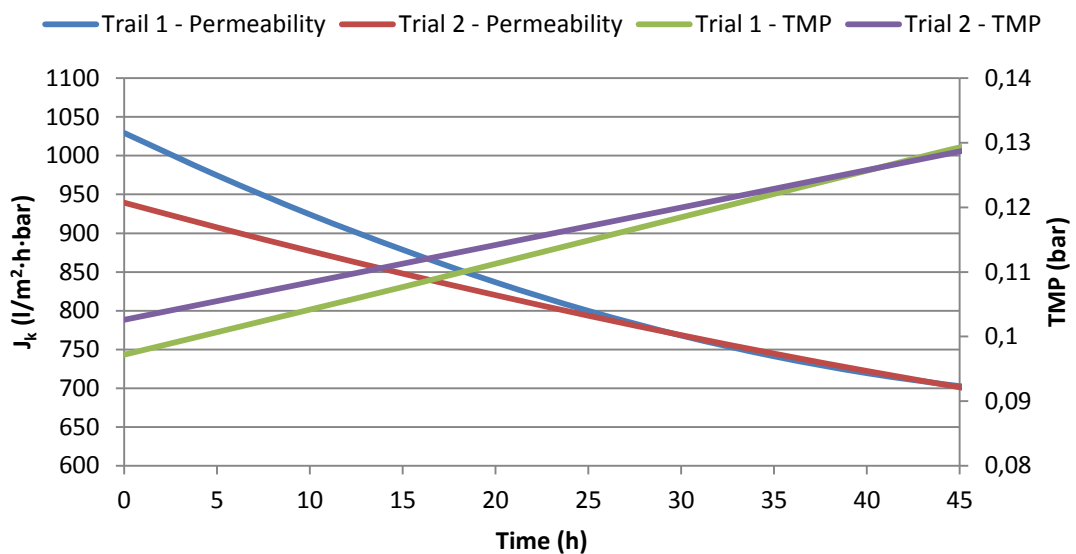
If only permeability is focused on, conditions during test 3 were optimal. The loss of permeability was 23 %. These conditions, however, are not optimal regarding water production or energy consumption. The average amount of treated water in tests 1...4 was 15.3 l/h, whereas in tests 5...8 the average flow through membrane was 21.2 l/h. Table 11 presents recovery, downtime, total volume of produced water, and energy consumption during each test. Operational time of tests varied between 41.5...49 hours. However, the results presented in Table 11 have been calculated according to 46 hour operation. Chemical consumption and time to conduct CEB can be estimated to be similar in all tests and they are thus excluded from the evaluation. The amount of chemicals used during trials is presented in chapter 6.2.1. The amount of water used for CEB was 1.4...2.4 % of total produced water.

**Table 11. Results from optimisation phase during trial 1. Results indicate that operating conditions during test 8 were optimal regarding water production and energy consumption.**

Test	Recovery (%)	Downtime (h)	Water produced (l/h)	Energy (kWh/m <sup>3</sup> )
1	72.5	13.4	10.8	11.0
2	81.2	9.1	12.2	10.1
3	73.3	12.8	10.9	11.5
4	81.8	8.8	12.5	10.1
5	80.0	10.0	16.4	8.1
6	87.3	6.4	18.3	7.2
7	81.2	9.2	16.9	7.8
8	87.8	6.0	18.5	7.2

Results presented in Table 11 show that the most optimal conditions for water production would be conditions in test 8. It should be remembered that these results are based on the amount of produced water. With a higher flux the wear of membrane is also higher, which might result in a shortened membrane lifetime. A longer operational period is needed to further estimate the utility of these conditions.

After the optimisation tests, the feed water was changed to flocculated water. Two weeks of operation were scheduled, but due to malfunction of PLC data from only one week was acquired. The operational data from trial 2 was compared to data from trial 1, shown in Figure 17. With flocculated water the initial drop in permeability was bigger, which was probably due to higher feed water turbidity. Similarly increase in TMP in the beginning was higher. However, the decline in permeability and increase in TMP were slower than with clarified water in trial 1, and already after two days of operation the permeability was the same for both feed waters, meaning that the rate of fouling with flocculated water was less than with clarified water.



**Figure 17. Comparison of permeability between trial 1 and trial 2. Initially the permeability during trial 1 is higher, but the fouling rate with flocculated water is less than with clarified water.**

Although permeability with flocculated water was worse compared to clarified water, the total amount of produced water is only slightly less, 10.8 l/h and 11.1 l/h, respectively. This difference can be explained by the differences with TMP. The initial difference in permeability is mainly due to increased TMP, whereas flow through the membrane is similar in both cases. As the difference in TMP is reduced so is the difference in permeability. As operating conditions, apart from feed water, were similar the required energy for m<sup>3</sup> of produced water was only slightly higher.

### 7.1.2 Effect of different cleaning chemicals

Three different chemical treatments were executed. The effect of each chemical treatment is presented in appendix 4. CEB with NaClO failed to show any improvement to the performance, contrary to what was suggested by literature. There was a slight improvement in permeability but the effect was brief. Treatment was done with 7 °C water, temperature was selected so that it was close to the temperature of permeate. However, the cold temperature might have affected the efficiency of this treatment. After treatment with NaClO, cleaning with citric acid was tried in three consecutive days with 7 °C water. With citric acid a slight improvement was always observed, however original permeability was never reached. Finally a combination of first citric acid and then mixture of NaOH and NaClO was tested and original permeability was achieved. These chemicals were used at higher temperature of 25 °C. These two CEBs were also done in a reverse order, NaOH and NaClO first and citric acid later, but no difference was observed in resulting permeability as shown in Table 12. All aforementioned chemical treatments lasted 30 minutes.

**Table 12. Effect of CEB with different treatments. In test 1 citric acid was used in first CEB and NaOH and NaClO in the second CEB, in test 2 the order is in reverse.**

Test	Before CEB		After 1 <sup>st</sup> CEB		After 2 <sup>nd</sup> CEB	
	TMP (bar)	Flux (l/m <sup>2</sup> h)	TMP (bar)	Flux (l/m <sup>2</sup> h)	TMP (bar)	Flux (l/m <sup>2</sup> h)
1	0,204	22,75	0,175	29,17	0,126	31,92
2	0,233	23,66	0,179	29,08	0,125	32,46
Original permeability					0,12	33,3

Using multiple chemicals for chemical treatment is not uncommon, as can be seen from appendix 1. At St. Saviours WTP two different chemicals are used to chemically clean the membranes: hypochlorite is used for organic substances and citric acid for inorganic substances (Redhead, 2008).

Yamamura et al. (2007) found that for a hydrophobic PVDF membrane the fouling happened in two stages. First the small molecules of composed mainly of humic substances adsorbed into the membrane pores and on the membrane surface. Secondly large molecules plug the pores and cause physically irreversible fouling. Humic substances can also bind larger substances to the membrane surface. Similar fouling can be hypothesised to have happened in this experiment. The small substances, consisting of NOM and small iron colloids adsorb to the membrane pores and the larger molecules start to accumulate on the existing particles creating physically irreversible fouling. Thus both caustic and acidic chemicals are needed to regain original permeability.

The duration of CEB at the beginning of the whole experiment was 30 minutes, however, during the experiment the duration was shortened to 12 minutes. Still, during that duration three backwash sequences were executed. The shorter CEB was observed to be as effective as the longer one. It was also observed that the effect of chemical cleanings was immediate. The permeability increased significantly during the first backwash sequence and subsequent cleaning cycles failed to improve the permeability. This would indicate that a simple CEB, where chemicals are added to backwash water, might be enough to remove foulants and regain original membrane permeability.

## **7.2 Water quality**

### **7.2.1 Observations during the experiment**

During the experiment, temperature of raw water coming to Pitkääkoski WTP declined from 4 °C to 2 °C. Despite of this, the average feed water temperature for membrane remained almost constant, approximately 4.4 °C. This was due to the ambient temperature of the space where the membrane module was placed. Thus the viscosity, and therefore resistance, stayed the same during each filtration test. However, the drop in temperature has caused variation in the performance of coagulation and SF16, as shown later. Otherwise the quality of the raw water remained the same.

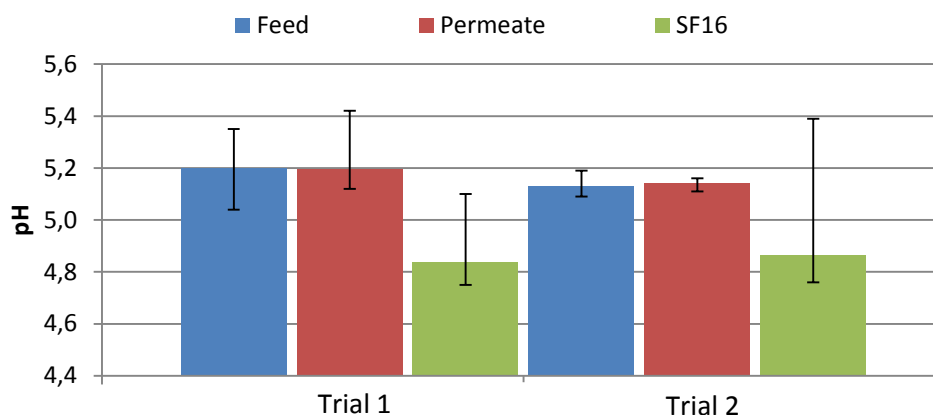
Despite the changes in operating conditions during the optimisation period, the permeate quality remained similar. With longer trial duration the differences might have been easier to observe. Formation of cake layer has been said to have an effect on permeate quality. Similarly to the effect of operating conditions, longer trial period and longer backwash interval might show different results. The formation of cake layer had no effect on the quality of permeate, and thus a longer interval between membrane cleanings did not result in a better or worse permeate quality. More detailed data from water quality analyses can be found in appendix 5.

### **7.2.2 pH and alkalinity**

LPM filtration was hypothesised to have no effect on pH and alkalinity but as both characteristics are important in preserving concrete structures in further treatment, they were measure for safe keeping. The average pH-values measured during the experiment confirm this hypothesis: the value of pH did not change during membrane filtration or sand filtration. The average pH-values measured during both trial 1 and trial 2 are



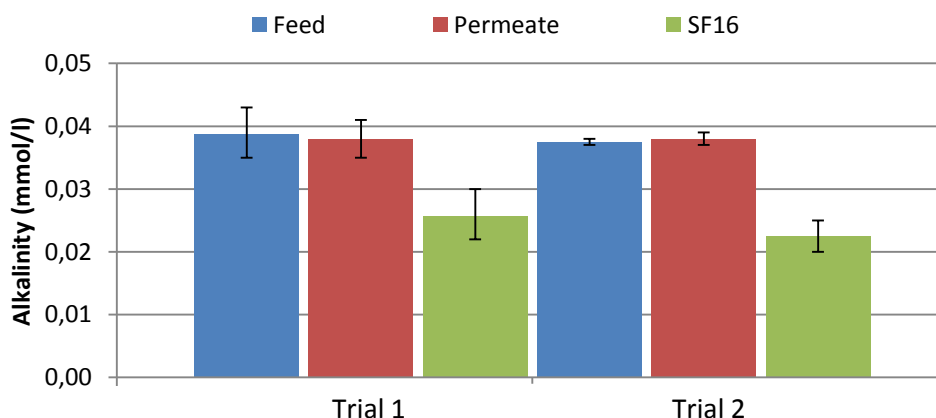
presented in Figure 18. Average values of measurements during trial 1 and 2 are presented with a bar whilst the error bars represent the extreme values measured.



**Figure 18. The average values of pH during both trials. Neither membrane or sand filtration affected the pH during filtration.**

From Figure 18 it can be observed that the mixing conditions of coagulant and the shorter retention time before treatment line 2, from which water was taken to membrane, has resulted in a smaller drop in the pH compared to treatment line 8. Clarified water in treatment line 8 before SF16 had a similar pH as SF16 (not shown) proving that pH was not reduced by sand filtration. The pH did not change during the 2<sup>nd</sup> trial either as presented in Figure 18. However, it can be seen that the fluctuation is smaller than with clarified water. Occasionally the pH of SF16 was very high during the 2<sup>nd</sup> trial phase as seen from error bars. This shows that mixing of coagulant was not optimal.

Similarly to pH the filtration was not expected to affect alkalinity of the water. However, since the alkalinity in the raw water is very low, the alkalinity after the LPM filtration was measured (Figure 19). From the values presented in appendix 5 and average values in Figure 19 it can be observed that LPM filtration did not affect the alkalinity of the water.

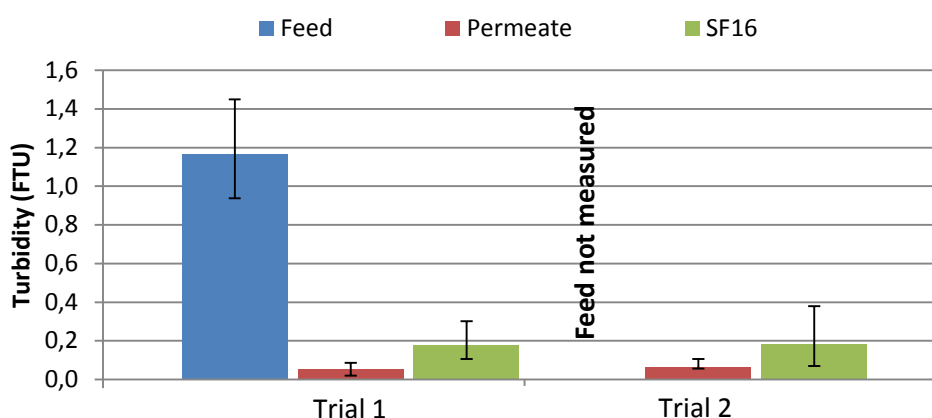


**Figure 19. The average values of alkalinity during trials.**

Again the longer retention time of treatment line 8 has resulted in a better and longer mixing of the coagulant which has reduced the alkalinity of the raw water. Similarly to pH it can also be observed that the variation of alkalinity during the second trial with coagulated water is smaller, due to the same factors as with pH.

### 7.2.3 Particle removal

The removal of turbidity and residual iron with membranes was hypothesised to be better compared to conventional treatment. The average turbidity values presented in Figure 20 confirm this hypothesis. Turbidity of flocculated water was not measured because of rapid sedimentation during the analysis but Poutanen (2012) has estimated that the turbidity is approximately 1.5 FTU. The turbidity values of permeate, excluding one measurement, were under 0.1 FTU whilst turbidity after SF16 was between 0.07...0.38 FTU.



**Figure 20. Average turbidity during piloting. Value of feed water in trial 2 is estimated.**

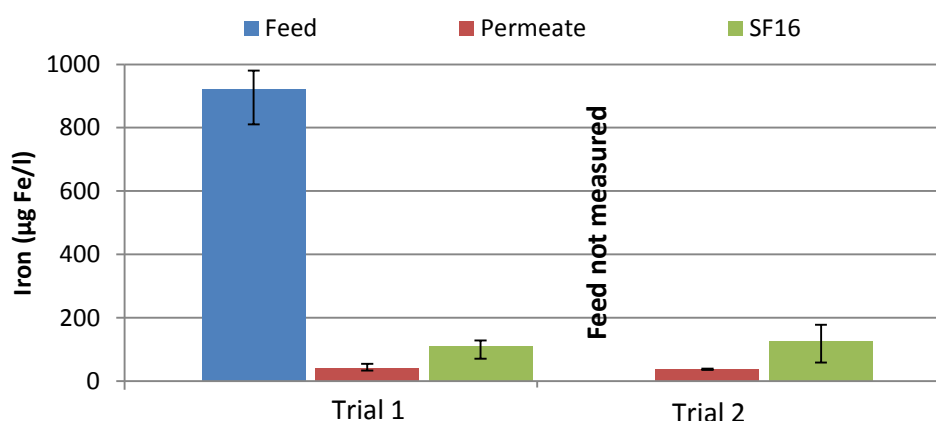
The reduction of turbidity in percentages is shown in Table 13. The average reduction of turbidity was better with membrane than with conventional treatment. From Figure 20 it can be seen that changing the feed water for the membrane has very little effect on permeate quality, the average turbidity during trial 1 was 0.06 FTU and during trial 2 the average value was 0.07 FTU. The turbidity of permeate was always smaller than the turbidity after sand filtration and below the recommendation of 0.1 FTU given in the National Water Quality Regulations (Ministry of Social Affairs and Health, 2000).

**Table 13. Reduction of turbidity in percentages during the trials. Membrane filtration was more consistent and effective in removing turbidity, as was expected.**

Trial 1			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	90.7	95.1	98.2
SF16	73.3	84.5	90.9
Trial 2			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	92.9	95.6	96.3
Sedimentation + SF16	74.7	87.8	95.4

It should also be noted that the fluctuation in turbidity after sand filtration is intense. One reason for fluctuation is the sand filter chosen, as SF16 has been found to clog easier than other sand filters. However, this fluctuation is peculiar for all sand filters in Pitkääkoski WTP. The fluctuation has also increased with decreasing water temperature, as suggested by Poutanen (2012).

The removal of residual iron with membranes was also effective. The amount of iron in flocculation was not measured but it is approximately 6000 µg Fe/l (Poutanen, 2012). The average amounts of residual iron in permeate for clarified water (trial 1) and flocculated water (trial 2) as feed water was 42.0 and 38.2 µg Fe/l, respectively. When compared to each other these values are within the range of uncertainty and thus it is difficult to estimate whether one is better than the other. Residual iron after SF16 was higher; the average amount was 150 µg Fe/l. The average values are presented in Figure 21.



**Figure 21. Average amounts of residual iron in the experiments. Value for feed water in trial 2 is approximately 6000 µg Fe/l.**

The removal percentages during both trials are shown in Table 14. The removal efficiency of membrane filtration was always better than conventional treatment. Similar to turbidity, the fluctuation in the amount of iron in permeate is low, whereas with SF16 the fluctuation is high. This is intuitive as residual iron increases turbidity.

**Table 14. Residual iron reduction in percentages. Again membrane was more effective compared to conventional treatment.**

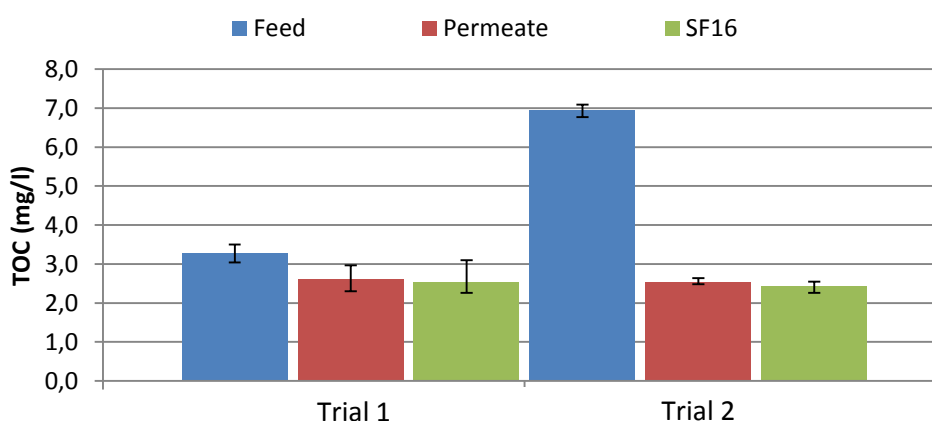
Trial 1			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	94.0	95.4	96.5
SF16	85.0	88.0	92.2
Trial 2			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	99.3	99.4	99.4
Sedimentation + SF16	97.0	97.9	99.0

The amount of iron in permeate was always below the quality regulation of 200 µg Fe/l (Ministry of Social Affairs and Health, 2000). It should also be noted that the cooling of raw water affects the amount of residual iron in sand filtration, which can be seen from the more intense fluctuation in maximum and minimum values during trial 2.

#### 7.2.4 Natural organic matter removal

NOM removal was measured with TOC, DOC, and AOC. Whilst TOC and DOC were measured only to observe the removal capability of the membrane, AOC was also measured to see the biofilm formation potential. Membranes alone are incapable of removing NOM, but combined with coagulation membrane should remove 50...60 % of NOM. Thus NOM removal was expected to be similar to conventional treatment.

TOC was measured by TOC analyser and LC-OCD analysis. The average results of TOC measured with TOC analyser during trials 1 and 2 are presented in Figure 22. The average TOC in membrane filtration was only slightly worse than sand filtration in trial 1 or sedimentation and sand filtration in trial 2. However, the average amount of TOC in permeate was the same in both trials, indicating that TOC removal was not enhanced by sedimentation before membrane filtration. TOC-values measured with LC-OCD were slightly smaller than values measured with TOC analyser, but same conclusions can be drawn from LC-OCD results.



**Figure 22. Average TOC values in trials. Conventionally treated water has slightly lower average amount of TOC compared to permeate.**

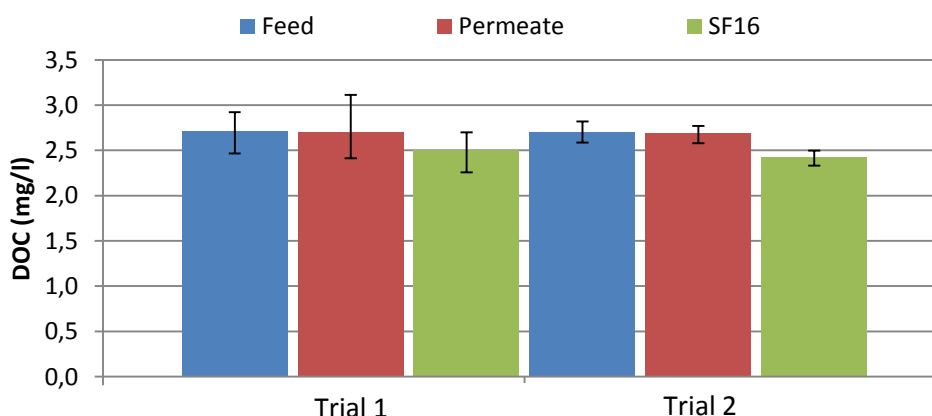
TOC reduction improved during the experiment, as can be seen from Figure 22 when comparing the average amount of TOC of conventionally treated water in trial 1 and trial 2. This is due to cooling of the raw water and in line with observations presented by Poutanen (2012). Similarly the residual TOC in permeate was lower in trial 2. Thus

it can be assumed that the lower TOC in trial 2 is due to more efficient coagulation and not the change of feed water. TOC reductions with both analysis methods during trials are shown in Table 15. Removal efficiency in trial 2 is slightly worse for both membrane and conventional treatment when measured with LC-OCD. This is due to lower amount of TOC in feed water when measured with LC-OCD. With LC-OCD the average TOC was 5.17 mg/l whilst TOC analyser gave TOC-value of 6.94 mg/l for feed water. However, these results confirm that conventional treatment was slightly better in removing TOC no matter what method was used. The results gotten in trial 2 with conventional treatment are similar to results presented by Poutanen (2012). The reduction with LPM filtration is close to the original hypothesis.

**Table 15. TOC reductions with membrane filtration and conventional treatment during both trials.**  
A = TOC analyser, L = LC-OCD analysis.

Trial 1						
	Minimum (%)		Average (%)		Maximum (%)	
Method	A	L	A	L	A	L
Membrane	14.1	13.7	20.5	18.6	24.2	23.3
SF16	21.5	17.5	25.1	21.5	28.3	27.6
Trial 2						
	Minimum (%)		Average (%)		Maximum (%)	
Method	A	L	A	L	A	L
Membrane	62.3	49.5	63.4	55.0	65.0	58.0
Sedimentation + SF16	63.4	53.7	65.1	57.7	66.6	60.4

When measured with TOC analyser, both membrane and sand filter are performing poorly in removing DOC as shown in Figure 23. As these treatment steps remove matter by screening and are thus incapable to remove dissolved substances, the poor performance was expected. Almost all TOC was DOC, which can be seen by comparing Figure 23 to Figure 22. The amount of DOC in clarified and flocculated water is almost the same and there is little difference in the permeate quality between trials.



**Figure 23. Average DOC values in trials 1 and 2 measured with TOC analyser.**

DOC measured from data acquired with LC-OCD, however, show very different results compared to DOC presented in Figure 23. LC-OCD results are different for two reasons. Firstly LC-OCD measured significantly smaller amounts of DOC in permeate and sand filtered water, 2.3 mg/l and 2.2 mg/l in trial 1 and 2.2 mg/l and 2.1 mg/l in trial 2, respectively. Secondly DOC during trial 2 in flocculated water was 4.7 mg/l, almost double compared to DOC of 2.7 mg/l measured with TOC analyser. DOC in trial 1 was the same with both methods. DOC was 2.7 mg/l and 2.9 mg/l for TOC analyser and LC-OCD, respectively.

The minimum, average, and maximum removal of DOC is presented in Table 16. It can be seen that when measured with TOC analyser no removal of DOC took place during membrane filtration, but conventional treatment was able to remove some of the DOC. However, the uncertainty of measurement is 20 % and thus all the results are within the marginal of uncertainty. According to LC-OCD analysis DOC was significantly reduced with membrane filtration and conventional treatment.

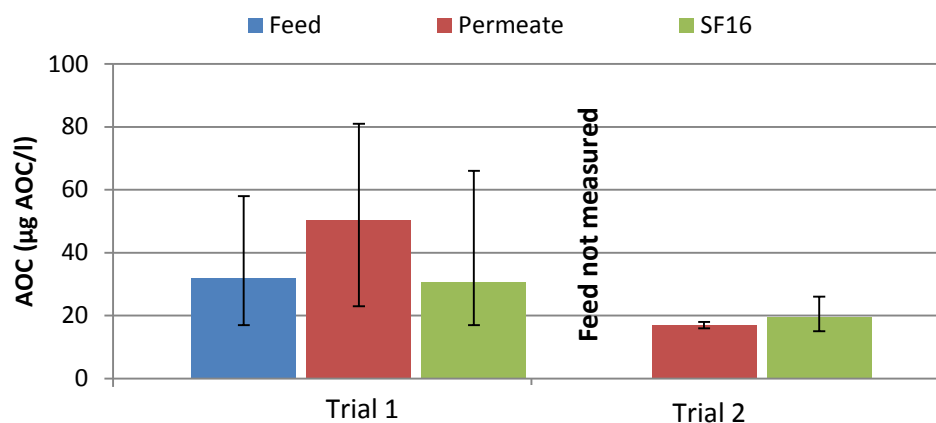
**Table 16. DOC removals from membrane filtration and conventional treatment in both trials.**  
A = TOC analyser, L = LC-OCD analysis.

<b>Trial 1</b>						
	Minimum (%)		Average (%)		Maximum (%)	
Method	A	L	A	L	A	L
Membrane	0.0	13.7	1.6	15.9	3.8	17.9
SF16	0.6	15.5	7.7	20.2	11.3	25.1
<b>Trial 2</b>						
	Minimum (%)		Average (%)		Maximum (%)	
Method	A	L	A	L	A	L
Membrane	0.0	51.7	0.7	53.0	1.8	53.9
Sedimentation + SF16	7.6	51.8	10.3	54.5	13.5	57.3

Similar to TOC results, conventional treatment is more effective in removing DOC compared to membrane filtration. The residual DOC in permeate is similar in both trials and the usage of sedimentation as membrane pretreatment has not improved DOC removal.

AOC is the part of organic matter that is consumed by bacteria and thus can be used as an indicator of the biofilm formation potential. Measuring AOC from flocculated water was not possible because of high iron content. Average amounts of AOC in permeate were larger than in feed water or sand filtered water during trial 1, but the fluctuation is very high. In trial 2 sand filtered water had a higher average AOC than permeate. This inconsistency can be due to uncertainty of the measurement.

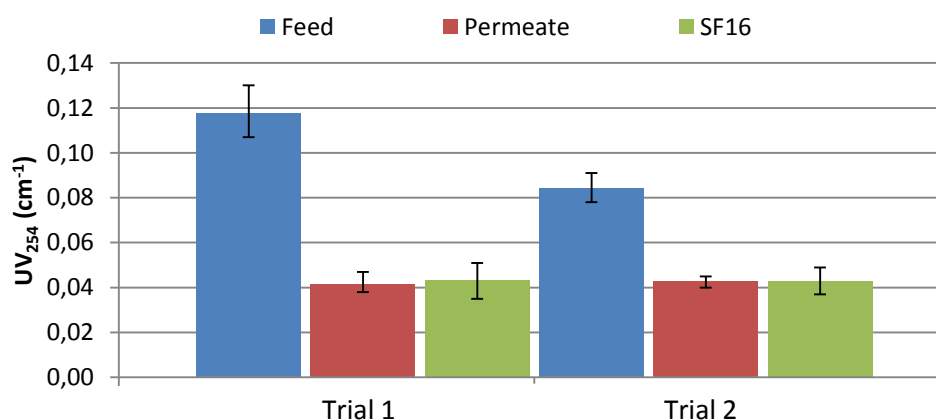
Average amounts of AOC from four and three measurements during trial 1 and 2, respectively, are presented in Figure 24. The amount of AOC in feed water during trial 1 was above 27  $\mu\text{g AOC/l}$ , value which was found to cause biofilm formation in NF membranes (Vrouwenvelder et al., 1998).



**Figure 24.** Average amounts of AOC in trials 1 and 2. During trial 1 there occurred much variation between measured values, during trial 2 the values were more consistent.

### 7.2.5 Natural organic matter fractions

As the removal of NOM is dependent on coagulation, the reductions of NOM fractions in membrane filtration were also expected to be similar to conventional treatment. The amounts of different NOM fractions were measured with three analyses.  $\text{UV}_{254}$  was measured to find out the aromaticity of the NOM, HPSEC analysis was done to find out the amount of different fractions according to molecular size, and LC-OCD test was done to study the hydrophobicity on NOM. The average  $\text{UV}_{254}$  values in both trials are presented in Figure 25. It can be seen that residual UV absorbance is similar for conventional treatment and membrane filtration.



**Figure 25.** Average UV absorbance in both trials. Membrane filtration reduced aromaticity as effectively as conventional treatment.

From Figure 25 it can be observed that membrane filtration produced more stable water compared to conventional treatment. The UV absorbance after conventional treatment was occasionally higher than what membrane could produce but conventionally treated water also had lower values than what membrane produced. Pre-treating the samples with centrifuge in trial 2 has resulted in a smaller UV absorbance which also affected the average removal during trial 2. From Table 17 it can be seen that the removal efficiency of UV absorbance is very high for both treatment methods. The average reduction in membrane filtration is slightly higher than what was measured for conventional treatment.

**Table 17. UV absorbance reductions show that membrane treatment was slightly more effective in removing UV<sub>254</sub> but the difference is small, however, the variation of measured values is smaller.**

Trial 1			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	61.1	64.5	70.5
SF16	57.4	63.2	69.4
Trial 2			
	Minimum (%)	Average (%)	Maximum (%)
Membrane	42.3	48.5	54.9
Sedimentation + SF16	38.8	48.1	58.4

When the reduction of UV<sub>254</sub> measured with spectrophotometer is compared to DOC removal measured with TOC analyser, it can be seen that membrane was more effective in removing aromatic compounds represented by UV<sub>254</sub>. This observation is confirmed by SUVA reductions given by LC-OCD analysis. When measured with LC-OCD, in trial 1 SUVA was decreased in membrane filtration and sand filtration with 17.0 % and 16.9 %, respectively, and in trial 2 the reductions were 47.7 % for membrane filtration and 43.9 % for sedimentation and sand filtration. Average SUVA values with both methods from each trial are presented in Table 18. SUVA was always below 4, indicating that NOM was mostly hydrophilic, except feed water SUVA in trial 1 measured with TOC analyser and spectrophotometer. This high SUVA might be explained by residual iron floc which disturbed the UV<sub>254</sub> measurement. In trial 2 the feed water was pre-treated with centrifuge which resulted in lower UV absorbance, and similar SUVA-values for both analyses.

**Table 18. Average SUVA values measured in both trials show that membrane treatment was effective in removing aromatic components. A = TOC analyser and spectrophotometer, L = LC-OCD analysis**

OCD analysis				
	Trial 1		Trial 2	
	A	L	A	L
Feed	4.33	2.03	3.12	3.11
Permeate	1.54	1.68	1.60	1.63
Conventional treatment	1.72	1.68	1.77	1.74



HPSEC analysis was performed to study the effect of membrane treatment on different molecular size of NOM. The size distribution in clarified water was fairly constant and thus the peaks from permeate and SF16 are compared to the average values of feed water. The peaks from chromatograph are divided into seven categories: (I) > 5000 g/mole, (II) 4000...5000 g/mole, (III) 3000...4000 g/mole, (IV) 1000...3000 g/mole, (V) 500...1000 g/mole, (VI) 350...500 g/mole, and (VII) < 350 g/mole. The peaks can be divided into large, intermediate, and small MW substances. First three peaks are categorised as large substances, peaks IV and V are intermediate substances, and peaks VI and VII are small substances. In some samples 8<sup>th</sup> peak was measured, but since it was not visible in all samples, the 8<sup>th</sup> peak was excluded.

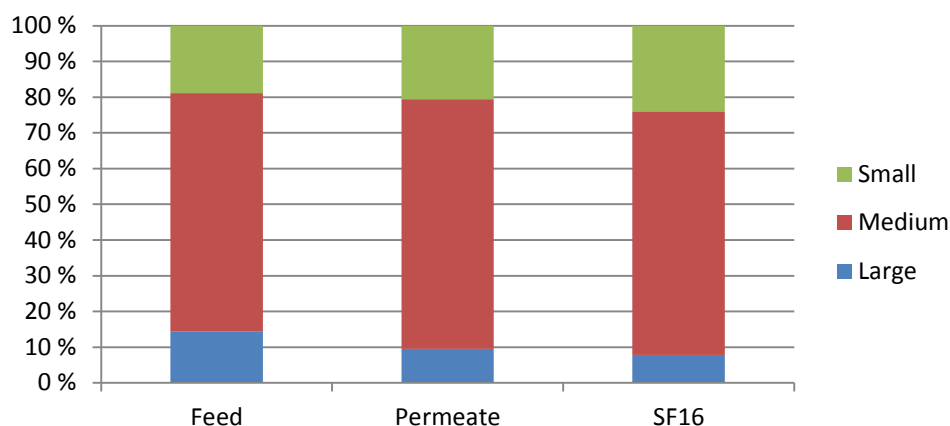
Generally the size distribution between the samples remained similar during the trials. Sum of all peak heights (SOPH) representing total amount of NOM showed a slightly decreasing trend throughout the experiment. Table 19 presents the average heights of different peaks during trial 1 and the reductions for membrane and sand filtration. Negative value means that the amount of corresponding fraction has increase during filtration.

**Table 19. Average peak heights and reductions during trial 1.**

Peak	Feed	Permeate		SF16	
	Average height	Average height	Reduction (%)	Average height	Reduction (%)
I	36.8	40.2	-9.0	33.2	10.0
II	96.2	74.0	23.1	54.3	43.6
III	336.5	214.0	36.4	137.3	59.2
IV	840.9	849.0	-1.0	592.4	29.6
V	1316.0	1569.3	-19.2	1374.2	-4.4
VI	566.9	658.8	-16.2	638.3	-12.6
VII	45.1	50.8	-12.6	51.5	-14.2
SOPH	3399.7	3640.2	-7.1	3085.4	9.2

Both membrane and sand filtration were successful in removing large substances but the amount of small substances was increased with both techniques. This means that during filtration the UV absorbance of small molecular weight compounds has increased. Coagulation has been shown to be effective against larger NOM compounds and possibly during filtration when flocs were broken the smaller, more loosely bound compounds escaped flocs and increased the peak height. Sand filter was effective in reducing the SOPH whereas during membrane filtration SOPH actually increased. This increase is however, within the margin of uncertainty of HPSEC analysis.

Figure 26 presents the share of large, intermediate, and small substances. From this figure it can be seen that both techniques reduce the share of large MW substances to fewer than 10 % but were ineffective in reducing medium-sized substances. To remove the smaller fractions other treatments, such as active carbon filters, should be employed.



**Figure 26. Size distribution in feed, permeate, and SF16 during trial 1. Sand filtration was more effective in removing large substances from water.**

Results are very similar in trial 2. Peak heights in feed water have increased, but the peak heights for permeate and conventional treatment have remained the same, meaning that the reduction has increased in both cases. Again conventional treatment is performing better compared to membrane filtration. Reduction in conventional treatment is positive in each peak except peak VII. Membrane filtration is showing slightly negative numbers in most size categories and SOPH is the same as for feed water meaning that the amount of NOM is similar. The share of size categories is similar to Figure 26. Average peak heights and reductions in trial 2 are shown in Table 20.

**Table 20. Average peak heights and reductions in trial 2.**

Peak	Feed	Permeate		Conventional treatment	
		Average height	Reduction (%)	Average height	Reduction (%)
1	46.0	32.0	30.4	37.0	19.6
2	66.3	80.3	-21.1	51.3	22.6
3	213.5	226.0	-5.9	140.0	34.4
4	881.5	914.5	-3.7	566.8	35.7
5	1614.5	1637.5	-1.4	1383.5	14.3
6	728.0	695.0	4.5	681.3	6.4
7	51.0	84.8	-66.2	89.0	-74.5
SOPH	3755.0	3780.5	-0.7	3091.8	17.7

LC-OCD analysis was conducted to measure the hydrophobicity on NOM. LC-OCD fractionates TOC into DOC and particulate organic carbon, and further fractionates DOC into hydrophilic and hydrophobic fractions. An example of the results can be found from appendix 5. DOC was fractionated into hydrophobic and hydrophilic

fractions. Hydrophilic fraction was found to be dominant; all DOC was found to be hydrophilic. Hydrophilic NOM is dominant because coagulation effectively removes hydrophobic fractions but has no effect to hydrophilic fractions (Chen et al., 2007; Grey et al., 2007). It was stated, however, that the analysis consistently gave too high values for hydrophilic NOM and thus certain caution has to be presented in drawing conclusions from this fractionation.

Hydrophilic NOM can be divided into refractory and biogenic fractions. Refractory fractions are humics and their building blocks. Humics and building blocks have a molecular weight of 1000 and 300...500 g/mol, respectively. Building blocks are natural breakdown products of humics. Biogenic fraction contains biopolymers and low MW organic acids and neutrals. Biopolymers, which have the largest molecular weight (> 20 000 g/mol) are typically polysaccharides but might also contain proteinic matter. Low MW organic acids and neutrals have a MW below 350 g/mol. The removal of these fractions in membrane filtration and conventional treatment is presented in Table 21. Conventional treatment is always more effective in removing refractory fraction, whilst MF is more efficient in removing biogenic fractions.

**Table 21. Fractionation of hydrophilic NOM. Hydrophilic NOM is divided to refractory and biogenic NOM. Membrane filtration removes biogenic NOM more effectively, whilst conventional treatment is more effective in removing refractory NOM.**

		Refractory NOM				Biogenic NOM					
		Humics		Building blocks		Biopolymers		Neutrals		Acids	
		ppb-C	%	ppb-C	%	ppb-C	%	ppb-C	%	ppb-C	%
Trial 1	Feed	1179.6	-	1006.9	-	35.4	-	638.9	-	112.4	-
	Permeate	899.7	23.7	936.2	7.0	38.1	-7.6	498.7	21.9	44.4	60.5
	SF16	770.7	34.7	909.3	9.7	43.0	-21.5	507.1	20.6	72.7	35.3
Trial 2	Feed	2997.5	-	1033.6	-	45.8	-	993.8	-	125.9	-
	Permeate	926.9	69.1	886.8	14.2	34.7	24.2	484.1	51.3	26.7	78.8
	SF16	794.6	73.5	831.8	19.5	40.1	12.6	517.7	47.9	38.8	69.2

LC-OCD results confirm most of the conclusions drawn from HPSEC results. Permeate quality is similar in both cases, the effect of sedimentation as a pretreatment is small. Sedimentation mostly removed large substances, presented by biopolymers and humics. The removal of largest MW substances by membrane and SF16 is negative in trial 1. From both LC-OCD and HPSEC results it can be seen that conventional treatment is more effective in removing the medium sized fraction compared to membrane filtration in both trials. According to LC-OCD, MF is effective in removing low MW substances, contrary to what was suggested by HPSEC. The results gotten with LC-OCD in trial to are similar to results gotten by Keucken et al. (2012) who experimented with Swedish surface water.

### 7.2.6 Microbiological quality

To measure the microbiological quality of the water, R2A agar cultivation and ATP measurements were done. Both of these tests measure the amount of bacteria and microorganisms and the removal of these was hypothesised to be excellent. The removal of virus was not tested, in theory some viruses might be removed by coagulation.

R2A agar analysis proved the excellent performance of MF membrane with both feed waters on removing microorganisms and bacteria. Colonies were formed in only one sample and thus the reduction in both trials is almost 100 %. Cultivations with conventionally treated water had significantly more formed colonies; the average amount of colonies was 55.6 pmy/ml, a reduction of 52 and 62 % in trials 1 and 2, respectively. It is, however, difficult to estimate what is the effect of coagulation on MF performance. In theory MF membrane with this pore size should be able to remove all bacteria, but reduction without coagulation was not tested in this experiment. The reduction of conventional treatment is not applicable to be used as reference point to show the performance of coagulation, because whilst effectively removing floc sand filters also have their own microbiological growth and this might affect the results of R2A agar after conventional treatment.

The results from ATP analyses confirm the good bacterial and microbial removal capability of the membrane. The minimum and maximum removal percentages gotten during the trials are presented in Table 22. At this point it should be remembered that the amount of ATP is very low and there exists a considerable amount of uncertainty with such low numbers. They do, however, strongly support the conclusions made from R2A agar cultivation.

**Table 22. ATP reductions during the trials. Membrane filtration is more effective in removing microbiological activity compared to conventional treatments.**

	<b>Minimum (%)</b>	<b>Maximum (%)</b>
<b>Trial 1</b>		
Membrane	40.4	80.0
SF16	26.7	49.5
<b>Trial 2</b>		
Membrane	53.8	92.9
Sedimentation + SF16	16.9	89.5

The membrane filtration was more effective in removing both R2A and ATP thus proving that the hypothesis claiming that membranes would effectively remove bacterial and microbial activity from the water was true. Membrane filtration performed better than sand filtration alone or in combination with sedimentation.

## **8 Summary**

This chapter discusses the results presented in chapter 7. First factors related to membrane performance are discussed and then the effect of membrane performance on operational expenses is reflected. After this, conclusions from water quality analyses are drawn. This chapter ends with conclusions on the whole thesis.

### **8.1 Discussion**

#### **8.1.1 Membrane performance**

To evaluate the possibility of retrofitting membranes to Pitkääkoski WTP a pilot study was conducted. By investigating existing retrofits two places to retrofit membranes were identified; after sedimentation to replace sand filtration and after flocculation to replace sedimentation and sand filtration. With both feed waters the membrane experienced a steady decline in permeability. The decline of permeability can be divided into two stages. First, during the first hour of operation, a very rapid decline occurs. This drop is caused by physically irreversible fouling which is unrecoverable without chemical cleaning. After the initial drop, steadier drop in permeability was observed. This decline is controlled with backwashing. The decline in permeability is, however, still rapid and did not reach a steady state during trials. During trial 2 the initial drop in permeability was higher, but after the initial drop the degradation of permeability was more gradual than during trial 1. Several factors can influence membrane fouling, and slight adjustments may result in less fouling.

One reason for the rapid decline can be membrane material. Yamamura et al. (2007) experienced problems when using similar PVDF MF membrane because the periodical backwashing was unable to stop the TMP increase. With a different type of membrane the decline in permeability might be more controllable with backwashing. This hypothesis is supported by the observation that most of NOM present in the feed water was hydrophilic. As was discussed earlier in chapter 3, hydrophilic fraction has been found to be the most fouling fraction for LPMs and fouling is mostly irreversible. Hydrophobic membranes especially have been found to foul easily at the presence of hydrophilic NOM fractions. Thus improvements may come from selecting a membrane material designed for low fouling rates by hydrophilic NOM fractions or by changing coagulation to target these fractions, as suggested by Carroll et al. (2000).

Another reason could be the high operating pressure which was used. A pressure of 0.9 bar was used in the experiment and to achieve the desired flux, the flow through membrane was controlled from the permeate side. The rate at which the flow was controlled was very high, similar fluxes could have been reached with a 0.6 bar pressure. Lower pressure decreases the amount of fouling as it reduced pore penetration. Aoustin et al. (2001) experienced lower flux decline with 100 kDa UF membrane when lower pressure was applied, whilst the final flux with both pressures is identical. They hypothesised that at higher operating pressure critical flux was reached at which deposition of particles started to control the flux.

Third reason for the drop in permeability might be that the backwash conditions were, despite considerable efforts made in trial 1, still not optimal. The pressure during backwashing was at best 1.0 bar, only slightly higher than the operating pressure. Most often the pressure was 0.66 bar. With backwash pressure of 1.0 bar, only a 15 second backwash could be conducted. Backwash pressure should be at least twice the operation pressure (Nakatsuka et al., 1996). Backwashing conditions were shown to have an effect on the decline in permeability. During optimisation period membrane experienced more gradual decline in permeability when higher backwash flux, and therefore higher pressure, was used. This effect was, however, very small and the decline is not explained only by backwashing conditions. It can also be hypothesised that permeate is not the best backwash water as suggested by Li et al. (2009). Demineralized water, or water after active carbon filtration, could be tested as backwash water to see if the rate of fouling would be smaller.

Membrane fouling might also be affected by the usage of ferric sulphate as coagulant. Yamamura et al. (2007) found iron as one substance causing physically irreversible fouling. Irreversible fouling is observed at the beginning of the filtration. This effect might be one reason why the drop in permeability during the first hour of operation was higher with flocculated water than with clarified water. Yamamura et al. observed no fouling due to aluminium and thus to eliminated the effects of iron aluminium-based coagulants should be tested. Still, iron floc which accumulated on the membrane surface was effectively removed and the biggest reason for the rapid initial decline is probably not originated from iron. The concentration of other impurities, such as hydrophilic NOM, was also higher in trial 2 than in trial 1.

After the rapid initial decline membrane experienced more gradual decline in trial 2 compared to trial 1. Panglisch et al. (2008) have suggested that colloidal or particular iron smaller than 0.45  $\mu\text{m}$  causes membrane fouling. A floc size measurement (results not presented) confirmed that only the smallest iron flocs escaped through sedimentation and it can be hypothesised that they have had an effect on membrane fouling even after the initial drop in permeability. Coagulants escaping sedimentation have been found problematic in another LPM filtration experiment (Kainua and Hentilä, 2011).

It was also noticed that as the raw water cooled down during this experiment, the particle removal with sand filtration worsened. As the raw water gets colder, the viscosity of water increases and thus mixing of coagulant is more uneven and the functionality of sedimentation is weaker. This results in smaller floc size and increases the clogging of sand filtration. Due to shorter mixing time in treatment line 1 the floc size is minimal in this treatment line. From these two observations it can be assumed that the operating conditions during the experiment were poor for membrane filtration as the floc size was very small. Small sized floc blocks membrane capillaries and/or forms a very thick cake layer. Whichever was true here, the result is a rapid decline in membrane permeability. Although the feed water to the membrane was warmed up by the ambient temperature, floc was always formed in the treatment process, and thus the warming in the feed tank did not viscosity during floc formation.

MF membranes are prone to pore blocking and adsorption (Zularisam et al., 2006). In trial 1 only small floc existed as bigger floc had settled. When flocculated water was used in trial 2 the large flocs were also present which might have enhanced the overall performance. Larger flocs form a cake layer on the membrane surface and this cake layer might have caught smaller flocs thus preventing them from blocking the pores. During backwashing this cake layer was removed, which resulted in a more gradual TMP increase.

Chemical treatment was conducted every other day to support regular backwashing. No chemical treatment alone was capable of regaining the original permeability of the membrane during trial 1. This would indicate that both organic and inorganic substances are present. A CEB with either citric acid or NaOH and NaClO was always capable to regaining approximately half of the original permeability. Different chemical cleanings were not tested in trial 2. Due to high load of iron the fouling profile might have been different.

At first the chemical treatments were conducted at temperatures similar to permeate quality, but both NaClO and citric acid cleanings were inefficient. Even when 20 °C water was used, citric acid failed to regain original permeability. CEB with NaClO failed to show any improvement when used with cold water and was not tested with warmer water. It can be hypothesised that with warmer water the result would have been more positive but still the original permeability might not have been achieved.

The chemicals that were used for CEB were recommended by the membrane manufacturer. These chemicals are, however, expensive and other chemicals could be tested. Also, as was already mentioned, the amount of chemicals and time needed to conduct a CEB should be studied further. It was noticed that the cleaning effect of a chemical was immediate. During the first minute of CEB, flow and TMP had already experienced a significant improvement, and this improvement was finalised by the first backwash cycle. Further treatment had no effect in permeability. This would indicate that a long, low-pressure backwash where chemicals are added might be enough to regain original permeability. Current equipment, however, is unsuitable for such operation.

### **8.1.2 Operational information during pilot**

Different operating conditions were compared to find the optimal operating conditions. Not surprisingly, conditions with higher flux and longer backwash interval proved to be most economical, as more water was produced with least amount of energy. To really estimate the effect of different operating conditions, however, longer tests should be conducted.

The recovery of membrane filtration was at best 87.8 % which is smaller compared to recovery of the conventional treatment currently operational at Pitkääkoski WTP. It is, however, not a proper comparison to compare the recovery percentage of the existing treatment to the one gotten from the pilot. Membrane configurations in WTPs using membrane filtration are much more complicated compared to configuration in the pilot. Recovery is normally enhanced by recirculating permeate or by using retentate from membrane filtration as the feed for another membrane. Recovery can also be increased by treating the backwash water. With an optimised system, recoveries over 99.5 % have been achieved (Panglisch et al., 2005; Gimbel et al., 2007; Heijman and Bakker, 2007).



Operating costs of the pilot were also of interest. The energy consumption at Pitkääkoski WTP for sedimentation and sand filtration is roughly estimated to be  $0.012 \text{ kWh/m}^3$ , which is much less than the energy consumption calculated in the pilot. Again, however, the arrangements in the piloting give very little indication on the real consumption of membrane filtration and the energy consumption should be compared to existing membrane processes. As was previously mentioned, the increase in energy consumption after retrofitting membranes to WTPs has been  $0.0\ldots0.22 \text{ kWh/m}^3$ . The energy consumption is highly dependable on pumping energy. The operating pressure in this experiment was 0.9 bar. Jacangelo and Noack (2005) have suggested that for hollow fibre polymeric membranes the operating pressure is  $1.4\ldots2.0$  bar. For immersed systems the operating pressure is lower.

To estimate the difference in OPEX regarding chemical costs, further investigation is needed. Two main reasons why cost for chemicals is difficult to estimate are the confusions with chemical consumption of CEB and with coagulant consumption during coagulation. It was observed during the experiment that a simple CEB where chemicals are added to the backwash water might be sufficient way to control fouling. With the equipment that was used for this experiment, adding chemicals to the backwash water is not possible. Estimation of chemical costs is also difficult because similar coagulation conditions were used for membrane filtration and conventional treatment. The amount of coagulant used was optimal for conventional treatment but as earlier was mentioned, the required amount of coagulant for membrane filtration might be less. The chemical cost for coagulation at Pitkääkoski is approximately one cent/ $\text{m}^3$ , slightly less than the chemical costs of  $1.04 \text{ cent/m}^3$  at Lello WTP (Sandvik and Ødegaard, 2010).

### **8.1.3 Water Quality**

To be able to compare the performance of the membrane to the conventional process currently used at Pitkääkoski WTP, an intensive water quality analysis programme was made. It was hypothesised that pH and alkalinity would not change during the filtration. However, as these characteristics are crucial in further treatment tests were conducted for precautionary reasons. As was presented in chapter 7.3.2 membrane filtration had no effect in either characteristic. However, it can be noted that there was a significant difference between treatment lines. Treatment line 1, from which the feed water for the pilot was taken, had a significantly higher pH and alkalinity compared to treatment line 8 from which sand filtered sample was taken. This suggests that mixing of the chemical

and retention time after adding the chemical is not uniform and thus it may have had an effect on the removal of NOM. However, TOC values measured during this experiment and TOC, UV<sub>254</sub>, and HPSEC values measured by Poutanen (2012) suggest that the effect is small. TOC before SF16 was measured few times, and the average value was 0.1 mg/l lower than TOC of the membrane feed water.

Another hypothesis was that membrane filtration would effectively remove particles from the water. This was confirmed by the quality analyses as the amount of both turbidity and residual iron were always below values from sand filtered water and also below water quality regulations. Very little difference was found between trials 1 and 2, demonstrating the good particle removal ability of the membrane. The good removal of turbidity and residual iron allows the further treatment steps to focus on removing NOM instead of particles. Residual turbidity has been found to consume oxygen during ozonisation and clog active carbon filters.

Regarding NOM removal membranes might be able to challenge current conventional treatment at Pitkääkoski WTP. When membrane was compared to sand filtration in trial 1 the reduction of TOC was 5 % less than with sand filter. The efficiency of membrane in removing TOC was enhanced in the second trial when flocculated water was used as feed water. In trial 2 the difference in reduction was only 1.7 %. Both differences can be explained by margin of uncertainty in analyses or the conditions in chemical mixing before flocculation. Earlier it was mentioned that chemical mixing is equal for all treatment lines, but it can be hypothesised that such a small difference could derive from the mixing conditions.

DOC shows a similar trend. Conventional treatment was more efficient in removing DOC but the difference is very small, and thus it is difficult to say if conventional treatment is actually performing better. Measured differences are always within the uncertainty of measurement. The removal of DOC with membrane filtration depends on the coagulation conditions. With conventional treatment the focus is on creating a settleable floc, but with MF filtration size larger than 0.5 µm might be enough to efficiently separate the floc. In order to find the true difference in NOM removal between conventional treatment and membrane filtration a more detailed sampling program should be carried out, where each treatment step would be analysed.

The average amount of AOC in the feed water during trial 1 was over the values where biofouling was found by Vrouwenvelder et al. (1998). They, however, used a NF membrane and it is possible that a higher concentration of AOC is needed to cause biofouling on a MF membrane. The amount of AOC could not be measured from flocculated water and thus it remains an open question whether changing the feed water to flocculated water resulted in increased biofouling or not. The amount of AOC in permeate was smaller in trial 2 but whether this is due to AOC consumption by biofilm is difficult to say. No literature was found on biofouling of MF membrane in potable water production that discussed the required amount of AOC to influence biofilm formation potential on MF membrane.

Membranes have a preferential rejection of the aromatic and hydrophobic NOM (Cho et al., 2000). Coagulation is also more effective on hydrophobic NOM which is indicated by the high rejection of UV versus DOC (Chen et al., 2007; Gray et al., 2007). These statements were true for this experiment which can be seen from the reductions of UV and DOC. UV absorbance was effectively reduced to almost half whereas DOC was not reduced at all during membrane filtration. The SUVA results from UV absorbance and DOC analyses conducted individually were confirmed by LC-OCD analysis. Membranes were not as effective in removing humics as conventional treatment, but as suggested by low SUVA of permeate and conventionally treated water both methods mainly removed humic substances. Coagulation is ineffective towards polysaccharides and proteins, presented as biopolymers in LC-OCD results (Kimura et al., 2005).

## **8.2 Conclusions**

Operation of conventional treatment at Pitkääkoski WTP started in 1950s. Since then the WTP has been expanded to its current capacity of 7000 m<sup>3</sup>/h. Due to rising water demand the WTP is looking to expand treatment capacity. The surface load of the conventional treatment line can be increased to a certain point but increasing over the physical limit would result in decreasing treated water quality regarding residual particles. Instead of updating the current treatment system, membrane filtration has been thought as an alternative. Membranes could also increase the buffer capacity of the WTP. Retrofitting Pitkääkoski WTP with LPMs was experimented in this thesis. Firstly a literature study was conducted in order to find where membranes could be retrofitted in the existing process and which treatment steps could be replaced. The most

advantageous options turned out to be substituting the existing sedimentation and sand filtration or sand filtration alone.

An intensive piloting phase was conducted where both replacements were tested. Operation data from the membrane filtration was recorded and a substantial amount of different water quality indicators were measured to examine the quality of water produced with membrane filtration. The operation of membrane filtration proved out to be very reliable, and very little maintenance was required. Despite changes in operating conditions permeate quality remained constant and membrane produced better quality water compared to conventional treatment in both trials. A comparison between LPM filtration and conventional treatment is presented in Table 23.

**Table 23. Comparison between LPM filtration and conventional treatment.**

	<b>LPM filtration</b>	<b>Conventional treatment</b>
Advantages	<p>Fully automatic, simple and reliable operation, easy to detect broken capillaries</p> <p>Small footprint with modules, easy to increase capacity</p> <p>Very good particle and microorganism removal despite fluctuating raw water</p>	<p>Pumps unnecessary during normal operation, low energy costs</p> <p>Well-known and proven process technology</p> <p>Low amount of effluent</p> <p>Pressureless operation</p>
Disadvantages	<p>Limited operating experience</p> <p>Increased downtime</p> <p>Amount of backwash waste</p> <p>Regular use of cleaning chemicals, chemical waste</p>	<p>Monitoring and maintenance effort required</p> <p>Not adaptable to rapid change in raw water, poor particle removal</p> <p>Control over water quality limited</p> <p>Difficult to control channelization of sand filter</p>

When flocculated water was used, the permeability of the membrane was initially lower compared with clarified water after sedimentation. However, after two days of operation the permeability was similar in both cases. The difference in permeability had a very small effect on the amount of water produced. Similarly the water quality was similar with both flocculated and clarified feed water. Sedimentation as a pretreatment had very little effect on the performance of the membrane. Other pretreatments that could easily be introduced are few; adsorbents could be added to enhance NOM removal. Ozonisation and active carbon filters are already operational at Pitkääkoski WTP, but the

treatment system is designed to operate by gravitation and if said treatments were to be used as pretreatments, massive pumps should be installed to reverse the flow direction.

Water quality analyses proved that membrane filtration produces better quality water compared to conventional treatment. Removal of particles and microbiological activity was better and if membranes were retrofitted to replace sedimentation and sand filtration, better water quality regarding turbidity would enable cost-savings in post-treatments. Residual iron would no longer consume oxygen, turbidity would not clog active carbon filters, and a multi-barrier system would be built to prevent microbiological contamination.

While the excellent particle removal has made membranes an interesting option for WTPs, membrane fouling still remains as a major drawback of the membrane filtration. Fouling, especially the one caused by NOM, has been found to be intensive and raise the operational expenses of potable water treatment. Increased amounts of NOM have been observed in raw water at Pitkääkoski WTP. NOM removal with membranes is similar to conventional treatment as both are reliable on coagulation. Regarding NOM fractions, LPM filtration was more effective in removing biogenic fraction, such as biopolymers and low molecular weight substances but conventional treatment was more effective in removing humic substances.

Previously coagulation conditions were modified to improve NOM removal of conventional treatment but the result was worse treated water quality regarding turbidity. If membrane filtration was used, adding coagulants would not result in worse permeate quality, but raised operational expenses as membranes would foul more and had to be cleaned more often. Which option is more expensive is difficult to say.

Operational costs for membrane filtration are estimated to be higher compared to existing treatment. Chemical consumption and energy consumption can be expected to raise or remain the same while maintenance costs are said to be lower. However, there are many surpluses with membrane retrofitting. The retention time of the treatment plant is shortened significantly which would result in an increase in capacity and if the capacity is further increased sometime in the future by adding more membranes, modular structure of membranes offer simple and fast solutions for capacity increasing.

More research should be conducted to deepen the understanding of retrofitting LPMs to Pitkääkoski WTP and how to solve the specific problems that the WTP has. Membranes have proven to be a reliable option for conventional treatment regarding water quality but issues related to fouling should be solved. Fouling was constant throughout the whole experiment and no steady-state was reached. To find out the steady-state flux longer experiments should be conducted. Time of operation that was used in this experiment was only two days and a longer period of two weeks should be used.

Different membrane materials and different pore sizes should also be tested. NOM was mostly hydrophilic; this fraction has been found to cause mostly irreversible fouling. A material that would foul least in these conditions should be found. Different pore sizes should be tested to see if further improvement in water quality can be reached without new problems appearing in operation of the LPM system. Different pore sizes might also lead to less fouling because membranes with smaller pore size have been found to be subjected to cake layer formation instead of internal fouling and cake layer can be more effectively removed by backwashing.

Furthermore, the chemicals used for both coagulation and chemical cleanings should be studied. The conditions in coagulation are optimal for conventional treatment, but different coagulants and different flocculation conditions have been proven to be more optimal for membrane filtration. Different cleaning chemicals should also be investigated as the chemicals used in this experiment are expensive and other chemicals have proven to be effective in other studies.

Selecting between granular media filtration and membrane filtration is still causing different opinions. Regardless, the clearest trend in particle separation is that membrane filtration is becoming more and more popular and the development of low pressure membranes is very rapid. Many studies have proven that membranes are efficient in removing particles and the removal is not affected by rapid changes in raw water. Turbidity is constantly under 0.1 FTU with both MF and UF membranes. In NOM reduction LPMs combined with coagulation can compete with conventional treatments.

Membrane materials are improving fast while becoming cheaper and it is only a matter of time before they are clearly a more economical option. As the land around current WTPs in Helsinki region is already effectively marked as an area of urban status, WTPs have no room to expand their buildings. This prevents building new sedimentation tanks

or any other type of expansion. Membrane filtration has a small footprint and would thus be a reasonable resolution that would satisfy all parties when the physical barriers of the current conventional treatment start to affect the treatment efficiency and water quality.

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## **Appendices**

Appendix 1. Operational data from different water treatment plants using MF/UF.

Appendix 2. Membrane data from different water treatment plants using MF/UF.

Appendix 3. Water quality from different water treatment plants using MF/UF.

Appendix 4. Membrane performance during the complete experiment.

Appendix 5. Raw data of water quality analyses.



## Appendix 1

Operational data from different water treatment plants using MF/UF. [1] Salehi et al., 2007. [2] Angers Loire Metropole, 2011. [3] Anonymous, 2012. [4] Pall, 2012. [5] Cromphout et al., 2011. [6] Almqvist et al., 2012. [7] Lozier, 2005. [8] Meier et al, 2006. [9] Skråvseth, A., 2009. [10] Lipp et al., 2003. [11] Panglisch et al., 2008. [12] City of Del Rio, 2004. [13] Najm et al., 2000 (pp. 29-45).

Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
Aalsterweg <sup>1</sup>	UF used for backwash treatment (Grit Chamber - Buffer - Microstrainer - UF - UV - Pure water cellar)	X-Flow	Period 60-70 s Flux 45 l/m <sup>2</sup> h	-	<b>CEB</b> Interval 3 h (HCl, H <sub>2</sub> O <sub>2</sub> )
Angers <sup>2</sup>	Ozonisation - Coagulation/Decantation - CAP added - UF - Chlorination - Storage	Aquasource	-	5000	-
Annen <sup>1</sup>	UF used for backwash treatment (Grit Chamber - Buffer - UF - UV - Pure water)	X-Flow	Period 45 s Flux 300 l/m <sup>2</sup> h	40	-
Anonymous 1 <sup>1</sup>	Groundwater - MF - Dechlorination	Memcor	<b>Air:</b> Interval 60 min Period 60 s <b>Hydraulic:</b> Interval 60 min Period 20 s Flux 60 l/m <sup>2</sup> h	1250	<b>CEB</b> (H <sub>2</sub> SO <sub>4</sub> , NaClO)
Anonymous 2 <sup>1</sup>	Groundwater - Pre-chlorination - MF	X-Flow	Interval 70 min Period 20-25 s Flux 250 l/m <sup>2</sup> h	189	<b>CEB</b> (HCl, NaOH)
Anonymous 3 <sup>1</sup>	Groundwater - Pre-chlorination - UF - Ozonation - GAC - Super chlorination - Dechlorination	-	<b>Air:</b> Interval 60 min Period 40 s <b>Hydraulic:</b> Interval 60 min Period 20-25 s	1758	<b>CIP</b> Interval 14 d (H <sub>2</sub> SO <sub>4</sub> )

Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
Anonymous 4 <sup>1</sup>	Groundwater - UF - Ozonation - GAC - Chlorination	X-Flow	Interval 150-200 min Period 50-60 s Flux 250 l/m <sup>2</sup> h	268	<b>CEB</b> Interval 18h (Caustic acid, HCl), Citric)
Anonymous 5 <sup>1</sup>	MF - Superchlorination	Memcor	<b>Air:</b> Interval 75 min Period 60 s <b>Hydraulic:</b> Interval 75 min Period 20 s Flux 60 l/m <sup>2</sup> h	1958	<b>CEB</b> (H <sub>2</sub> SO <sub>4</sub> , NaClO)
Anonymous 6 <sup>1</sup>	Clarification with PACL - GAC - Pre-chlorination - UF	X-Flow	Interval 150-200 min Period 50-60 s Flux 250 l/m <sup>2</sup> h	80	<b>CEB</b> Interval 18 h (Caustic acid, HCl, Citric)
Anonymous 7 <sup>1</sup>	Air Stripping - UF - Chlorination	X-Flow	Interval 50 min Period 50 s	208	<b>CEB</b> Interval 48 h (HCl) 96-110 h (NaOH)
Anonymous 8 <sup>3</sup>	Pre-ozonation - Coagulation - Flocculation - Sedimentation - Post-ozonation – Sand filtration - Microsieve - UF	Aquasource	Interval 45-60 min Period 30-40 s Flux 250-300 l/m <sup>2</sup> h	10417	<b>CEB</b> Every BW (NaClO) <b>CIP</b> Interval 160 d
Beverly Beach <sup>4</sup>	Well - MF - Storage	Pall	<b>Air:</b> Interval 60 min Period 120 s <b>Hydraulic:</b> Period 30 s	29.5	<b>CEB</b> Interval 60 min (Cl) <b>CIP</b> Interval 21-28 d
Chaparral <sup>1</sup>	Screen Strainer - Strainer - UF - GAC - Reservoir	Zenon	-	4750	-
Clay Lane <sup>1</sup>	Ozone - GAC - Strainers - Basin - Primary UF – Storage (UF also used for backwash treatment)	X-Flow (X-Flow)	Interval 40 -150 min Period 50 s	6667 (208)	<b>CEB</b> Interval 10-40 min (NaOH, Acid)

Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
de Gavers <sup>5</sup>	Canal - Nitrification - Flocculation/Direct filtration - Gavers lake - UF - GAC filtration - Storage (UF on one treatment line, flocculation/direct filtration on other)	-	Period 60 s Flux 250 l/m <sup>2</sup> h	UF: 333 WTP: 1375	<b>CEB</b> Interval 30 min, (NaOH+H <sub>2</sub> O <sub>2</sub> , H <sub>2</sub> SO <sub>4</sub> +H <sub>2</sub> O <sub>2</sub> ) <b>CIP</b> Interval 90 d (H <sub>2</sub> C <sub>2</sub> O <sub>4</sub> )
Dierfurt <sup>1</sup>	Well - Strainer - Ultrafiltration	INGE	Interval: 1-2 weeks	72	-
Ennerdale <sup>1</sup>	Microstrainer - MF - NaOCl - Contact basin - NaOH, Orthophosphate - Storage (UF also used for backwash treatment)	Memcor (Memcor)	Interval 60 min (Interval 30 min)	2458	<b>CIP</b> Interval 3 months (NaOH)
Gothenburg <sup>6</sup>	Precipitation - Sedimentation - GAC filter - UF - Lime, sodium, chlorine gas, and chlorine dioxide	X-Flow	Interval 60 min Period 35 s	7750	<b>CEB</b> Interval 24 h (NaOH, NaClO, H <sub>2</sub> SO <sub>4</sub> )
Granbury <sup>7</sup>	Intake - Softening - UF - RO - Clear Well	Hydranautics	-	-	<b>CEB</b> (ClO)
Heel <sup>1</sup>	UF used for backwash treatment (Filtration - Tank - UF - Basin)	X-Flow	Period 45 s Flux 147 l/m <sup>2</sup> h	240	<b>CEB</b> Interval 20 BW Period 240 s (HCl, H <sub>2</sub> O <sub>2</sub> )
Heemskerk <sup>1</sup>	Sieve - Coagulation - Sedimentation - Filtration - AC - UF - Reverse Osmosis	X-Flow	Period 30 s Flux 300 l/m <sup>2</sup> h	2000	<b>CEB</b> Interval 8 h (NaOCl)
Helmond <sup>1</sup>	UF used for backwash treatment (Grit Chamber - Buffer - UF - Filtration - UV - Pure water)	X-Flow	Period 60 s Flux 165 l/m <sup>2</sup> h	57	<b>CEB</b> (HCl, H <sub>2</sub> O <sub>2</sub> )
Hermeskeil <sup>8</sup>	Intake - Limestone filter - UF	-	-	140	<b>CEB</b> (HCl + NaOH)
Hydron Fevoland <sup>1</sup>	UF used for backwash treatment (UF - UV - Pure water cellar)	X-Flow	Period 35 s	-	<b>CIP</b> Interval 60 d (duration 20h)

Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
Iverness <sup>8</sup>	Intake - Micro strainer - Baffled Flocculation - Primary UF - Storage (UF also used for backwash treatment)	X-Flow (X-Flow)	Interval 40 min Period 45 s	1435	<b>CEB</b> Interval 109 BW (ClO (200 mg/l Cl <sub>2</sub> ), NaOH)
Keldgate <sup>8</sup>	Well - Pre-filtering - UF – Storage (UF also used for backwash treatment)	X-Flow	-	3750	<b>CEB</b> (NaOCl, HCl, NaOH)
Lello <sup>9</sup>	Pre-filtration - PAX added - UF - Chlorination - Storage	X-Flow	Interval 120 min	500	<b>CEB</b> Interval 8 BW (NaClO, H <sub>3</sub> PO <sub>4</sub> , NaOH)
Macharen <sup>1</sup>	UF used for backwash treatment (Grit chamber - Buffer - UF - NF - UV - Pure water)	X-Flow	Period 120 s Flux 200 l/m <sup>2</sup> h	-	<b>CEB</b> (HCl, H <sub>2</sub> O <sub>2</sub> )
Maennedorf <sup>1</sup>	Ozonation - AC filter - Strainer - UF	INGE	-	733	-
Manitowoc <sup>7</sup>	Intake - Strainer - MF - Clear Well	Memcor	Interval 30 min Period 150 s	1734	<b>CIP</b> Interval 4 d (NaOH, H <sub>2</sub> SO <sub>4</sub> )
Minneapolis <sup>8</sup>	Lime mixing, PAC, Potassium - Recarbonation - Storage - Ferric chloride coagulation - Flocculation - UF - Storage	X-Flow	Interval 25 min	11040	<b>CEB</b> when required (NaOCl, NaHSO <sub>3</sub> , HCl)
Muotathal <sup>8</sup>	Prefiltration - UF	Membratec	Interval 30 min	108	<b>CEB</b> Interval 8-24 h (NaOCl) <b>CIP</b> Interval 30-360 d (NaOH)
Nietap <sup>1</sup>	UF used for backwash treatment (Grit Chamber - UF - UV - Pure water cellar)	X-Flow	Period 20 s Flux 138 l/m <sup>2</sup> h	90	<b>CEB</b> (HCl, NaOH)
Nonnweiler <sup>10</sup>	Intake tower - Limestone filtration - UF - Disinfection	X-Flow	Interval 60 min period 40 s	54	<b>CEB</b> Interval 4-8 h (H <sub>2</sub> O <sub>2</sub> ) <b>CIP</b> (NaOH)



Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
Ouddorp <sup>1</sup>	Intake - Microstrainer - Filtration - Infiltration - Groundwater - Aeration - Filtration - AC Filter - UF -Storage	X-Flow	Period 5-25 s Flux 250 l/m <sup>2</sup> h	808	<b>CEB</b> Interval 90-120 min (NaOCl)
Pusan <sup>1</sup>	Coagulation - Sedimentation - Strainer - UF - Reverse Osmosis	INGE	-	340	<b>CEB</b> (NaClO)
Roetgen <sup>11</sup>	Intake - Pre-filtration - In-line Flocculation - Ultrafiltration - Limestone - Disinfection - Storage (UF also used for backwash treatment)	X-Flow (INGE)	Interval 60-70 min (Interval 25 min)	6000 (600)	<b>CEB</b> Interval 24 h (NaOH, acid) <b>Backwash CEB</b> Interval 48 h
San Felipe <sup>12</sup>	Well - Pre-filtering - UF - Storage (UF also used for backwash treatment)	Aquasource	Interval 60 min Period 30-75 s	2842	-
San Patricio <sup>1</sup>	Rapid mixing - Flocculation - Sedimentation - MF - Storage	Pall	Period 45 s	1250	<b>CEB</b> Cl <b>CIP</b> Interval 180 d (Caustic and citric solution)
Sandhurst <sup>7</sup>	Intake - Coagulation/Flocculation - MF - Ozonation - BAC filtration - Storage	Memcor	Interval 25 min Period 150 s	5250	<b>CEB</b> (NaOH)
Saratoga <sup>7</sup>	Strainer - MF - Chlorine disinfection	Memcor	Interval 22 min Period 150 s	788	-
Singapore <sup>8</sup>	Intake - Aeration - Pre-coagulation - Coagulation (alum) - UF - Ozonation - PAC Contactor - Chlorination - Storage	Zenon	Interval 20 min Period 30 s	7500	-
Spannenburg <sup>1</sup>	UF used for backwash treatment (Buffer - UF - Filter - Pure water cellar)	X-Flow	-	100	<b>CEB</b> Interval 24 h (HCl) 24 h (H <sub>2</sub> O <sub>2</sub> ) 96 h (NaOCl) <b>CIP</b> Interval 30-40 d

Plant	Treatment process	Membrane	Backwash	Capacity (m <sup>3</sup> /h)	Chemicals
Thyon <sup>8</sup>	Prefiltration - UF	Membrattec	Interval 30 min	70	<b>CEB</b> Interval 8-24 h (NaOCl) <b>CIP</b> Interval 30-360 d (NaOH)
Torgon <sup>8</sup>	Prefiltration - UF	Membrattec	Interval 30 min	40	<b>CEB</b> Interval 8-24 h (NaOCl) <b>CIP</b> Interval 30-360 d (NaOH)
Vigneux Sur Seine <sup>13</sup>	Pre-chlorination - Clarification/settling - GAC filtration - Ozonation - UF -Storage	-	-	-	-
Vionnaz <sup>8</sup>	Prefiltration - UF	Membrattec	Interval 30 min	80	<b>CEB</b> Interval 8-24 h (NaOCl) <b>CIP</b> Interval 30-360 d (NaOH)

## Appendix 2

Membrane data from different water treatment plants using MF/UF. [1] Salehi et al., 2007. [2] Angers Loire Metropole, 2011. [3] Anonymous, 2012. [4] Pall, 2012. [5] Cromphout et al., 2011. [6] Almqvist et al., 2012. [7] Lozier, 2005. [8] Meier et al, 2006. [9] Skrøvseth, A., 2009. [10] Lipp et al., 2003. [11] Panglisch et al., 2008. [12] City of Del Rio, 2004. [13] Najm et al., 2000 (pp. 29-45).

Plant	Membrane material	Nominal pore size	Filtration Mode	Filtration Flux (l/m <sup>2</sup> h)	TMP (bar)	Surface area (m <sup>2</sup> )	Manufacturer	Treatment Goal
Aalsterweg <sup>1</sup>	-	-	-	11,0-111,0	-	900	X-Flow UFC M5	-
Angers <sup>2</sup>	-	0.01	-	-	-	45000	Aquasource	Elimination of turbidity and microbiological retention.
Annen <sup>1</sup>	-	-	-	75	-	400	X-Flow UFC M5	-
Anonymous 1 <sup>1</sup>	-	-	-	31	0,4	50400	Memcor S10V	-
Anonymous 2 <sup>1</sup>	PVC	-	-	95	0,25	1680	X-Flow S-225-FSFC	Removal of <i>Cryptosporidium</i>
Anonymous 3 <sup>1</sup>	PVDF	-	-	60	0,23	12480	-	Removal of virus, bacteria, particles, ions
Anonymous 4 <sup>1</sup>	-	-	-	80,0-90,0	0,4	35,2	X-Flow S 225	Removal of <i>Cryptosporidium</i>
Anonymous 5 <sup>1</sup>	-	-	-	35	0,55	55440	Memcor S10V	Removal of <i>Cryptosporidium</i>
Anonymous 6 <sup>1</sup>	-	-	-	80-90	0,4	35,2	X-Flow S 225	Removal of <i>Cryptosporidium</i>
Anonymous 7 <sup>1</sup>	-	0.02	-	110	0,2-0,4	1680	X-Flow Xiga	Removal of virus and bacteria
Anonymous 8 <sup>3</sup>	PES (original CA)	0.01	Inside-Out	60,0-70,0	0,1-0,6	168000	Aquasource	Removal of turbidity, suspended matter, bacteria, and virus
Beverly Beach <sup>4</sup>	PVDF	0.1	Outside-In	-	2	-	Pall	-
Chaparral <sup>1</sup>	-	-	-	51	-	-	Zenon 500D	-
Clay Lane <sup>1</sup>	-	-	-	124	0,4-0,8	-	-	Removal of <i>Cryptosporidium</i> and viruses

Plant	Membrane material	Nominal pore size	Filtration Mode	Filtration Flux (l/m <sup>2</sup> h)	TMP (bar)	Surface area (m <sup>2</sup> )	Manufacturer	Treatment Goal
de Gavers <sup>5</sup>	-	-	-	44-54	-	-	-	-
Dierfurt <sup>1</sup>	-	-	-	80	-	-	INGE Dizzer 5000 MB	-
Ennerdale <sup>1</sup>	-	-	-	160 (62,0-100,0)	2	-	Memcor MF (Memcor UF)	-
Gothenburg <sup>6</sup>	-	0.02	-	102	0,26	-	X-Flow Xiga	Removal of virus and bacteria
Granbury <sup>7</sup>	PES	0.01	Inside-Out	-	0,3	12077	Hydranautics HydraCap	-
Heel <sup>1</sup>	-	-	-	36	-	1218	X-Flow	-
Heemskerk <sup>1</sup>	-	0.02	-	113	-	26880	X-Flow Xiga	Removal of turbidity and microbiology
Helmond <sup>1</sup>	-	-	-	65	-	840	X-Flow UFC M5	-
Hermeskeil <sup>8</sup>	-	-	Inside-Out	-	-	-	-	-
Hydron Fevoland <sup>1</sup>	-	-	-	67	-	240	X-Flow	-
Iverness <sup>8</sup>	-	-	-	69 (42,8)	-	20580 (3360)	X-Flow (X-Flow)	Removal of <i>Cryptosporidium</i> , turbidity, and colour
Keldgate <sup>8</sup>	PES	0.01	-	-	-	37000	X-Flow	Removal of Protozoa
Lello <sup>9</sup>	-	0.01	-	80	-	-	X-Flow	Removal of humus
Macharen <sup>1</sup>	-	0.02	-	50	-	540	X-Flow Xiga	-
Maennedorf <sup>1</sup>	-	-	-	110	-	-	INGE Dizzer 5000 MB	-
Manitowoc <sup>7</sup>	-	0.2	-	47,0-97,0	-	17550	Memcor	Removal of particulate matter and pathogens with focus on turbidity, coliform bacteria, <i>Giardia</i> , and <i>Cryptosporidium</i> .

Plant	Membrane material	Nominal pore size	Filtration Mode	Filtration Flux (l/m <sup>2</sup> h)	TMP (bar)	Surface area (m <sup>2</sup> )	Manufacturer	Treatment Goal
Minneapolis <sup>8</sup>	PES	0.02	Inside-Out	97	-	141120	X-Flow	-
Muotathal <sup>8</sup>	-	-	-	-	-	-	-	-
Nietap <sup>1</sup>	-	-	-	103	-	870	X-Flow	-
Nonnweiler <sup>10</sup>	PES	0.01	-	45	0,2-0,5	1400	X-Flow	-
Ouddorp <sup>1</sup>	-	0.02	-	120	-	6720	X-Flow Xiga	-
Pusan <sup>1</sup>	-	-	-	80	0,2	-	INGE Dizzer 5000 MB	-
Roetgen <sup>11</sup>	-	-	-	60 (65)	0,13 (0,3)	69120 (7020)	X-Flow (INGE)	-
San Felipe <sup>12</sup>	-	-	Inside-Out	-	1,4	-	Aquasource	-
San Patricio <sup>1</sup>	-	-	-	99	-	15000	Pall MF	-
Sandhurst <sup>7</sup>	PP	0.2	Inside-Out	36,67	0,2-0,8	143410	Memcor	Removal of turbidity and pathogen
Saratoga <sup>7</sup>	PP	-	Inside-Out	44,2	2,5	18110	Memcor 90M10C	-
Singapore <sup>8</sup>	-	-	-	68	-	13950	Zenon 1000	Removal of pathogen and algae
Spannenburg <sup>1</sup>	-	-	-	80	-	1290	X-Flow	-
Thyon <sup>8</sup>	-	-	-	-	-	-	-	-
Torgon <sup>8</sup>	-	-	-	-	-	-	-	-
Vigneux Sur Seine <sup>13</sup>	-	-	-	-	-	-	-	Removal of microorganisms
Vionnaz <sup>8</sup>	-	-	-	-	-	-	-	-



## Appendix 3

Water quality from different water treatment plants using MF/UF. [1] Salehi et al., 2007. [2] Angers Loire Metropole, 2011. [3] Anonymous, 2012. [4] Pall, 2012. [5] Cromphout et al., 2011. [6] Almqvist et al., 2012. [7] Lozier, 2005. [8] Meier et al, 2006. [9] Skråvseth, A., 2009. [10] Lipp et al., 2003. [11] Panglisch et al., 2008. [12] City of Del Rio, 2004. [13] Najm et al., 2000 (pp. 29-45).

Plant	Water Source	pH	TOC (mg/l)	UV <sub>254</sub> (cm <sup>-1</sup> )	Temperature (°C)	Alkalinity (mmol/l)	Turbidity (NTU)	Colour	TDS (mg/l)
Aalsterweg <sup>1</sup>	Surface	-	< 2.0	< 0.02	18.0	-	< 2.0	-	-
Angers <sup>2</sup>	Surface	5.0-6.5	-	-	-	-	-	-	-
Annen <sup>1</sup>	Ground	-	-	-	-	-	-	-	-
Anonymous 1 <sup>1</sup>	-	-	-	-	-	-	-	-	-
Anonymous 2 <sup>1</sup>	-	-	-	-	-	-	-	-	-
Anonymous 3 <sup>1</sup>	-	-	-	-	-	-	-	-	-
Anonymous 4 <sup>1</sup>	-	-	-	-	-	-	-	-	-
Anonymous 5 <sup>1</sup>	Surface	8.6	5.0	0.07	11.5	0.32	1.8	5.0 (CU)	134
Anonymous 6 <sup>1</sup>	Surface	-	-	-	-	-	-	-	-
Anonymous 7 <sup>1</sup>	Surface	7.6	-	-	10.1	0.17	0.7	5.8 (Hazen)	-
Anonymous 8 <sup>3</sup>	Surface	7.1	-	-	11.6	2.98	0.13	-	-
Beverly Beach <sup>4</sup>	Ground	-	-	-	-	-	-	-	-
Chaparral <sup>1</sup>	Surface	-	-	-	-	-	-	-	-
Clay Lane <sup>1</sup>	Surface	-	7.0	-	15.0	1	100	-	-
de Gavers <sup>5</sup>	Surface	8.1	2.9	-	-	1.51	8.0	-	-
Dierfurt <sup>1</sup>	Surface	7.6	2.9	-	-	-	0.03	-	-
Ennerdale <sup>1</sup>	Surface	7.0	-	-	10.0	-	-	-	-
Göthenburg <sup>6</sup>	Backwash	-	-	-	10.0	-	1100-1500 (FTU)	-	-

Plant	Water Source	pH	TOC (mg/l)	UV <sub>254</sub> (cm <sup>-1</sup> )	Temperature (°C)	Alkalinity (mmol/l)	Turbidity (NTU)	Colour	TDS (mg/l)
Granbury <sup>7</sup>	Backwash	-	-	-	-	-	-	-	-
Heel <sup>1</sup>	Backwash	-	4.5	-	12.5	-	-	-	-
Heemskerk <sup>1</sup>	Backwash	7.7-8.0	-	-	14.0	-	1 – 5 (FTU)	-	-
Helmond <sup>1</sup>	Backwash	8.0	-	-	11.0	-	434 (FTU)	-	-
Hermeskeil <sup>8</sup>	Backwash	7.0	-	-	11.0	-	0	-	-
Hydron Fevoland <sup>1</sup>	Backwash	8.0	-	-	11.0	-	-	-	-
Iverness <sup>8</sup>	Backwash	-	-	-	-	-	-	-	-
Keldgate <sup>8</sup>	Ground	-	-	-	11.0	-	0.8	-	-
Lello <sup>9</sup>	Ground	7.2	1.1	-	11.5	3.04	0.6	0	-
Macharen <sup>1</sup>	Ground	7.3	2.9	-	12.4	2.74	0.28	8.9	-
Maennedorf <sup>1</sup>	Ground/Surface	7.1	-	-	11.5	3.37	10.8	0.8	-
Manitowoc <sup>7</sup>	Ground	-	-	-	11.0	-	0.7	-	-
Minneapolis <sup>8</sup>	Ground/Surface	7.0	2.2	-	-	3.07	0.5	1.2 (Pt/Co)	-
Muotathal <sup>8</sup>	Ground	-	-	-	-	-	-	-	-
Nietap <sup>1</sup>	Surface	-	-	-	-	-	-	-	-
Nonnweiler <sup>10</sup>	Surface	-	-	-	-	-	-	-	-
Ouddorp <sup>1</sup>	Surface	8.0	-	-	-	-	7.6	23 (°Cr-Co)	-
Pusan <sup>1</sup>	Surface	7.2	5.4	-	7.7	0.31	-	-	-
Roetgen <sup>11</sup>	Surface	7.1	-	-	1.6 – 8.0	-	-	30 (mg Pt/l)	-
San Felipe <sup>12</sup>	Surface	7.0 – 9.7	3.0 – 8.0	-	5.0 – 25.0	-	5.0 – 100	-	-
San Patricio <sup>1</sup>	Surface	-	-	-	-	-	-	-	-
Sandhurst <sup>7</sup>	Ground	6.5 – 7.0	5.0	0.4	5.0 – 15.0	-	5.0	-	-
Saratoga <sup>7</sup>	Ground	-	<2.0	<0.02	18	-	2.0	-	-
Singapore <sup>8</sup>	Surface	-	-	-	-	-	1.0	-	-

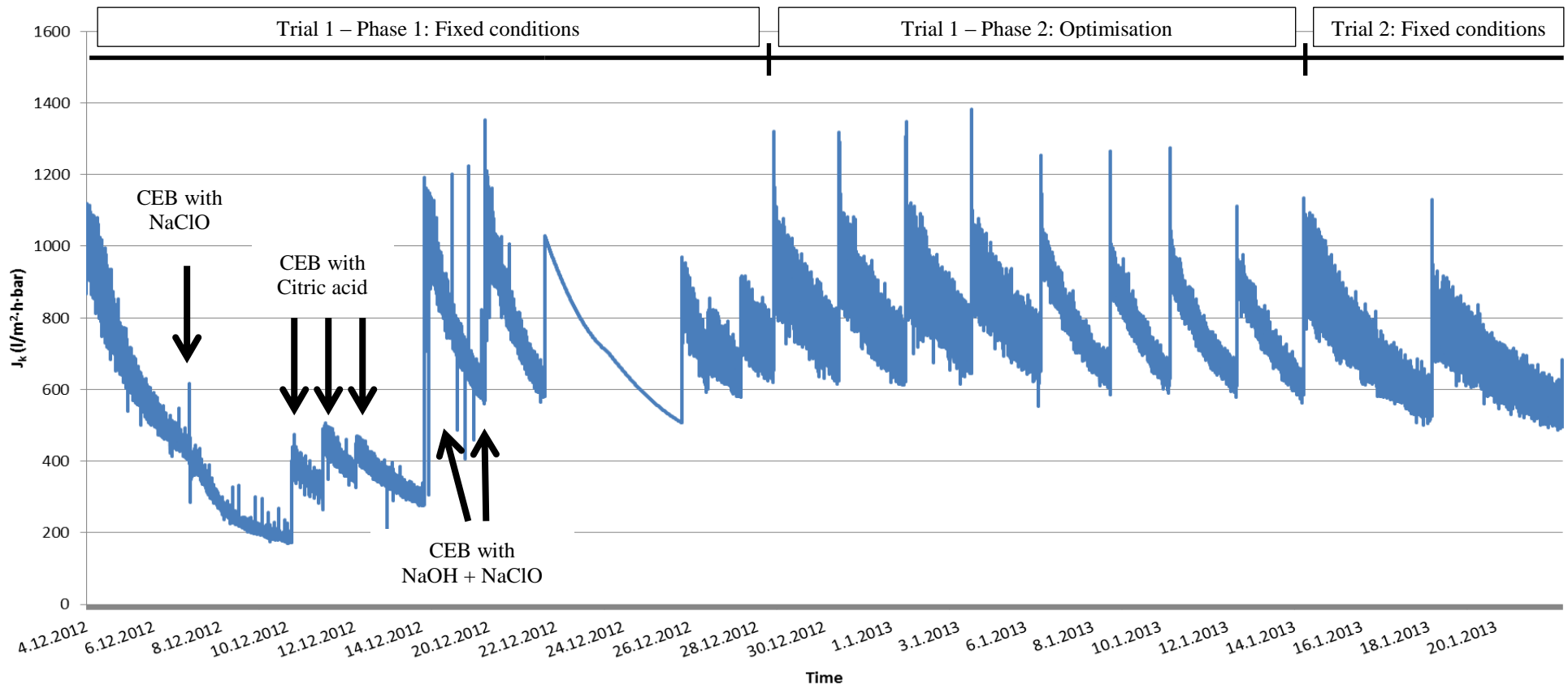


Plant	Water Source	pH	TOC (mg/l)	UV <sub>254</sub> (cm <sup>-1</sup> )	Temperature (°C)	Alkalinity (mmol/l)	Turbidity (NTU)	Colour	TDS (mg/l)
Spannenburg <sup>1</sup>	Surface	8.0	-	0.05-0.07	-	-	3.0 – 13.0	-	-
Thyon <sup>8</sup>	Surface	-	Low	-	5.0 – 25.0	-	5.0	-	-
Torgon <sup>8</sup>	Surface	7.6 – 8.1	0.6 – 1.8	-	3.0 – 18.0	1.1	1.0 – 100	-	-
Vigneux Sur Seine <sup>13</sup>	Surface	8.0	-	-	-	0.57	2.25	14.9	-
Vionnaz <sup>8</sup>	Surface	-	1.0 – 10	0.05 – 1.0	7.0 – 32.0	0.24 – 0.9	1.0 – 10.0	-	329 – 1752



## Appendix 4

Membrane performance during the complete experiment.

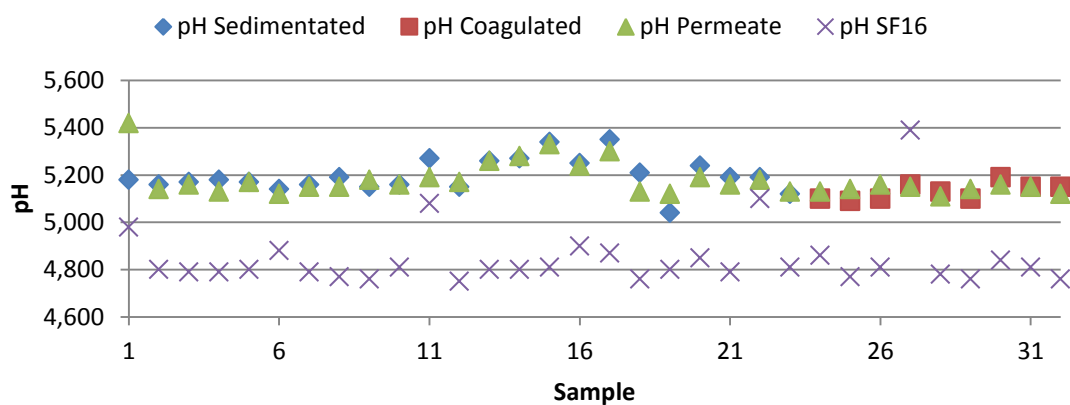




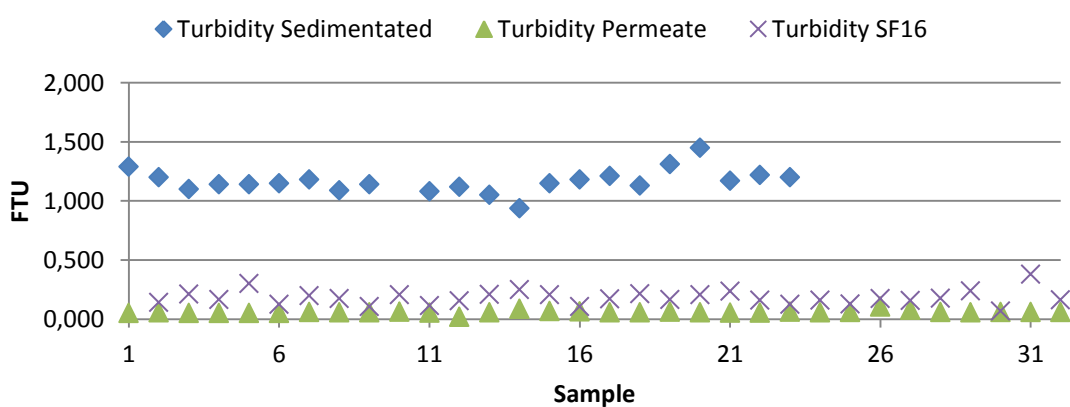
## Appendix 5

Raw data of water quality analyses.

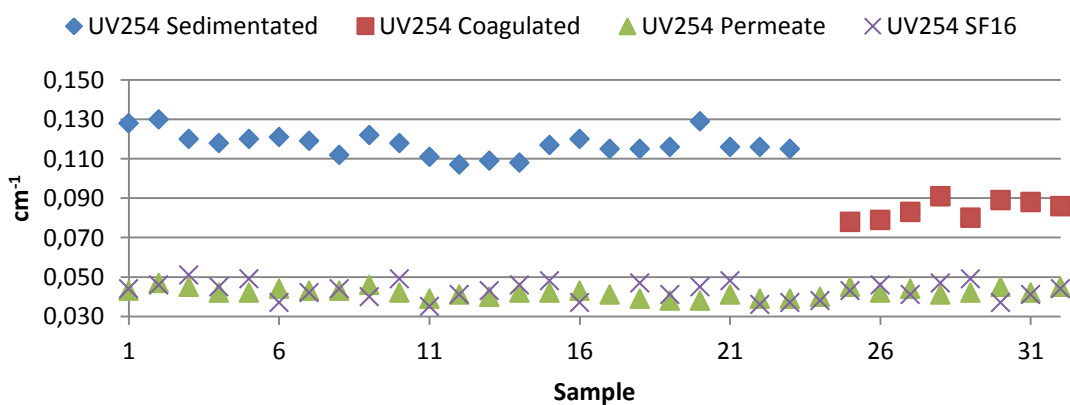
pH



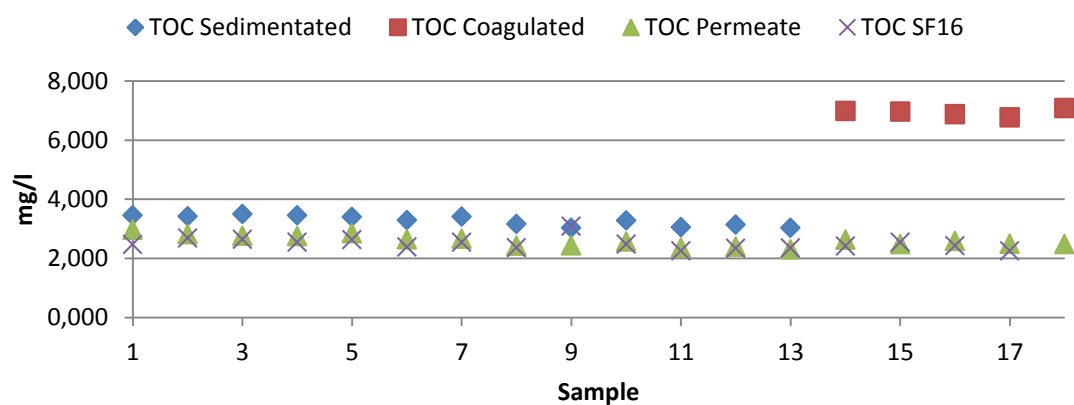
Turbidity



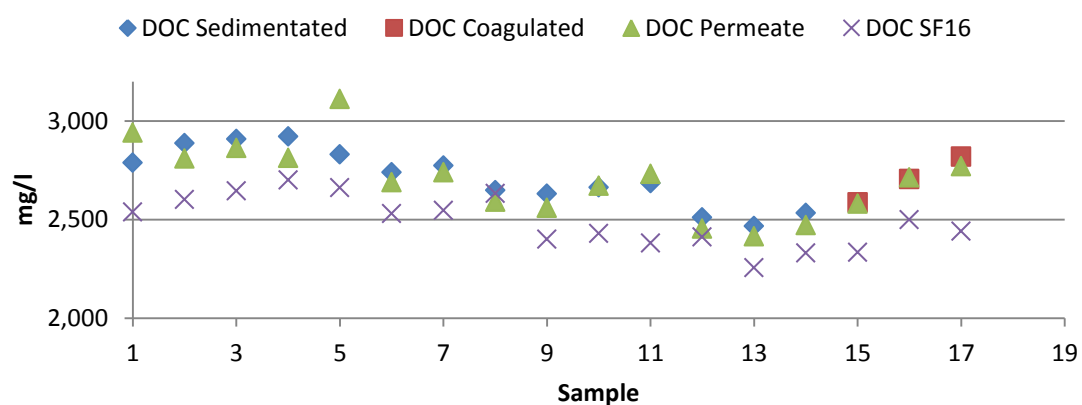
UV<sub>254</sub>



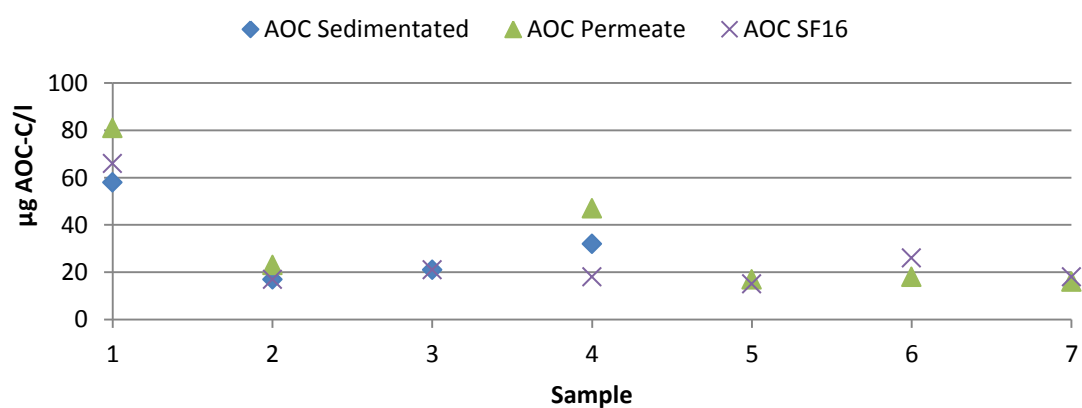
## TOC



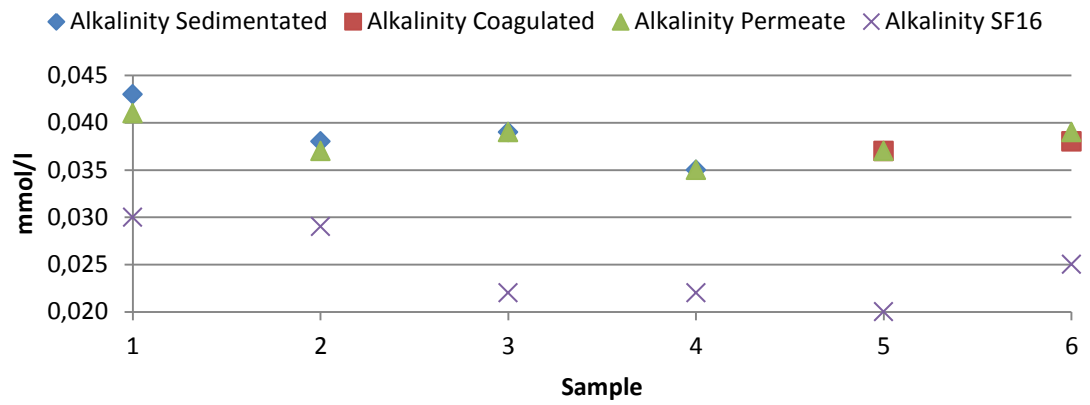
## DOC



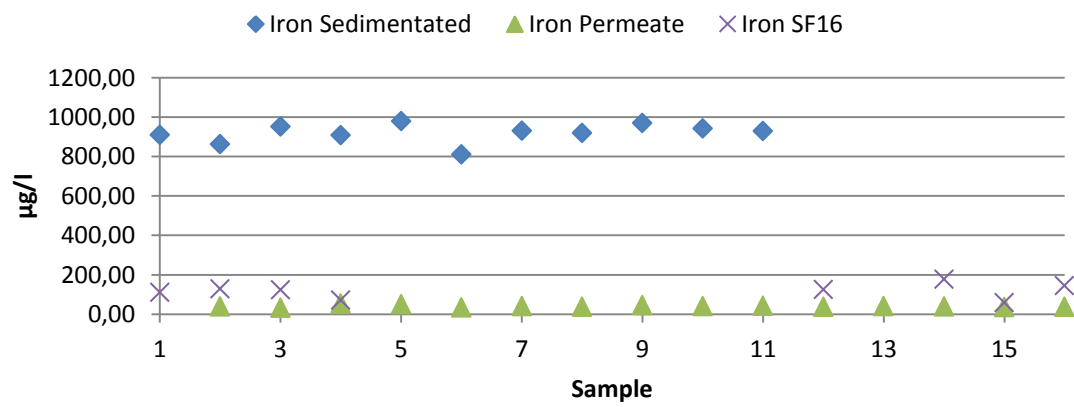
## AOC



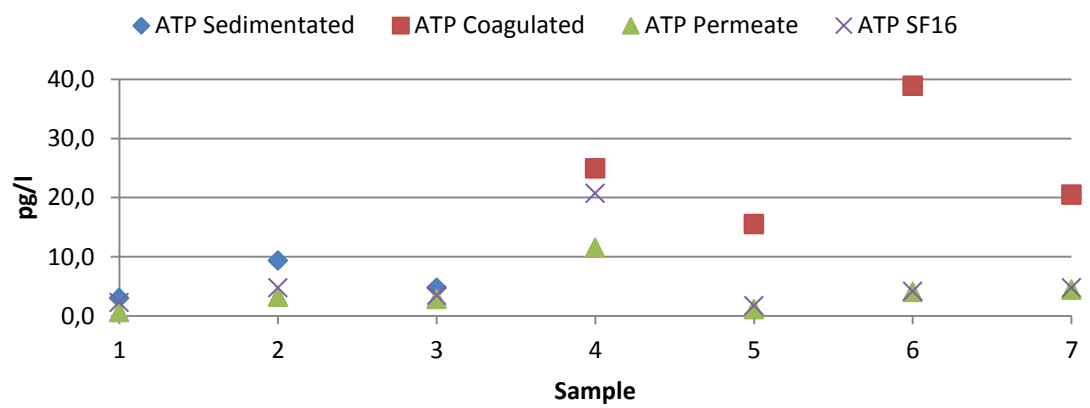
## Alkalinity



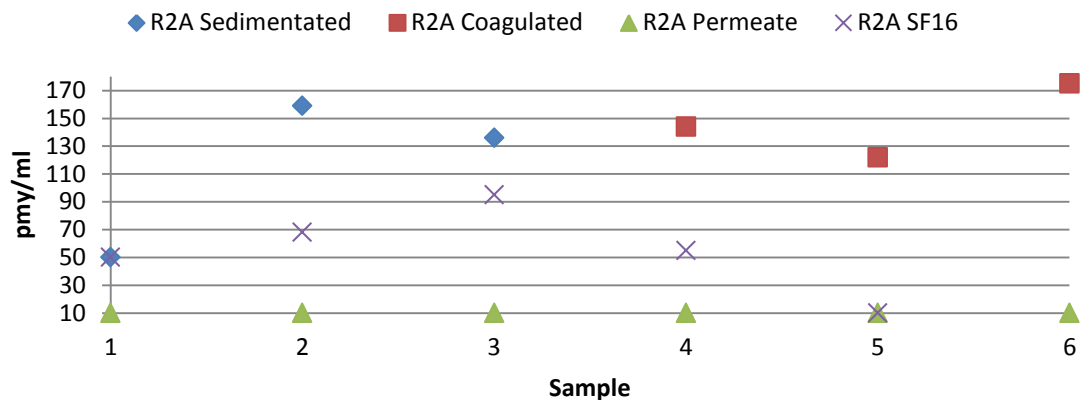
## Iron



## ATP



R2A



Example of LC-OCD result

