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# Optimisation of a small-scale ultrafiltration system

Separation of humic substances  
from surface water

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## Abstract

Norrvatten is the fourth largest drinking water producer in Sweden. They produce and distribute drinking water to 14 municipalities north of Stockholm. For some time, Norrvatten has studied alternative processes in order to optimise and increase the production capacity and efficiency of their drinking water treatment plant at Görvålnverket.

In this project, a small-scale pilot plant has been used to study and optimise a coagulation and ultrafiltration hybrid process step in order to remove humic substances from surface water. The effect of pH, reaction time, and different feed water qualities (Görvåln, Fyrisån and Görvåln full scale sand filtrate) were analysed through a series of experiments performed with the pilot plant. The most optimal placement of an eventual ultrafiltration step was also studied.

The results from the experiments suggested that pH in the range 6.1 to 6.7 had no large effect on the removal efficiency of fluorescent dissolved organic matter (fDOM). Instead, differences in removal efficiency could be observed between coagulant dosages which indicates that this parameter is of more importance than pH. At higher pH ( $> 7$ ) removal was significantly lower. The reaction time had no effect on the removal efficiency of the plant. However, the transmembrane pressure increased ten times faster during the experiments with a shorter reaction time. These results suggested that the floc formed were smaller and thereby more tightly packed in the ultrafilter which in turn increased the pressure in the membrane. An fDOM removal of at least 50 %, though no more than 60 %, was achieved with all feed waters except for the sand filtrate which had a removal efficiency of 18 %. However, highly concentrated humic waters such as Fyrisån proved to be challenging for the plant to handle since the pressure built up rather quickly in the membrane. The sand filtrate feed water experiments indicated that an ultrafiltration step after a sand filtration process would be effective. However, further studies are required to be able to determine the most optimal placement of the ultrafiltration process.

In conclusion, the results achieved with the pilot plant show promising signs of an ultrafiltration process being a viable alternative for Norrvatten to increase their drinking water treatment plant's efficiency and capacity.

## Sammanfattning

Norrvatten är den fjärde största dricksvattenproducenten i Sverige. De producerar och distribuerar dricksvatten till 14 kommuner norr om Stockholm. Norrvatten har under en tid studerat alternativa processer för att optimera och öka produktionskapaciteten och effektiviteten i deras dricksvattenreningsverk vid Görvälnverket.

I detta projekt har en pilotanläggning använts för att studera och optimera ett hybridprocessteg bestående av ett koagulerings- och ultrafiltreringssteg med syfte att avlägsna humusämnen från ytvatten. Effekten av pH, reaktionstid och olika matarvattenkvaliteter (Görväln, Fyrisån och sandfiltrat) analyserades genom en serie experiment utförda med pilotanläggningen. Den mest optimala placeringen av ett eventuellt ultrafiltreringssteg i reningsprocessen studerades även.

Resultaten från experimenten påvisade att pH mellan 6.1 och 6.7 inte hade någon effekt på reningen av fDOM. Mer signifikanta skillnader i rening kunde observeras mellan de olika koaguleringsdoserna, vilket indikerar att denna parameter är viktigare än pH. Reaktionstiden hade ingen märkbar effekt på anläggningens reningseffektivitet. Däremot ökade transmembrantrycket tio gånger snabbare under experimenten med den kortare reaktionstiden. Dessa resultat antyder att de bildade flocken var mindre och därmed blev mer tätt packade i ultrafiltret vilket i sin tur lett till det snabbt ökande trycket i membranet. En avskiljning av minst 50 %, men inte mer än 60 %, av det ingående fDOM kunde uppnås med alla matarvatten utom sandfiltratet, som hade en reningseffektivitet på 18 %. Vattnet från Fyrisån visade sig vara en utmaning för anläggningen att hantera, då trycket snabbt byggdes upp i membranet vilket tyder på att anläggningen inte klarar för starkt förorenade vatten. Resultaten från sandfiltratexperimenten visade att ett ultrafiltreringssteg efter sandfiltren skulle kunna vara en möjlig placering av ultrafiltren. Ytterligare studier krävs dock för att med säkerhet kunna bestämma den mest optimala placeringen.

Sammanfattningsvis visar resultaten från experimenten lovande tecken på att en ultrafiltreringsprocess är ett möjligt alternativ för att öka Norrvattens dricksvattenreningsverks effektivitet och kapacitet.

## Populärvetenskaplig sammanfattning

Att vatten är viktigt vet vi alla. Men innan vattnet når våra kranar, duschar och toaletter behöver det renas från bland annat virus, bakterier och organiskt material. Norrvatten är Sveriges fjärde största vattenreningsverk och renar vatten åt 700 000 människor norr om Stockholm. Med växande samhällen och ökande mängder föroreningar i vatten, ställs vattenverken inför nya utmaningar. Norrvatten har under en längre tid jobbat på att öka sitt vattenverks kapacitet och effektivitet för att möta dessa förändringar.

Man har bland annat undersökt möjligheten att integrera ett ultrafiltrationssteg i reningsprocessen för att förbättra avlägsnandet av organiskt material från vattnet. Ett ultrafilter består av många små porer som vatten och andra små ämnen kan passera, medan större partiklar och delar av organiskt material separeras bort. Med en ultrafiltreringsprocess hoppas man kunna minska belastningen på andra delar av reningsverkets processer och även minska halterna organiskt material i det renade vattnet.

En ultrafilterpilotanläggning sattes ihop och användes för att undersöka effekten av pH, reaktionstid och olika vattenkvaliteter. Effektiviteten bestämdes utifrån mängden avlägsnat organiskt material. Resultaten visade att pH värden över 7 påverkade effektiviteten av pilotanläggningen negativt. Vid pH mellan 6.1 och 6.7 kunde ingen effekt på reningen av organiskt material observeras. En reaktionstid på 150 sekunder visade sig resultera i mer gynnsamma förhållanden än en reaktionstid på 0 sekunder. Försöken med olika typer av vatten visade att pilotanläggningen klarar att avlägsna upp till 60% av det ingående vattnets halt av organiskt material. Däremot blev anläggningen överbelastad vid försöken med starkt förorenade vatten.

Överlag visade resultaten från experimenten lovande tecken på att en ultrafiltreringsprocess är ett möjligt alternativ för att öka Norrvattens reningsverks effektivitet och kapacitet.

## Table of Contents

<b>Acknowledgements</b> .....	<b>i</b>
<b>Abstract</b> .....	<b>ii</b>
<b>Sammanfattning</b> .....	<b>iii</b>
<b>Populärvetenskaplig sammanfattning</b> .....	<b>iv</b>
<b>List of Tables</b> .....	<b>vii</b>
<b>List of Figures</b> .....	<b>viii</b>
<b>Abbreviations</b> .....	<b>ix</b>
<b>1. Introduction</b> .....	<b>1</b>
1.1 Aim.....	1
1.2 Solving method.....	1
1.3 Delimitations .....	1
<b>2. Technical background</b> .....	<b>2</b>
2.1 The purification process at Görvålverket .....	2
2.2 Surface water quality .....	3
2.2.1 Natural organic matter .....	3
2.2.2 Natural organic matter in drinking water treatment plants .....	4
2.3 Membrane filtration of surface water.....	6
2.3.1 Classification of membranes .....	6
2.3.2 Membrane fouling.....	6
2.3.4 Crossflow and dead-end mode.....	7
2.4 Coagulation and ultrafiltration hybrid process .....	8
2.4.1 Conventional coagulation.....	8
2.4.2 In-line coagulation.....	8
2.4.3 Operating parameters .....	9
2.5 Integration of membrane filtration in treatment processes.....	10
<b>3. Experiments</b> .....	<b>11</b>
3.1 Development of the pilot plant .....	11
3.2 Operation of the pilot plant .....	12
3.2.1 Pre-treatment of feed water .....	12
3.2.2 Main treatment of feed water .....	13
3.2.3 Permeate analysis and data management.....	14
3.3 Maintenance of the pilot plant .....	14
3.3.1 Backwashing .....	15
3.3.2 Storage.....	15
3.4 Optimisation and evaluation of operating parameters.....	15

3.4.1 pH .....	16
3.4.2 Reaction time .....	16
3.5 Comparison of different feed waters .....	17
3.5.1 Fyrisån .....	17
3.5.2 Sand filtrate .....	18
3.5.3 Blend of Fyrisån & sand filtrate .....	18
3.5.4 Görväln .....	18
<b>4. Results and discussion .....</b>	<b>19</b>
4.1 Optimal operating parameters.....	19
4.1.1 pH .....	19
4.1.2 Reaction time .....	22
4.2 Influence of feed water .....	23
4.3 Placement of the ultrafiltration process .....	26
4.4 Verification of data.....	27
4.5 Experimental uncertainties and future studies.....	29
<b>5. Conclusion.....</b>	<b>31</b>
<b>6. References .....</b>	<b>32</b>
<b>7. Appendices.....</b>	<b>35</b>



## List of Tables

<i>Table 1: A compilation of all experiments conducted for the optimisation of pH and coagulant dosage. ....</i>	<i>16</i>
<i>Table 2: A compilation of the experiments conducted for the evaluation of the effect of reaction time. ....</i>	<i>16</i>
<i>Table 3: A compilation of experiments conducted for the evaluation of the effect of different feed waters. ....</i>	<i>17</i>
<i>Table 4: A compilation of experiments conducted with Fyrisån water. ....</i>	<i>17</i>
<i>Table 5: A compilation of the experiments conducted with sand filtrate. ....</i>	<i>18</i>
<i>Table 6: A compilation of the experiments conducted with a 2:1 blend of Fyrisån water and sand filtrate. ....</i>	<i>18</i>
<i>Table 7: A compilation of the experiments conducted with Görväln water. ....</i>	<i>18</i>
<i>Table 8: A compilation of experiments conducted with Görväln water and aluminium sulphate as coagulant... </i>	<i>18</i>
<i>Table 9: The operating conditions and fDOM removal for the pH experiments.....</i>	<i>19</i>
<i>Table 10: The operating conditions and fDOM removal for the reaction time experiments.....</i>	<i>22</i>
<i>Table 11: A compilation of the experiments where either the achieved fDOM levels were lower than 32, or more than 50 % of the initial fDOM was removed. ....</i>	<i>25</i>

## List of Figures

<i>Figure 1: Purification process at Görvålnverket. ....</i>	<i>2</i>
<i>Figure 2: An illustration of the pilot plant system. ....</i>	<i>11</i>
<i>Figure 3: An illustration of the effect of sulfuric acid and coagulant on the pH of raw water. ....</i>	<i>12</i>
<i>Figure 4: The effect of pH and coagulant dosages on fDOM removal ....</i>	<i>20</i>
<i>Figure 5: The effect of alkalinity and coagulant dosages on fDOM removal ....</i>	<i>21</i>
<i>Figure 6: The effect of pH on the TMP slope for the different coagulant dosages.....</i>	<i>21</i>
<i>Figure 7: The effect of reaction time on fDOM removal.....</i>	<i>22</i>
<i>Figure 8: Shows the effect of reaction time on the TMP escalation.....</i>	<i>23</i>
<i>Figure 9: The remaining fDOM for the different coagulant dosages and feed waters ....</i>	<i>23</i>
<i>Figure 10: The fDOM removal in percentages for the different feed waters ....</i>	<i>24</i>
<i>Figure 11: SUVA against coagulant concentration for the different feed waters ....</i>	<i>26</i>
<i>Figure 12: The correlation between experimental alkalinity data and laboratory or theoretical values. ....</i>	<i>27</i>
<i>Figure 13: The correlation between experimental UV data and laboratory or theoretical values.....</i>	<i>27</i>
<i>Figure 14: The correlation between the theoretical and measured coagulant dosages. ....</i>	<i>28</i>
<i>Figure 15: The correlation between experimental pH data and laboratory or theoretical values. ....</i>	<i>28</i>
<i>Figure 16: Correlation between fDOM and UVA254 ....</i>	<i>29</i>

## Abbreviations

Al	Aluminium
ALG	aluminium sulphate
ALK	alkalinity
cDOM	coloured DOM
CFF	crossflow filtration
DEF	dead-end filtration
DOC	dissolved organic carbon
DOM	dissolved organic matter
DWTP	drinking water treatment plant
fDOM	fluorescent dissolved organic matter
GAC	granular activated carbon
MQ	milli-Q water
NOM	natural organic matter
NTU	nephelometric turbidity units
PAC	polyaluminium chloride
SUVA <sub>254</sub>	specific ultraviolet absorbance at 254 nm
TMP	transmembrane pressure
TOC	total organic carbon
UF	ultrafilter
UV <sub>254</sub>	ultraviolet absorbance at 254 nm

## 1. Introduction

This thesis project has been carried out at Norrvatten which is the fourth largest drinking water producer in Sweden. They produce and distribute drinking water to 14 municipalities north of Stockholm. For some time, Norrvatten has studied processes that could potentially optimise and increase the production capacity and efficiency of their drinking water treatment plant (DWTP) at Görvålnverket. In particular, studies on the integration of membrane filtration have been conducted in order to reduce the amount of humic and other organic substances in the water, and also to reduce the heavy load on subsequent carbon filters.

### 1.1 Aim

The aim of this project is to gain more knowledge about the implementation of an ultrafiltration (UF) process step in the water treatment plant at Görvålnverket. More precisely, the questions at issue are:

- Optimisation and evaluation of the effect of pH and reaction time.
- Evaluation and comparison of the efficiency of the UF with different feed waters.
- Evaluation of the most optimal placement of the UF.

### 1.2 Solving method

To evaluate and understand the operation of the UF pilot plant, several experiments were conducted where the effect of operating parameters and feed water qualities were studied.

### 1.3 Delimitations

- Characteristics of natural organic matter (NOM) was not analysed.
- Seasonal variabilities in water quality were not taken into consideration due to the time frame of the project.

## 2. Technical background

The feed water at Norrvatten, Görvålverket, is surface water collected from lake Mälaren which is Sweden's third largest lake. Norrvatten produce about 1 600 litres of water per second which is distributed to the 700 000 people living in the 14 municipalities that receive their water from Görvålverket. The feed water at Görvålverket goes through several treatment processes before being distributed.

### 2.1 The purification process at Görvålverket

The purification process of the water at Görvålverket have been illustrated in Figure 1. The feed water first passes through a micro strainer (1) where fish, algae and other larger particles are removed. The water is then led to a mixer (2), where aluminium sulphate coagulant is added. Next, the water is led to a flocculator (3) where the suspended particles in the water clump together to form so called flocs. The flocs bind to humic substances, soil particles, microorganisms and more. Once the flocculation process is finished, the water travels to a sedimentation basin (4) where the flocs sink to the bottom and are separated from the main water flow. The water is then first filtered through a sand filter (5) and then a filter with granular activated carbon (6) as the media. UV-reactors (7) are then used to disinfect the water. Lastly, it is pH adjusted (8) to make it slightly basic before distribution. (Norrvatten a n.d.)

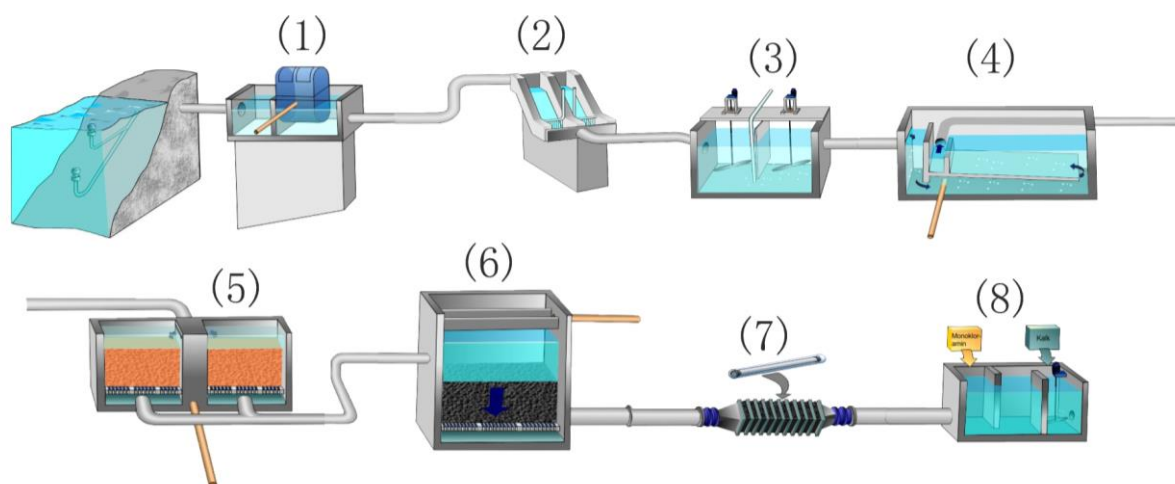


Figure 1: Purification process at Görvålverket.

## 2.2 Surface water quality

Norrvatten continuously conducts analyses to make sure that the drinking water from their plant meet the quality standards which are set by the Swedish Food Agency. Some common parameters that are assessed by drinking water facilities are total organic carbon (TOC), dissolved organic carbon (DOC), ultraviolet absorbance ( $UV_{254}$ ), pH, turbidity, and fluorescence. (Sillanpää, Matilainen, and Lahtinen 2015)

Alkalinity is another water quality that is important since it is an indicator of the buffering capacity of water at pH in the basic range. Water with high alkalinity has a good buffering capacity and does not easily change in pH. The pH of low alkalinity water, on the other hand, can quickly and easily be changed when acid or base are added. (Fernandez 2018)

### 2.2.1 Natural organic matter

Considering that Norrvatten uses lake Mälaren as their water source, the feed water contains a fair amount of natural organic matter (NOM) since surface waters contain higher levels of organic matter than groundwater (Löfgren 2003). A large portion of NOM, about 35 % to 70 % depending on the water source, is composed of humic substances which arise when plants and animals decompose in the water cycle. Waters containing high amounts of humic substances are often characterised by a yellow-brownish colour. The remaining portion of NOM consists of non-humic substances, these substances and their contribution to the yellow-brown colour is almost insignificant. (Machenbach and Ødegaard 2007)

**Humic substances.** Humic substances are hydrophobic biopolymers and are of great importance for the acidity of lakes and watercourses since many surface waters have a pH below 6 due to these substances (Swedish University of Agricultural Sciences, n.d.). They are grouped into three categories depending on their solubility in aqueous bases and acids: fulvic acids, humic acids and humin. Fulvic acids is the dominating type, they are soluble at any pH, and give waters a light yellow-brown colour. Humic acids have a grey-brown colour, are soluble in basic conditions, and precipitate at a pH below 2. Humin are black in colour and are insoluble in water. (Machenbach and Ødegaard 2007)

The colour intensity can be used as an indicator of the chemical properties of the humic substances since they are related to the molecular weight and carbon content, which increase as

the colour intensity increases, and the oxygen content and solubility, which decrease as the colour intensity increases. (Machenbach and Ødegaard 2007)

### 2.2.2 Natural organic matter in drinking water treatment plants

Trends are showing that organic matter in the surface waters of the world, including lake Mälaren in Sweden, are steadily increasing. In Mälaren, the concentration of organic substances have shown to follow a cyclic pattern due to environmental factors such as the turnover rate and internal processes in the lake (Köhler 2021). Increased concentrations of organic substances likely have a negative effect on drinking water purification since it is well known that NOM give water an undesired colour, taste, and odour. However, studies are still inconclusive as to whether or not they have an effect on human health (Ødegaard 2002). Drinking water treatment processes will therefore have to adapt to the new conditions since the current ones might not be efficient enough. (Köhler et al. 2016)

NOM reduces the efficiency of membranes by causing fouling which results in more frequent backwashing and cleaning of the membranes. In cases where activated carbon is used, NOM competes for adsorption sites with other contaminants (Machenbach and Ødegaard 2007). In coagulation processes, the need for coagulant dosage is mainly driven by the NOM content. Disinfection processes are also affected by the presence of NOM as they cause disinfection by-products to be formed and increase the disinfectant dose. These are some of the reasons as to why NOM has become an important variable and should be measured and removed. (Matilainen, Vepsäläinen, and Sillanpää 2010)

**Measurement of NOM.** The amount of humic substance in a water source can be measured in many ways (Löfgren, 2003). The TOC/DOC content is an important water quality variable that is often measured to assess the concentration of NOM in the water since they are the most convenient parameters to measure. (Sillanpää, Matilainen, and Lahtinen 2015)

Another common method to measure NOM is by UV-light adsorption. Studies have indicated that UV absorbance is correlated to humic substances (Keucken et al. 2017). UV-light adsorption is especially useful since it measures the ratio of hydrophobic NOM to hydrophilic NOM, which provides information about the ratio of humic to non-humic substances. A specific UV-absorbance (SUVA) higher than 4 indicates hydrophobic (humic) material, and a SUVA lower than 3 indicates mainly hydrophilic (non-humic) material. SUVA is the UV absorbance at 254 nm divided by the DOC concentration. Research has suggested that coagulation, in order

to remove NOM, will likely be effective on waters with a high SUVA, which is useful for determining what NOM-removal method to implement. (Sillanpää, Matilainen, and Lahtinen 2015)

Fluorescence can be used to measure fluorescent dissolved organic matter (fDOM) which is the fraction of coloured DOM (cDOM) that fluoresces when excited by UV radiation (Wang et al. 2020). cDOM is the coloured fraction of DOM which mainly consists of humic substances. (Kida et al. 2018)

Turbidity is an optical measurement of the relative clarity of water. It measures particles, organic matter, cDOM and other microorganisms that causes cloudiness of the water (U.S. Geological Survey n.d.). The turbidity is measured in nephelometric turbidity units (NTU) and should be less than 0.5 NTU in the outgoing drinking water according to Swedish drinking water quality standards. (Livsmedelsverket 2015)

**Removal of NOM.** There are several treatment processes that may be implemented to remove NOM. The processes that are currently considered the most feasible from an economic standpoint are coagulation and flocculation followed by sand filtration. These processes remove most of the NOM, but some is still left which is why more efficient alternatives are being investigated.

Ozonation coupled with biofiltration is an option for NOM removal. During ozonation, higher molecular weight NOM are split into lower molecule weight compounds which increases the fraction of more easily degradable organic carbon. The compounds causing discolouration of the water are also destroyed during this process. Ozonation is often followed by a biofiltration process where the organic carbon compounds are removed. (Brugger, n.d.)

A sorption process using a media such as granular activated carbon (GAC) is another viable option for NOM removal. Studies have shown that the addition of GAC to a conventional treatment process can substantially increase the removal of intermediate and low molecular weight compounds. (Marais et al. 2018) However, high molecular weight compounds are not efficiently removed during this process which is believed to be due to the large molecules not being able to access the internal pore structure of the GAC. (Velten et al. 2011)

However, in recent years, especially membrane-based drinking water processes has been of interest. Membranes are diverse and can therefore be chosen to fit the needs of the specific



water source (Lidén 2016). Membrane filtration can be implemented into water treatment plants both for the removal of NOM and also to decrease the load on subsequent purification steps in the process. (Ødegaard 2002)

### 2.3 Membrane filtration of surface water

Membrane filtration for water treatment was first introduced in the 1960's and has since the 1990's been tested in many drinking water treatment plants. These plants have partly been designed for the removal of natural organic matter (NOM) which is especially needed in countries with cold climates and soft surface waters. Some areas that are typically affected by this are Scandinavia, Russia, and Northern Great Britain. Colour removal, which also suggests the removal of humic substances, is therefore an important part of water treatment plants in these areas. (Ødegaard 2002) (Thorsen 1999)

#### 2.3.1 Classification of membranes

Membrane filtration can be used to separate a broad range of molecular weights and sizes, from larger particulates to dissolved organics and salts.

**Microfiltration.** The membranes with the largest pore size, microfilters, have pores larger than 0,1 µm and function similarly to the more traditional sieve. They remove suspended particles, large bacteria, and algae. (Cassano and Basile 2013)

**Ultrafiltration.** UF have pore sizes ranging from 1 nm to 100 nm. The separation that can be achieved with a UF is therefore highly dependent on the pore size. This type of membrane removes finer particles than MF and also viruses and large molecules. (Cassano and Basile 2013)

**Nanofiltration.** Nanofiltration generally have a nominal pore size of 1 nm and separate even smaller molecules. However, they are often characterised by their ability to separate salts rather than their pore size. (Lidén 2016)

#### 2.3.2 Membrane fouling

Membrane fouling is still an issue in the operation and applications of membranes in water treatment plants. Fouling is the accumulation of impurities such as particulates and compounds on the surface or within the pores of the membrane which eventually decreases the permeability.

Membrane fouling has many adverse effects of the efficiency of treatment plants, it affects the permeate quality, flux, operating cost and shortens the lifetime of the membrane. (Arhin et al. 2016)

Membrane fouling can be caused by a variety of source of fouling. Known types of fouling include biofouling, inorganic fouling, organic fouling, and particulate fouling. Biofouling is the formation of a biofilm on the membrane surface caused by organisms such as algae. Organic fouling is believed to be caused by NOM. There is still a lot of on-going research on how to control fouling caused by NOM since it is very common in natural waters and is very diverse. The diversity of NOM makes it very difficult to compare the studies made on the fouling mechanisms of NOM. In-organic fouling is mainly caused by particles containing metals. Lastly, particulate fouling is caused by suspended solids such as clay and soil. (Arhin et al. 2016)

Many of the previously mentioned treatments in *2.2.2 Influence of natural organic matter in DWTPs under Removal of NOM* are effective for mitigation and prevention of fouling since they remove NOM which is the main foulant of surface water.

#### 2.3.4 Crossflow and dead-end mode

There are two possible ways to configure the flow in membrane filtration; (1) crossflow filtration (CFF) and (2) dead-end filtration (DEF).

In CFF, the feed passes across the membrane instead of going through the filter. The direction of the flow prevents the accumulation of particles on the membrane by washing away the filter cake. This is especially useful for feed water that is highly turbid. However, since the UF step is often later in the process, this advantage may not come in handy due to already low turbidity. (Keucken et al. 2017)

Most often, DEF is applied in drinking water treatment plants rather than CFF, meaning the feed water enters on one side of the membrane and exits on the other side. In these applications, a filter cake may be formed on the feed side of the UF, caused by the accumulated particles and organic matter that do not pass through the membrane. Regular backwashing, normally 2 times per hour, of a UF is therefore often a requirement to prevent the filter cake from becoming too compact. (Lidén 2016)

## 2.4 Coagulation and ultrafiltration hybrid process

Hybrid processes where coagulation is coupled with membranes have become useful for complying with the quality standards of drinking water. Coagulation prior to a UF has shown to increase the permeate quality and reduce membrane fouling (Keucken et al. 2017). There are mainly two ways in which coagulation can be integrated; (1) conventional coagulation where coagulated organic matter is settled before the membrane or (2) in-line coagulation where coagulated organic matter is separated on the membrane surface. (Guigui et al. 2002) (Bergamasco et al. 2011)

### 2.4.1 Conventional coagulation

Conventional coagulation has been the most widely used in water treatment plants and involves flocculation and settling of the formed flocs. The coagulant is added into the feed water at the beginning of the process and rapidly mixed, the mixing is then slowed down to a slow-mixing to allow flocculation which lets the flocs settle. The process uses gravity to remove most of the flocs and any remaining flocs are removed from the feed by subsequent filtration. This process is effective for removal of both NOM and turbidity. (Department of Environment and Conservation 2011)

### 2.4.2 In-line coagulation

Membrane filtration in combination with coagulation has been shown to have the potential to prevent fouling and decrease coagulant doses. Especially in-line coagulation -the use of coagulants without flocculation or sedimentation- has shown benefits such as enhanced NOM removal and reduced resistance caused by the fouling layer. However, the removal efficiency of NOM is highly dependent on the coagulant, membrane type, feed water, and coagulation and filtration conditions such as pH, concentration, and reaction time. The process must therefore be optimised and designed with regards to these operating parameters. (Blankert, Betlem, and Roffel 2007)

Recently, in-line coagulation has been used more widely since, compared to conventional coagulation, this process does not require flocs to settle which in turn reduces the footprint of the process and coagulant dose. The conventional coagulation process is also more energy intensive than the in-line coagulation. (Keucken et al. 2017)

### 2.4.3 Operating parameters

Several conditions and parameters have been shown to affect the efficiency of coagulation and UF hybrid processes, especially the coagulant dosage, pH, reaction time, mixing speed and temperature. (Arhin et al. 2016) (Dayarathne et al. 2020)

**Coagulant dosage.** There are mainly two approaches that can be taken when deciding on the dosage for a coagulation process, either optimised coagulation or sweep flocculation. Sweep flocculation occurs when an excessive amount of coagulant is added to create large flocs. However, optimised coagulation has shown to be more effective since it uses exact dosing of the coagulant to neutralise and destabilise the particles' charges to allow flocculation (Department of Environment and Conservation 2011). A previous study, executed at Norrvatten in 2020, showed that the optimal coagulant dosage for a coagulation-UF process was 6 mg/L for waters with a TOC content of 8.5 mg/L. However, for water with a higher TOC content of 15.5 mg/L, the dosage had to be increased to about 8 mg/L. (Voightländer 2020)

**pH.** Several studies have shown how a change in pH can affect the chemistry and efficiency of coagulation processes, floc formation, and also how a specific pH can reduce membrane fouling (Arhin et al. 2016)(Feng et al. 2015). However, a change in pH does not only affect coagulation, but it also affects the characteristics of NOM. For instance, by affecting its solubility in water. Aluminium based coagulants are generally most effective in removing NOM at around pH 6, which have been confirmed by recent studies where the optimal pH for coagulation and NOM removal was at pH 5.5-6.5 (Yan et al. 2008) and 5-6.5 (Dayarathne et al. 2020). In a study where the pH was adjusted from 4 to 9, the most effective NOM removal was achieved at pH 6. Fouling was more serious in acidic conditions due to the flocs formed being denser and less porous. (Feng et al. 2015)

The coagulant in a coagulation process has an effect on the resulting pH. The type of coagulant has shown to be an important factor that should be taken into consideration when designing a coagulation process. Aluminium sulphate (alum) has been the most commonly used coagulant in DWTPs. However, an increased use of poly aluminium chloride (PAC) can be seen since this type of coagulant has many benefits when compared to the more traditional alum. For instance, PAC has higher basicity, lower residual levels of aluminium in the permeate, effective in a wider pH range, consume less alkalinity, and the increase of chloride is lower than the respective increase in sulphates from alum (Gebbie 2001). PAC is also Since the distributed

water from Görvålnverket has a pH of 8.2-8.4, a higher operating pH is preferred as it reduces the consumption of chemicals such as acid and base. (Norrvatten b n.d.)

**Reaction time.** The reaction time is the time that the feed water spends in a specific process. For coagulation processes, a study has shown that the most optimal reaction time for slow-mixing is at 15 minutes and for rapid-mixing it is at 2 minutes. However, it has also been shown that the intensity of mixing could be of higher importance than the time, which could have an effect on the optimal reaction time. (Zhang et al. 2013)

**Temperature.** Several studies have shown temperature to have an impact on the coagulation process. At lower temperatures, the formed flocs are often weaker due to less motion of particles at low temperatures, which results in lower collision energy and less collisions between particles. This, in turn, leads to higher dosages of coagulant being required. However, the TOC removal reportedly has not been significantly affected by low temperatures. Low molecular substances were easier to remove at higher temperatures. Temperature may be a parameter that affects coagulation but controlling this parameter is not sufficient. However, it is still necessary to be aware of the seasonal effects it might have on the effectiveness of the treatment process. (Dayarathne et al. 2020)

## 2.5 Integration of membrane filtration in treatment processes

A membrane filtration step can be implemented in several places along the purification process. Ultrafiltration is often used as a pre-treatment step with the purpose of removing organic matter from the following process steps. However, for treatment of feed water with high concentrations of organic matter, a UF could be placed later in the process for further reduction of NOM. (Lidén 2016)

It could preferably be placed after a traditional filtration process such as sand filtration, mainly to control membrane fouling and to improve water quality. Coupling sand filtration and UF has shown to significantly reduce the irreversible membrane fouling and NOM was also reduced further. (Guo et al. 2018)

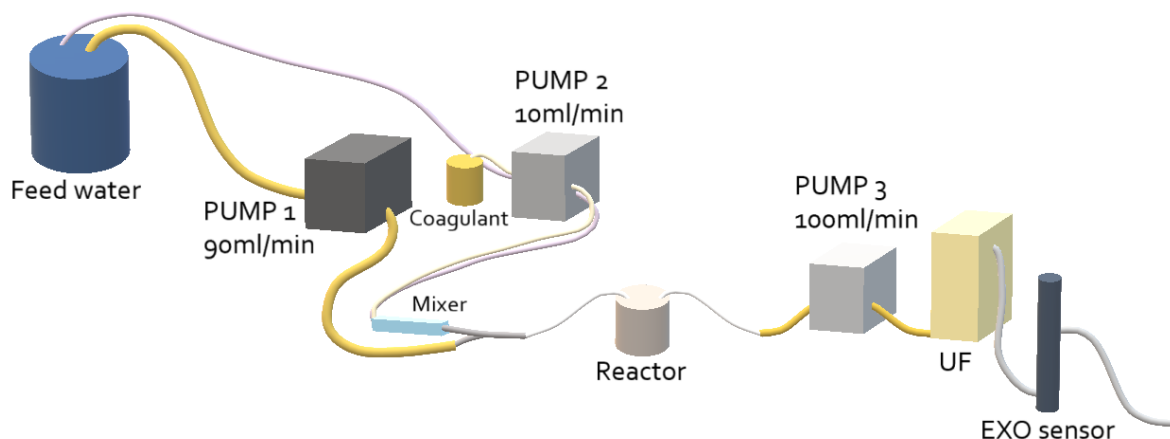
A study performed in Japan showed that GAC can effectively be used as a pre-treatment to UF in order to prevent irreversible fouling of the membrane. The frequency of chemical cleaning could also be reduced with the coupling of GAC and UF. (Tsujimoto et al. 1998)

### 3. Experiments

The experiments were conducted with a UF pilot plant that was assembled at the beginning of the project. The following chapters have addressed the development, operation, and maintenance of the pilot plant. Additionally, all experiments that have been executed have been described in words and compiled in tables.

#### 3.1 Development of the pilot plant

The pilot plant was assembled in two main steps. The first step was to prepare all the necessary tubing for the plant. The second step was to assemble the plant in the same way as it had been in a previous study (Voigtländer 2020) performed at Norrvatten. Assembling of the pilot plant was done by connecting the pumps, membrane, and reactor, to create the whole system as is illustrated in Figure 2 below. Pictures of the real-life plant at Norrvatten have been compiled in *Appendix A*.



*Figure 2: An illustration of the pilot plant system.*

The tubes from pump 2 were led to a mixing tube with a three-way connector on the outflow end. Next, the tube from pump 1 was connected to the three-way connector. These two streams were then combined and led to the reactor, where the coagulation process would take place. After coagulation, the stream was led to pump 3 before reaching the ultrafilter. After filtration, the permeate was led to an EXO sensor for analysis.

## 3.2 Operation of the pilot plant

Once the pilot plant had been reassembled, the experiments were conducted. The operation of the UF pilot was divided into three parts: (1) pre-treatment of feed water, (2) purification of feed water, and (3) analysis of permeate. The pre-treatment consisted of pH adjustment and coagulant solution preparation. The purification process consisted of the plant itself. The analysis part consisted of the EXO sensor and laboratory analyses.

### 3.2.1 Pre-treatment of feed water

The pre-treatment of the feed water consisted of pH adjustment and preparation of coagulant solutions. The final pH was determined by both the additions of acid/base and the coagulant dosage. The estimated requirement of a 0.05 M sulfuric acid and coagulant were based on titration curves produced in PHREEQC before the experiments were conducted. The values from PHREEQC were given in units that were converted to millilitres of  $\text{SO}_4$  per litre for the sulfuric acid, and milligrams of aluminium for the coagulant. For conversion calculations, see *Appendix B*.

The coagulant dosages of interest were between 4.5-24.5 mg/L of feed water. The coagulant was diluted 100 times in the plant, considering the flow rate of coagulant in the plant being 1 mL/min and the total flow rate being 100 mL/min. Coagulant solutions were therefore prepared 100 times more concentrated than required. The solutions were prepared a maximum of a couple hours before use. For a description of how the solutions were prepared, see *Appendix B*.

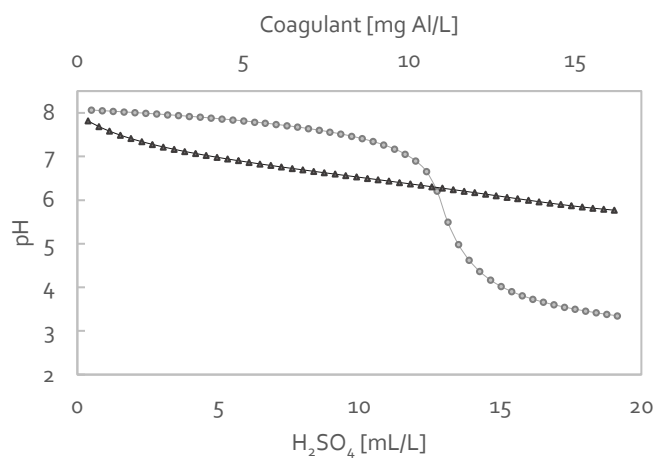


Figure 3: An illustration of the effect of addition of ● sulfuric acid and ▲ coagulant on the pH of raw water. With an initial temperature of 1.4 °C, TOC level of 8 mg/L and a pH of 8.1.

As can be seen in Figure 3, higher coagulant dosages result in lower pH values. The experiments using high dosages were therefore pH adjusted with a 0.1 M sodium carbonate, in order to avoid pH values below 6.2 in the membrane.

### 3.2.2 Main treatment of feed water

The main treatment of the water consisted of the plant itself where coagulation and filtration took place. Depending on the experiment that was conducted, the procedure could vary. However, the general procedure that was followed is as follows.

- Raw water was pH adjusted and prepared in 5 L containers for the experiments.
- Coagulant solutions at a desired dilution were prepared.
- The water quality of the feed water was measured by filling the EXO sensor and measuring for approximately 5 minutes.
- A flow of feed water through the pilot plant was established, whilst bypassing the membrane. Once no more air bubbles could be seen in the plant, the feed water was led through the membrane.
- Flow rates for the coagulant solution and the total flows were checked volumetrically by using a measuring cylinder. If necessary, pump heads were adjusted accordingly. Pictures of the pumps, and the process of calibrating them, have been attached in *Appendix C*.
- The pilot plant was then ready for the experiments. The coagulant tube was moved from the feed water to the coagulant solution and the tubes in the feed water were moved to the pH adjusted feed water.
- Measurements were taken with 20 second intervals on the EXO sensor, and the chosen experimental conditions for dosing were run for about 40 minutes for each experiment. Starting and ending times were noted.
- Once 35 minutes had passed, a 0.5 L permeate sample was taken for the Norrvatten laboratory.
- The total flow rate was checked at the beginning of every experiment and the TMP was also monitored. Swiping of the EXO sensor was automatically performed intermittently.



- A backwash was performed when the TMP reached over a certain pressure. At the end of a day, the membrane was prepared for storage with a chemically enhanced backwash (CEB). Both procedures are explained in more detail in *3.3 Maintenance of the pilot plant*.

### 3.2.3 Permeate analysis and data management

Analysis of the permeate was done with the EXO sensor, and permeate and selected raw water samples were sent to the lab for additional analysis for parameters such as pH, alkalinity, absorbance, and TOC, chloride, and sulphate content. The pH, absorbance and alkalinity values from the lab results and the EXO sensor were compared for further verification.

The sonde measures fDOM using a fluorescence signal. This signal is affected by both NOM content and temperature.

**Correction for temperature.** The fDOM data from the EXO sensor were corrected for temperature through the following formula:

$$fDOM_{tcorr} = fDOM_{QSU} + fDOM_{QSU} \cdot 0.012 \cdot (T_{feed\ water} - 25)$$

**Correction for absorbance.** The fDOM data was then also corrected for any absorbed light due to coloured water through the following formula:

$$fDOM_{abscorr+tcorr} = fDOM_{tcorr} + fDOM_{tcorr} \cdot (0.2508 \cdot UV_{254}/100)$$

The transmembrane pressure was another operating parameter that was used for the analysis of the results and the efficiency of the plant. The TMP slope, which describes how much the pressure increases per minute of operation, have been used in order to compare the experiments, see Appendix D. The slope was produced by calculating the total increase in pressure and dividing it by the operated time in minutes, according to the following formula:

$$TMP_{slope} = \frac{TMP_{final} - TMP_{initial}}{t_{end} - t_{start}}$$

## 3.3 Maintenance of the pilot plant

The maintenance of the pilot plant consisted of regular backwashes and thorough cleaning, that were performed in order to preserve it and prevent permanent fouling.

### 3.3.1 Backwashing

Backwashing of the membrane was performed with Milli-Q water at least once every 2 hours, depending on the pressure build-up. At the beginning, when the membrane was still new, a backwash was performed once the pressure reached a value of over 0.6 bar. Over time, the pressure in the membrane built up quicker and therefore, the backwashes were performed once the pressure reached values above 0.9 bar.

The backwash was performed by first disconnecting the permeate outlet of the membrane from other parts of the pilot plant. Then, the backwashing was performed by intermittently increasing the pressure to about 0.7 bar for efficient removal of particles stuck to the membrane. Once no more particles were seen in the outflow, the backwashing was considered complete.

If a backwash was performed in the middle of an ongoing experiment, the water in the membrane was first allowed to be replaced with the feed water before being connected to the outflow of the EXO sensor. This was done in order to not disrupt the measurement by mixing Milli-Q water with the feed water in the EXO sensor.

### 3.3.2 Storage

Prior to storage, at the end of each day of use, a standard chemical enhanced backwash was performed in the following steps:

- A Milli-Q backwash was performed according to the previous description, until no more flocs were released.
- A CEB with 1 litre of 150-200 ppm sodium hypochlorite was performed.
- Lastly, a final backwash with Milli-Q water was performed to flush out the hypochlorite and any other residues that might be left.

## 3.4 Optimisation and evaluation of operating parameters

Experiments have been conducted to determine how the efficiency of the UF-pilot is affected by parameters such as pH and reaction time, in order to find the most optimal operating conditions.

### 3.4.1 pH

Experiments were conducted with raw water from Görvåln, and at pH values around 6.0-7.5 which were achieved by the combined effect of the added amount of coagulant and acid/base. The reaction time for all experiments was 150 seconds which was achieved with a reactor volume of 250 mL. These experiments were conducted in order to find the most optimal combination of acid and coagulant dosage, with regards to the remaining fDOM in the permeate. These experiments should give an indication of how pH and alkalinity affect the purification efficiency.

The following experiments were conducted with varying amounts of acid and coagulant dosages, as can be seen in Table 1, with the aim to analyse if part of the added coagulant was consumed only for reducing the pH instead of acting as a coagulant. The sulfuric acid or bicarbonate were added to the 10 L containers with feed water, and mixed beforehand.

Table 1: A compilation of all experiments conducted for the optimisation of pH and coagulant dosage.

<b><i>Experiment</i></b>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]	Na <sub>2</sub> CO <sub>3</sub> [mL]
<i>pH experiment 1</i>	4.5	40, 50, 60	-
<i>pH experiment 2</i>	6.5	25, 35, 45	-
<i>pH experiment 3</i>	8.5	0, 17, 30	-
<i>pH experiment 4</i>	6.5	-	5, 15, 20

### 3.4.2 Reaction time

The reaction time experiments were conducted with raw water from Görvåln, and the reaction time was adjusted by using either a 250 mL reactor or in absence of a reactor. All experiments were executed with a fixed coagulant dosage of 6.5 mg Al/L and H<sub>2</sub>SO<sub>4</sub> additions of 25, 35, and 45 mL, see Table 2.

Table 2: A compilation of the experiments conducted for the evaluation of the effect of reaction time.

<b><i>Experiment</i></b>	Reaction time [s]	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]
<i>Reaction time experiment 1</i>	0	6.5	25, 35, 45
<i>Reaction time experiment 2</i>	150	6.5	25, 35, 45

### 3.5 Comparison of different feed waters

In these experiments, a comparison of water from Görväln, Fyrisån and the sand filtrate at Norrvatten has been performed. An overview of the experiments conducted in this part of the project can be seen in Table 3 below. All experiments were conducted with a fixed reaction time of 150 seconds and a pH of 6.2-6.8.

Table 3: A compilation of experiments conducted for the evaluation of the effect of different feed waters.

<b>Experiment</b>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]	Na <sub>2</sub> CO <sub>3</sub> [mL]
<i>Fyrisån</i>	6.5-24.5	0-45	5, 10, 15
<i>Sand filtrate</i>	3.8	-	0, 5, 15
<i>Fyrisån &amp; sand filtrate</i>	10.5, 12.5	30, 35	-
<i>Görväln</i>	4.5, 6.5, 8.5	30, 35, 50	-
<i>Görvaln (ALG)</i>	3.8-9.1	9, 22, 30	5, 15

#### 3.5.1 Fyrisån

Due to the presence of particles, the water from Fyrisån was filtered through a sand filter before being used in the pilot plant, see *Appendix A* for pictures and a description of how the filtration was performed. The experiments have been listed in Table 4 below.

Table 4: A compilation of experiments conducted with Fyrisån water.

<b>Fyrisån</b>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]	Na <sub>2</sub> CO <sub>3</sub> [mL]
<i>F1</i>	6.5	45	-
<i>F2</i>	8.5	40	-
<i>F3</i>	10.5	35	-
<i>F4</i>	12.5	30	-
<i>F5</i>	14.5	0	-
<i>F6</i>	16.5	0	-
<i>F7</i>	18.5	0	-
<i>F8</i>	20.5	-	5
<i>F9</i>	22.5	-	10
<i>F10</i>	24.5	-	15

### 3.5.2 Sand filtrate

The sand filtrate experiments were conducted with a fixed coagulant dosage of 3.8 mg Al/L. The sand filtrate from the purification process at Norrvatten has already been pH adjusted, these experiments have therefore been conducted with Na<sub>2</sub>CO<sub>3</sub> additions as can be seen in Table 5.

Table 5: A compilation of the experiments conducted with sand filtrate.

<i>Sand filtrate</i>	Coagulant [mg/L]	Na <sub>2</sub> CO <sub>3</sub> [mL]
<i>S1</i>	3.8	0
<i>S2</i>	3.8	5
<i>S3</i>	3.8	15

### 3.5.3 Blend of Fyrisån & sand filtrate

These experiments were conducted with a 2:1 blend of Fyrisån water and sand filtered water from Norrvatten. The two experiments have been compiled in Table 6 below.

Table 6: A compilation of the experiments conducted with a 2:1 blend of Fyrisån water and sand filtrate.

<i>Fyrisån &amp; sand filtrate</i>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]
<i>FS1</i>	10.5	35
<i>FS2</i>	12.5	30

### 3.5.4 Görvältn

For the experiments with feed water from Görvältn, previously conducted experiments were used. The experiments that were used can be seen in Table 7 below.

Table 7: A compilation of the experiments conducted with Görvältn water.

<i>Görvältn</i>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]
<i>G1</i>	4.5	50
<i>G2</i>	6.5	35
<i>G3</i>	8.5	30

The measurements in the last experiment were conducted with aluminium sulphate as coagulant, instead of polyaluminium chloride. A compilation of the experiments can be seen in Table 8 below.

Table 8: A compilation of experiments conducted with Görvältn water and aluminium sulphate as coagulant.

<i>Görvältn (ALG)</i>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> [mL]	Na <sub>2</sub> CO <sub>3</sub> [mL]
<i>GA1</i>	3.8	30	-
<i>GA2</i>	4.5	22	-
<i>GA3</i>	5.7	9	-
<i>GA4</i>	7.6	-	5
<i>GA5</i>	9.1	-	15

## 4. Results and discussion

Results from the experiments are illustrated in tables and diagrams, in the same order as they were presented in the previous section. In order to estimate the pilot plant's full-scale applicability, the full-scale plant data have been used as a starting point for the evaluation of the results, with the goal of reaching the same efficiencies of the full-scale plant. The results from all experiments have been compiled in tables in *Appendix E*.

### 4.1 Optimal operating parameters

Data gathered from the experiments conducted in this part of the project could give an indication of how pH, coagulant dosage and reaction time affects the efficiency of the pilot plant. The efficiency has mainly been judged based on the fDOM removal which is in QSU units if not indicated otherwise.

#### 4.1.1 pH

With the results from the four pH experiments, the combined and individual effect of pH and coagulant dosage was analysed. In Table 9 below, the results from the two measurements with the lowest and highest operating pH, within each experiment, have been compiled. The table indicate the remaining fDOM in the permeate and the fDOM removal in percentages.

*Table 9: The operating conditions and fDOM removal for the pH experiments.*

<b>Experiment</b>	Coagulant [mg/L]	pH	fDOM in permeate	fDOM removal [%]
<i>pH experiment 1</i>	4.5	6.1	42.3	36
		6.5	43.2	33
<i>pH experiment 2</i>	6.5	6.2	31.6	49
		6.5	33.0	47
<i>pH experiment 3</i>	8.5	6.3	30.6	55
		6.7	33.0	51
<i>pH experiment 4</i>	6.5	7.0	39.2	38
		7.6	50.0	23

A coagulant dosage of 4.5 mg/L, with an acid addition of 16 mL, resulted in an fDOM removal of 33 %, and an acid addition of 60 mL resulted in a removal of 36 %. The acid addition was thus almost four times larger, with a pH ranging from 6.5 to 6.1, and resulted in a relatively small change of 3 % more removal. The same pattern could be seen for pH experiment 2 and 3. This indicates that the pH does not notably affect the fDOM removal in the pH range (6.1-

6.7) that could be analysed in these experiments. Therefore, it is preferable so stay as close to the upper limit as possible since this will reduce the chemicals needed for the UF and also for the last pH adjustment step when increasing the pH before distribution.

More significant differences in fDOM removal could be seen between the coagulant dosages. Over 10 % more fDOM was removed with a coagulant dosage of 6.5 and 8.5 mg/L, compared to the results from the experiments with a coagulant dosage of 4.5 mg/L. However, not much of a difference could be seen between 6.5 and 8.5 mg/L. The 8.5 mg/L dosage resulted in an average of 5 % more removal. This indicates that the optimal coagulant dosage is reached near 6.5 mg/L. Any additional coagulant above this concentration is thereby not efficiently used to reduce fDOM. This can be verified by looking at the coagulant efficiency, which is the amount of fDOM removed per mg Al/L. The higher this value, the better, since this indicates that the coagulant has been used more efficiently. This value decreases from 4.7 to 3.6 when going from 6.5 mg/L to 8.5 mg/L.

pH experiment 4 was conducted with additions of a basic solution instead of an acidic. The results show that the fDOM removal efficiency decreased as the alkalinity and pH increased. In pH ranges around 7 and above, the pH and alkalinity seem to have a negative effect on the fDOM removal, as can be seen in Figure 4 and 5 below.

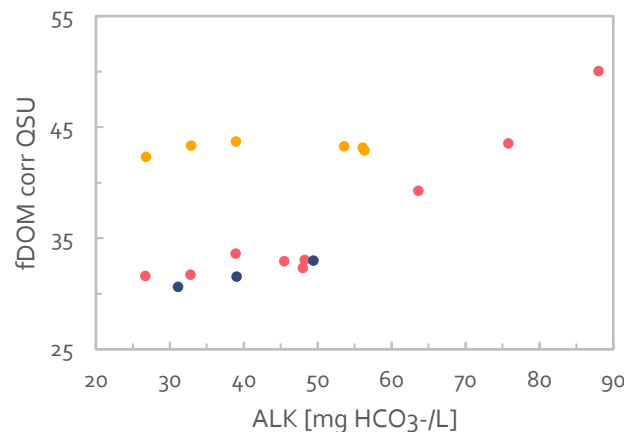


Figure 4: The effect of pH and coagulant dosages on fDOM removal (● 4.5 mg/L, ● 6.5 mg/L, ● 8.5 mg/L).

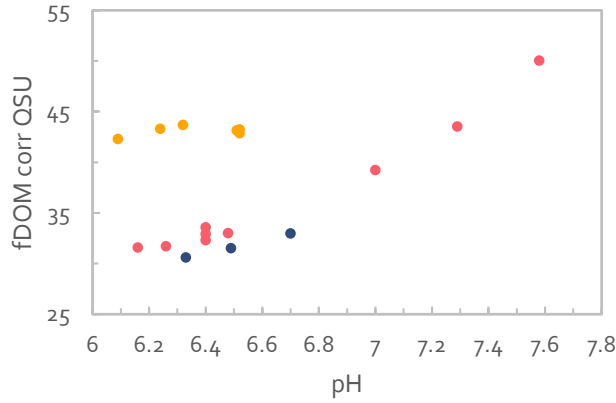


Figure 5: The effect of alkalinity and coagulant dosages on fDOM removal (● 4.5 mg/L, ● 6.5 mg/L, ● 8.5 mg/L).

The fDOM levels for each respective coagulant dosage are quite similar irrespective of the pH, therefore, it seems the removal of fDOM is not affected by pH. In basic conditions, on the other hand, the fDOM removal seems to be correlated to the pH since the fDOM removal efficiency decreases as the pH rises. The results from pH experiment 1-4 thus verify previous studies on the subject, which show that coagulation is most effective in slightly acidic pH values. At these pH values, the dominant coagulation mechanism, charge neutralisation between the coagulant and NOM, is the most effective.

The TMP increase rate was not correlated to either coagulant dosage or pH in acidic conditions, see Figure 6. However, in basic pH ranges, it increased as the pH increased. This is believed to be correlated to the higher fDOM levels in the permeate which suggests a less effective coagulation. From an economical aspect, lower pH values are preferred in order to reduce the frequency of backwashes which consume chemicals, in addition to being time consuming and decreasing the efficiency of the pilot plant.

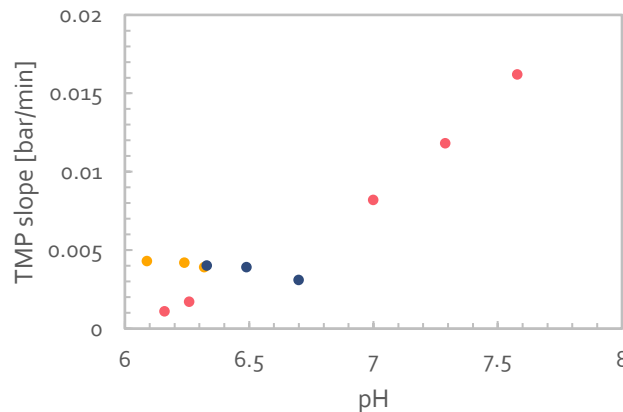


Figure 6: The effect of pH on the TMP slope for the different coagulant dosages (● 4.5 mg/L, ● 6.5 mg/L, ● 8.5 mg/L).



#### 4.1.2 Reaction time

The results from the reaction time experiments showed similar removal efficiencies, as can be seen in Table 10 and Figure 7. Experiment 1 had a 6-8 % higher removal efficiency than experiment 2. However, the TMP increased ten times faster in those experiments, see Figure 8. This could be due to the fact that the flocs did not have as favourable conditions to form and grow. During the measurements in experiment 2, flocs were allowed to form and grow for some time in a reactor.

Table 10: The operating conditions and fDOM removal for the reaction time experiments.

<b>Experiment</b>	Reaction time [s]	pH	fDOM in permeate	fDOM removal [%]	Average TMP slope
<i>Reaction time experiment 1</i>	0	6.1	30.0	54	0.0139
		6.4	30.0	54	
<i>Reaction time experiment 2</i>	150	6.2	31.6	49	0.0014
		6.4	33.6	46	

Meanwhile, in the experiments without a reactor, there was no time for mixing, which could have resulted in the coagulant being less efficient in forming good flocs. The floc formed would not have the prerequisites to form and grow, which could have resulted in less and smaller flocs being formed. Smaller flocs get more tightly packed in the membrane which would explain the high rate at which the TMP increased.

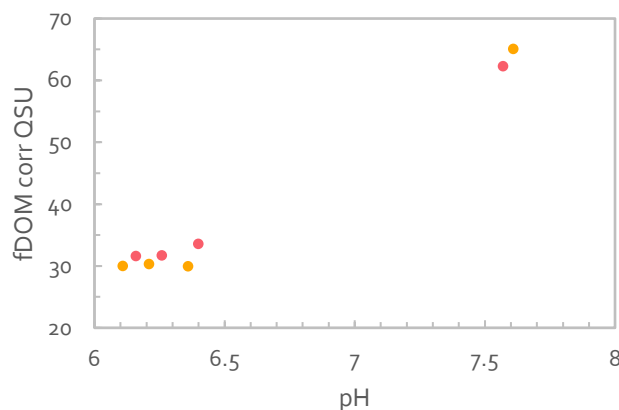


Figure 7: The effect of reaction time on fDOM removal (● 0 s and ● 150 s).

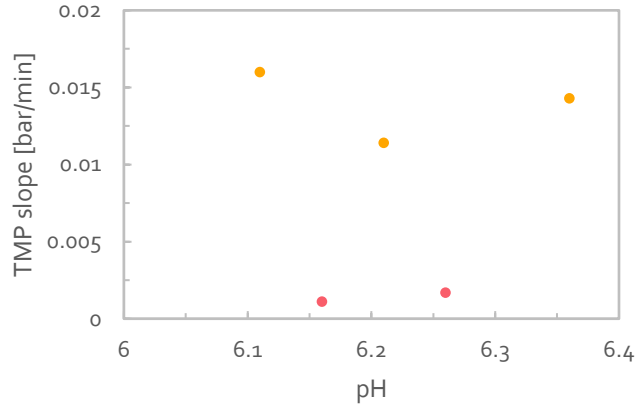


Figure 8: Shows the effect of reaction time on the TMP slope (● 0 s and ● 150 s).

## 4.2 Influence of feed water

The influence of feed water was analysed by conducting experiments with feed waters of different water qualities. Water from Fyrisån was of special interest since this water was about twice as contaminated as the Görväln water. The experiments with Fyrisån water would therefore function as load tests and give an indication of how the pilot plant's efficiency was affected by feed water. As can be seen in Figure 9 and Figure 10 below, each feed water seems to reach a maximum removal efficiency at about 55-60 %, where the curves level out.

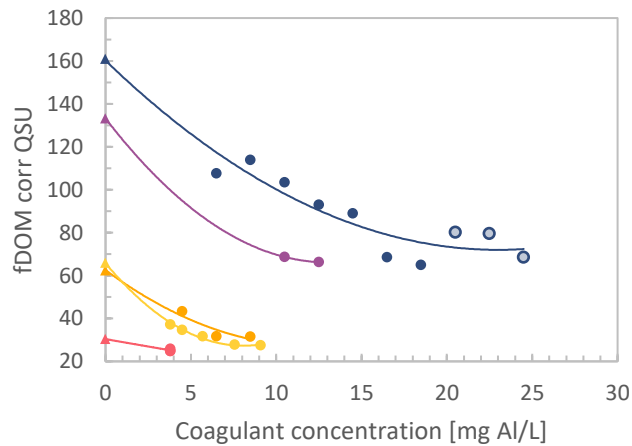


Figure 9: The remaining fDOM for the different coagulant dosages and feed waters (● Görväln (ALG), ● Görväln (PAC), ● sand filtrate, ● Fyrisån & sand filtrate, ● Fyrisån). The data points with a light centre are experiments where sodium carbonate have been used for the pH adjustment.

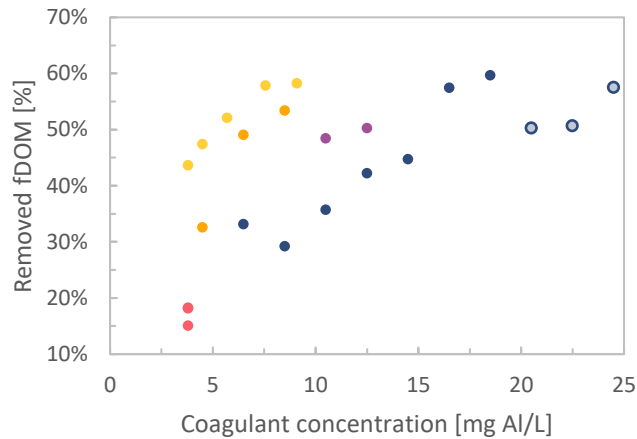


Figure 10: The fDOM removal in percentages for the different feed waters (● Görväln (ALG) ● Görväln (PAC), ● sand filtrate, ● Fyrisån & sand filtrate, ● Fyrisån). The data points with a light centre are experiments where sodium carbonate have been used for the pH adjustment.

When looking at the fDOM removal in percentages, the pilot plant seems to be as efficient with both Görväln and Fyrisån feed waters, as is shown in Table 11 below. The maximum removal efficiency for Fyrisån that was reached in these experiments was at 60 % with a coagulant dosage of 18.5 mg/L. The maximum removal efficiency for Görväln (PAC) was at 55 % with a coagulant dosage of 8.5 mg/L. Since no more than 60 % could be removed in any of the experiments, irrespective of the initial water quality, it could suggest that about 40 % of the total fDOM content is not separable with the plant when using aluminium based coagulants.

The results were compared by looking at the coagulant efficiency. The sand filtrate had a coagulant efficiency at an average of 1.4. Meanwhile, the coagulant efficiency was around 14.2 for Görväln experiments, which indicates that the fDOM was more easily precipitated and separated with this feed water. Fyrisån feed water had an average coagulant efficiency of 4.4, which suggests that Fyrisån water was more difficult to coagulate than Görväln water.

The efficiency of the plant with sand filtrate as feed water was at the most 18 %, which is relatively low in comparison to the other feed waters. This could be explained by the fact that the easily precipitated NOM have already been separated in the sand filters. However, the permeate from these experiments was of the best quality when looking at the fDOM content in the permeate.

Table 11: A compilation of the experiments where either the achieved fDOM levels were lower than 32, or more than 50 % of the initial fDOM was removed.

<i>Feed water</i>	Coagulant [mg/L]	pH in permeate	fDOM in permeate	fDOM removal [%]	Coagulant efficiency [fDOM removed per mg/L]
<i>Fyrisån</i>	16.5	6.63	68.5	57	5.6
	18.5	6.54	64.9	<b>60</b>	5.2
	20.5	6.64	80.1	50	3.9
	22.5	6.67	79.5	51	3.6
	24.5	6.60	68.4	57	3.8
<i>Sand filtrate</i>	3.8	6.27	24.9	18	1.5
	3.8	6.40	24.8	<b>18</b>	1.5
	3.8	6.61	25.8	15	1.2
<i>Fyrisån &amp; sand filtrate</i>	12.5	6.48	66.3	50	5.4
<i>Görvaln (PAC)</i>	6.5	6.16	31.6	49	15.6
	8.5	6.33	30.6	<b>55</b>	12.1
<i>Görvaln (ALG)</i>	5.7	6.41	31.6	52	17.8
	7.6	6.33	27.8	<b>58</b>	13.9
	9.1	6.48	27.5	58	11.6

Figure 11 below displays how the value for SUVA decreases as the coagulant concentration increases. Lowered SUVA values indicate that humic substances have been eliminated from the water and that mainly non-humic substances remain. The curves for the different feed waters seem to level out and stagnate around a SUVA value of 1.75. This complies with the fact that the coagulation process becomes more difficult and less efficient in waters of low SUVA values. This value can be used as an indication of how much further the coagulant concentration can be increased in order to achieve more fDOM removal. Once reached, the coagulation process loses its efficiency since the separable substances have already been separated.

Assuming that a SUVA of 1.75 is the “limit”, the experiments with Fyrisån water could potentially have been run with higher coagulant dosages since the lowest SUVA that was achieved for this water was at about 2. The pH is assumed to already have been optimised during these experiments since all Fyrisån experiments had a final pH within the pH range that has previously been shown to not affect the efficiency of the pilot plant. Though, the pH values were quite close to the upper limit of 6.7 of the pH range which suggest that the pH could still have had a negative effect on the fDOM removal. However, it is also possible that this water had higher amounts of non-humic substances.

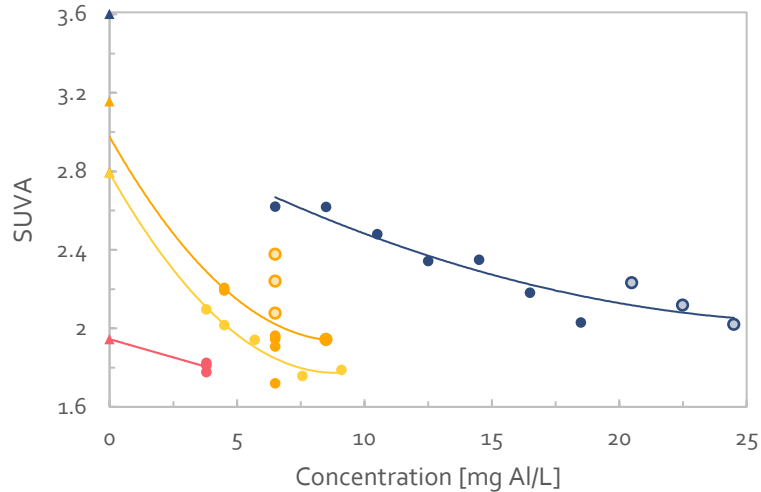


Figure 11: SUVA against coagulant concentrations for the different feed waters (● Görväln (ALG) ● Görväln (PAC), ● sand filtrate, ● Fyrisån). The data points with a light centre are experiments where sodium carbonate have been used for the pH adjustment.

The TMP was measured for each of the experiments in this section, however, this data was not used since they were not comparable to previous data due to several backwashes being performed in the middle of some of the experiments. Neither the TMP itself nor the TMP slope (the rate of escalation of TMP) could therefore be used to analyse the effect of feed water on the membrane.

However, during the experiments, it was very clear that the membrane could not handle the high fDOM levels of Fyrisån water very well. The TMP increased rather quickly which led to many backwashes being required in order to not overload the membrane.

#### 4.3 Placement of the ultrafiltration process

The experiments performed with sand filtrate as feed water showed that a UF process step could be placed after sand filtration. The pilot plant could remove an additional 18 % of fDOM from the sand filtrate. The plant did not seem to have any issues with the TMP since two consecutive measurements could be performed without the need of a backwash.

If the UF were to be placed between the sand filtration and carbon filters, it would increase the removal of organic substances whilst also reducing the load on the subsequent carbon filters. However, the possibility of placing the UF after the carbon filters is also an alternative. This could potentially decrease the risk of biofouling of the membrane.

#### 4.4 Verification of data

The theoretical values and the data gathered from the EXO sensor were verified by comparing them to the laboratory results. The errors in percentages, for each individual data point, have been compiled in *Appendix F*.

The alkalinity values were somewhat scattered around the dashed line as can be seen in Figure 12. The dashed line corresponds to the values where the theoretical and measured value is the same. The values with the biggest differences were with the Fyrisån experiments that had been pH adjusted with sodium carbonate. This suggests that sodium carbonate has an effect on the alkalinity of the water.

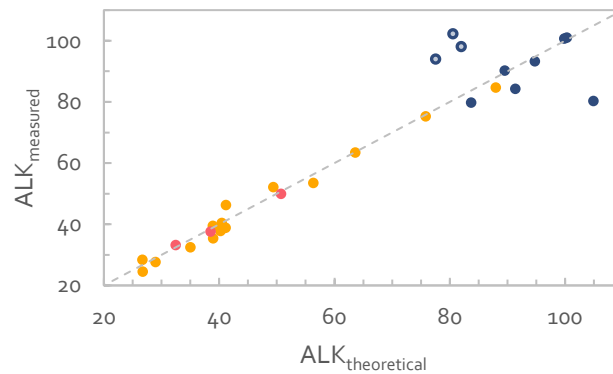


Figure 12: The correlation between experimental alkalinity data and laboratory or theoretical values. The colours correspond to the different feed waters (● Görväln, ● sand filtrate, ● Fyrisån).

The measured UV values were on average 2.68 units lower than the theoretical values, and the average error in percentages was 27.48 %. However, since a strong correlation ( $r^2=0.9036$ ) could be seen between the data points it indicates that the error is a constant error that can be adjusted for, see Figure 13.

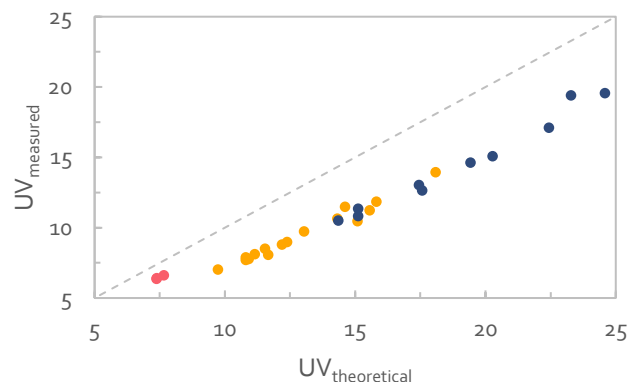


Figure 13: The correlation between experimental UV data and laboratory or theoretical values. The colours correspond to the different feed waters (● Görväln, ● sand filtrate, ● Fyrisån).

Figure 14 below, show that most of the achieved coagulant dosages were close to the values aimed for (theoretical values). The achieved (measured) values were found by comparing chloride concentrations before the coagulant additions to the concentrations after the additions. The experiments are therefore assumed to be correct regarding the coagulant dosage. On average, the values were 0.84 mg/L and 9.9 % off from the theoretical values.

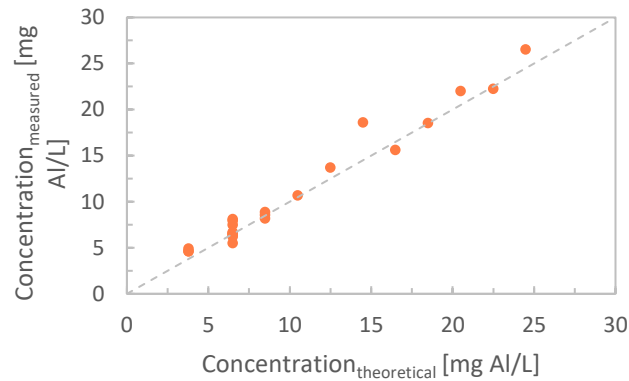


Figure 14: The correlation between the theoretical and measured coagulant dosages.

The measured pH values from the EXO probe were on average 0.3 units higher than the corresponding values from the laboratory, see Figure 15. The  $r^2$ -value of the data points was 0.9456, which indicates that there is a linear relationship between the measurements. The reason for all of the measured data being systematically higher than the corresponding laboratory values could therefore be due to calibration issues in the EXO sensor and/or the use of different electrodes that are more or less adapted for measuring in these types of solutions.

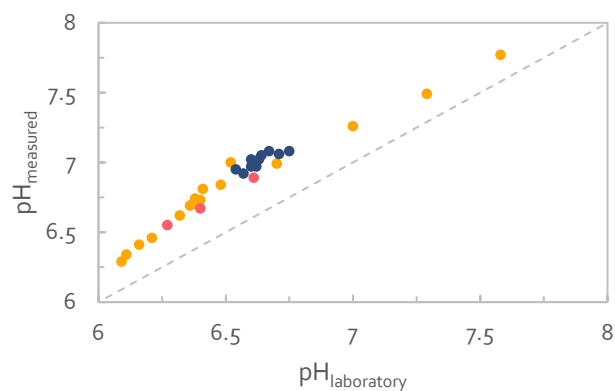


Figure 15: The correlation between experimental pH data and laboratory or theoretical values. The colours correspond to the different feed waters (● Görvåln, ● sand filtrate, ● Fyrisån).

At Norrvatten, the coagulant dosages are continuously adjusted based on the measured  $UVA_{254}$  of the incoming water. As can be seen in Figure 16, a strong correlation could be seen between the measured fDOM values and their respective  $UVA_{254}$  which means that fDOM can be used in the same way as  $UVA_{254}$  is used to adjust the coagulant dosing. The  $r^2$ -value for water from Fyrisån was 0.9682 and for Görvåln it was 0.9816, a strong correlation is therefore present between the  $UVA_{254}$  and fDOM of the water.

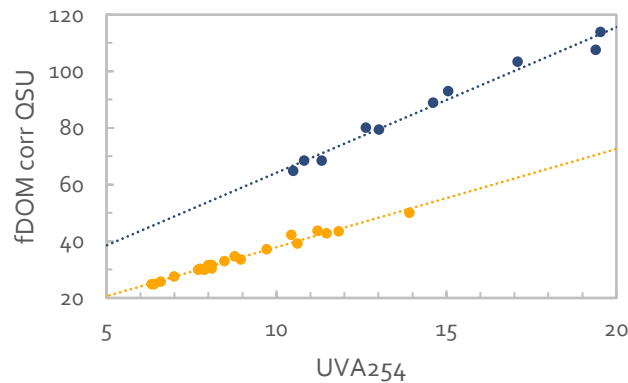


Figure 16: Correlation between fDOM and  $UVA_{254}$  for feed waters from ● Görvåln and ● Fyrisån.

#### 4.5 Experimental uncertainties and future studies

The membrane seemed to have irreversible fouling issues since the initial TMP increased during the period of use which suggests that floc might have accumulated in the membrane. At the beginning, the TMP was about 0.25 bar directly after a backwash, but later on in the project this value increased to about 0.5 bar. This could lead to some uncertainties in all of the results and more importantly, the comparability of the results. For future studies, the membrane should either be replaced more often, or a more efficient backwashing routine should be followed.

During the initial experiments with feed water from Fyrisån, the fDOM levels first decreased slightly before rising again. This raised some question about the stirring speed. The stirring was therefore slowed down in order to allow propped formation of floc and also to avoid breaking up formed floc. This measure solved the issue of the rising fDOM levels and could therefore be an interesting parameter to look into in future studies.

Mainly the combined effect of coagulant dosage and final pH have been analysed. Therefore, it would be of interest to perform more excessive experiments on the effect of pH by not only



measuring the resultant pH, but also the pH of the feed water in order to establish if it is in fact the pH or the coagulant dosage, or both, that affects the coagulation efficiency.

Only two different reaction times were analysed during this project. To be able to draw a more definitive conclusion about the effect of reaction time, several reaction times should be analysed.

The most optimal placement of the ultrafilter process step needs to be studied further. Experiments with feed water from the carbon filters should be performed in order to determine the efficiency of the plant when using this type of water.

## 5. Conclusion

The pH did not have a direct effect on the efficiency. When the plant was operated in slightly acidic conditions (pH 6.0-6.7), no significant difference in fDOM removal was observed. More significant differences could be seen between the coagulant dosages which indicates that this parameter was of importance.

As for the reaction time, it was clear that a reaction time of 150 seconds resulted in more favourable floc formation since the TMP in these experiments increased ten times slower than in the experiments with a reaction time of 0 seconds. However, the fDOM removal was similar for both reaction times.

From the experiments with different feed waters, it was concluded that an fDOM removal efficiency of 60 % could not be exceeded with the pilot plant. The initial feed water quality did not have an effect on the fDOM removal. However, more contaminated waters such as Fyrisån required many backwashes since the TMP increased quickly, this suggests that these waters were challenging for the plant. Thus, from an economical and environmental point of view, the plant is not efficient with highly contaminated waters such as Fyrisån.

The experiments conducted with sand filtrate suggest that an ultrafiltration step after a sand filtration process would be effective. However, further studies are required to be able to determine the most optimal placement of the ultrafiltration process in a drinking water purification process.

In conclusion, the results achieved with the pilot plant show promising signs of an ultrafiltration process being a viable alternative for Norrvatten to increase their DWTPs efficiency and capacity.

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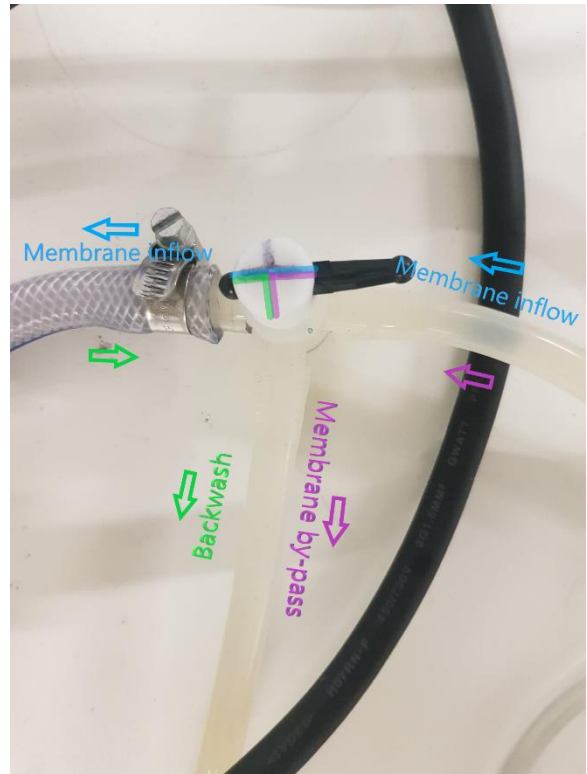
## 7. Appendices

### Appendix A: Pilot plant

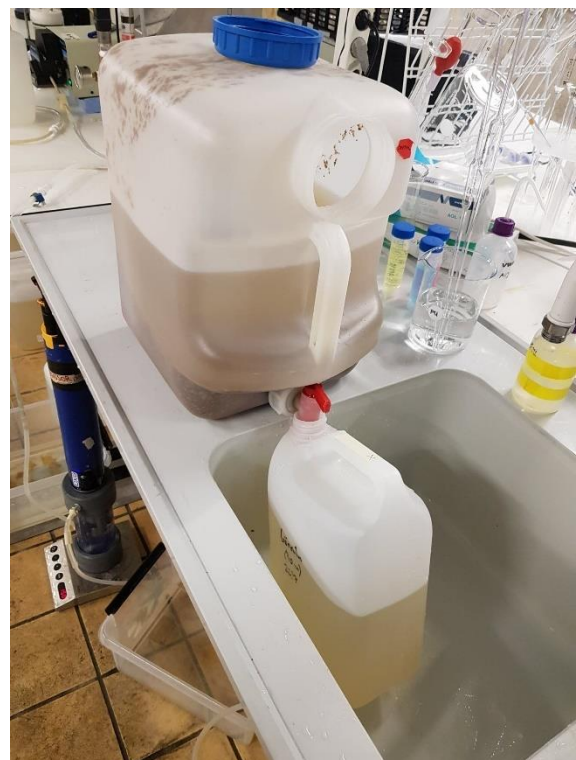
The two pictures below show the pilot plant when it was in use during the project.



To the right, a picture of a three-way valve from a part of the pilot plant is shown. This valve was adjusted during backwashes and membrane bypasses. The blue arrows show how the valve was set during the experiments. The green arrows show the setting for backwashes, and purple for experiments where the membrane was bypassed.



The water from Fyrisån was filtered through a sand filter before use, as can be seen in the picture to the right.



## Appendix B: Conversion calculations & solution preparations

**Conversion calculations.** Conversion calculations for the sulfuric acid and coagulant can be seen below.

$$\frac{SO_4 \left[ \frac{mg}{L} \right]}{10^3 \cdot M_{W,H_2SO_4} [mg/mol]} = mol \text{ added } H_2SO_4/L$$

$$\frac{[mol \text{ added } H_2SO_4/L]}{0.001 [total \text{ mol } H_2SO_4/L]} = \frac{x [ml \text{ added } H_2SO_4/L]}{20 [total \text{ ml } H_2SO_4/L]}$$

$$x [ml \text{ added } H_2SO_4/L] = \frac{SO_4 \left[ \frac{mg}{L} \right]}{0.001 [total \text{ mol } H_2SO_4]} \cdot 20 [total \text{ ml } H_2SO_4]$$

$$x [ml \text{ added } H_2SO_4/L] = \frac{SO_4 \left[ \frac{mg}{L} \right]}{M_{W,H_2SO_4} [mg/mol]} \cdot 20 [total \text{ ml } H_2SO_4]$$

The values for the coagulant were given as milligrams of aluminium per litre, which required conversion to milligrams of poly aluminium chloride per litre, as follows:

$$\frac{Al [mg/L]}{Al(OH)_{1.2}Cl_{1.8} [mg/L]} = \frac{M_{w,Al}}{M_{w,Al(OH)_{1.2}Cl_{1.8}}}$$

**Coagulant solutions.** The coagulant solutions were prepared in volumes of 200 mL, in order to be enough for about three consecutive experiments. The concentration of the concentrated poly aluminium chloride solution was 98g Al/L. To prepare a solution for an experiment where 5mg Al/L was the concentration of interest, a solution of 500mg Al/L would be prepared. The formula below was used to calculate how much of the concentrated Al solution should be diluted to 200 mL with Milli-Q water.

$$C1_{Al,initial} \cdot V1_{Al,initial} = (C2_{Al,final} \cdot 100) \cdot V2_{Al,final}$$

$$98\,000 \text{ mg Al/L} \cdot V1_{Al,initial} = (5\text{mg Al/L} \cdot 100) \cdot 0.2 \text{ L}$$

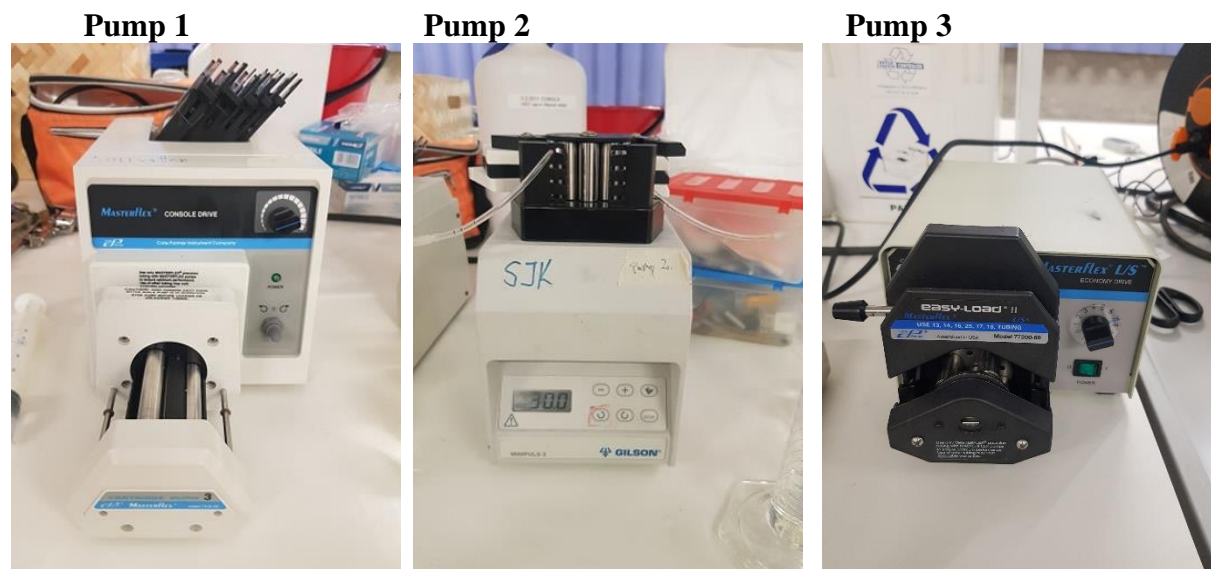
$$V1_{Al,initial} = 0.001 \text{ L} = 1 \text{ ml}$$

According to the calculations above, 1 mL of the concentrated Al solution would have to be diluted in 199 mL of Milli-Q water to reach a concentration of 500mg Al/L, which would result in a concentration of 5mg Al/L in the plant.



## Appendix C: Calibration of pumps

The process of the calibration of the three pumps is shown below.

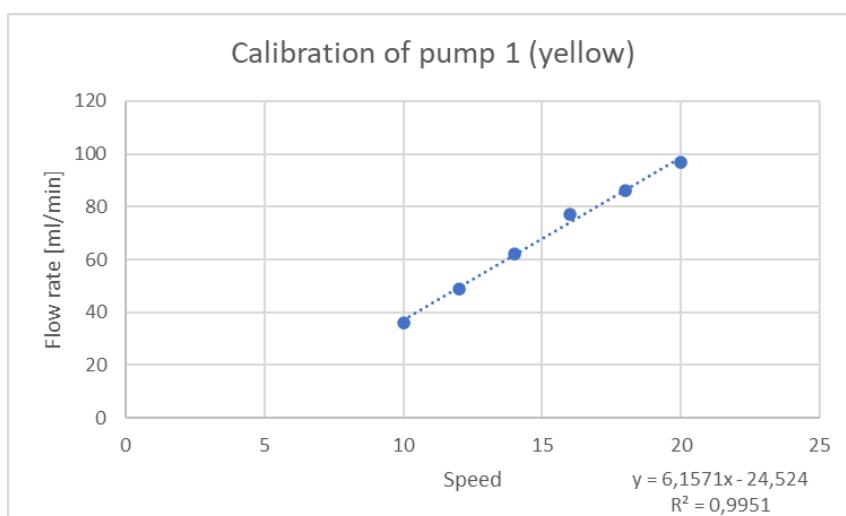


### Pump 1

*Yellow tube (90 mL/min)*

10 speed: 36 mL/min  
12 speed: 49 mL/min  
14 speed: 62 mL/min  
16 speed: 77 mL/min  
18 speed: 86 mL/min  
20 speed: 97 mL/min

*For a flow rate of 90 mL/min a speed of 18.6 is required.*

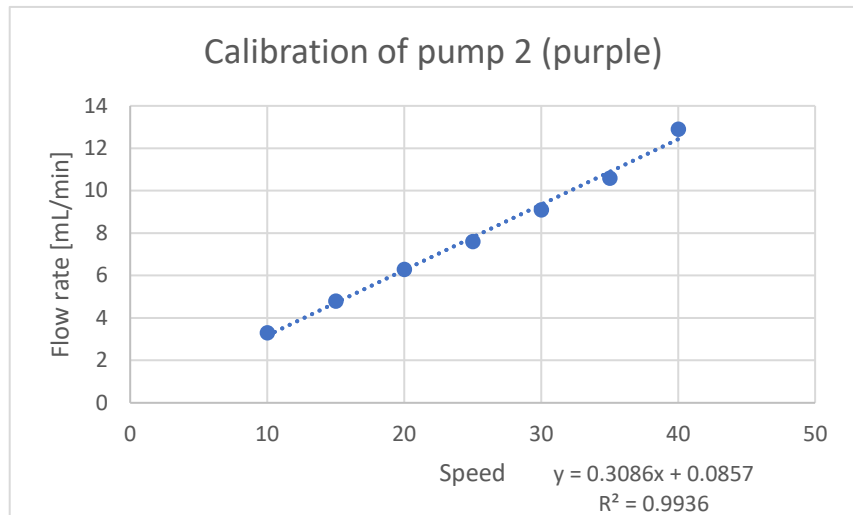


## Pump 2

Purple tube: (9 mL/min)

10 speed: 3.3 mL/min  
15 speed: 4.8 mL/min  
20 speed: 6.3 mL/min  
25 speed: 7.6 mL/min  
30 speed: 9.1 mL/min  
35 speed: 10.6 mL/min  
40 speed: 12.9 mL/min

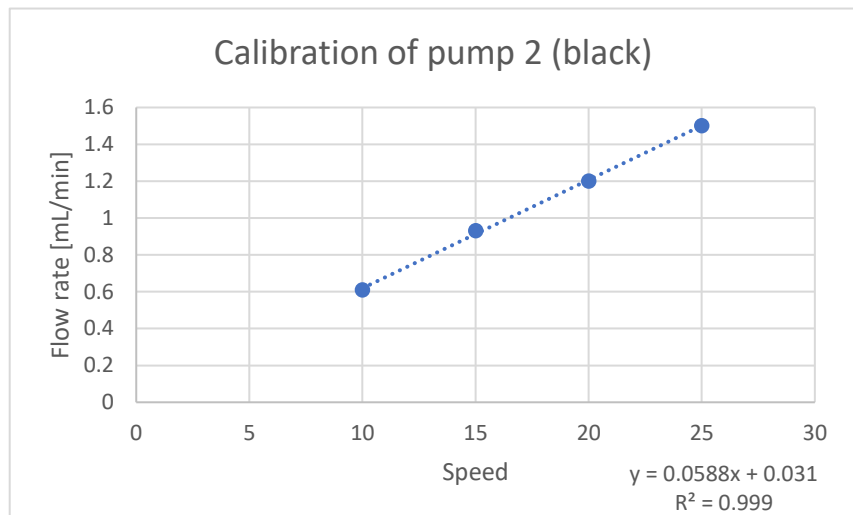
For a flow rate of 4.5 mL/min a speed of 14.2 is required. If fastened looser, a speed of 15.5 is required.



Black tube: (1 mL/min)

10 speed: 0.61 mL/min  
15 speed: 0.93 mL/min  
20 speed: 1.2 mL/min  
25 speed: 1.5 mL/min

For a flow rate of 1 mL/min a speed of 16.5 is required. If fastened tighter, a speed of 15.5 is required.

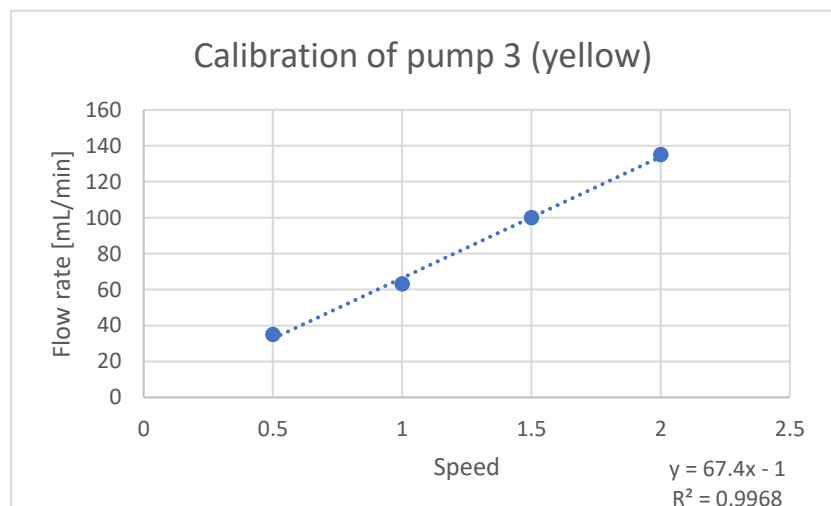


## Pump 3

(total of 100 mL from pump 1+pump 2)

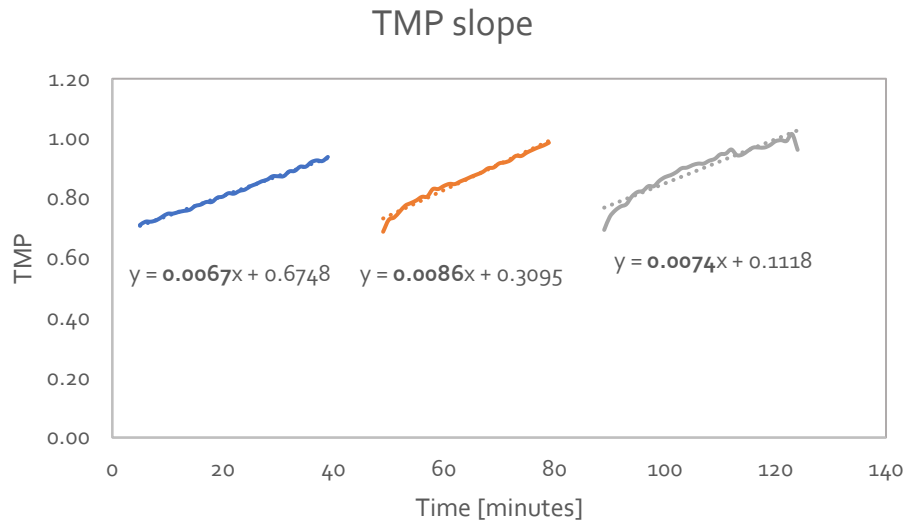
Yellow tube:

0.5 speed: 35 mL/min  
1 speed: 63 mL/min  
1.5 speed: 100 mL/min  
2 speed: 135 mL/min



## Appendix D: TMP slope calculation

The TMP slope was produced by plotting and then creating a trendline for the data points of interest, according to the figure below. The TMP slope is the values in bold.



## Appendix E: Compilation of experimental results

<i>Feed water</i>	Coagulant [mg/L]	H <sub>2</sub> SO <sub>4</sub> 0.05M [ml/10L]	Na <sub>2</sub> CO <sub>3</sub> 0.1M [ml]	pH laboratory	fDOM in permeate	ALK [mg HCO <sub>3</sub> /L]	TOC	Chloride [mg/L]	Sulphate [mg/L]	UV m <sup>-1</sup>	TMP
<i>Görvältn</i>	4.5	16	-	7	42.86	53.44	<del>42.97</del>	27.09	38.87	11.48	-
<i>Görvältn</i>	0	-	-	7.67	62.26	70.14	7.70	17.51	32.03	23.80	-
<i>Görvältn</i>	6.5	25	-	6.73	33.58	39.35	5.21	29.07	43.54	8.96	<del>0.003</del>
<i>Görvältn</i>	6.5	45	-	6.41	31.57	28.30	4.13	29.54	50.91	8.06	0.0011
<i>Görvältn</i>	4.5	40	-	6.62	43.68	35.32	5.09	24.69	51.04	11.22	0.0039
<i>Görvältn</i>	4.5	60	-	6.29	42.31	24.42	4.76	25.05	57.99	10.44	0.0043
<i>Görvältn</i>	0	-	-	7.65	67.64	74.12	8.05	17.68	32.40	25.40	
<i>Görvältn</i>	8.5	-	-	6.99	32.97	52.05	4.38	32.90	31.63	8.48	0.0031
<i>Görvältn</i>	8.5	17	-	6.8	31.51	42.88	4.12	32.22	40.35	8.00	0.0039
<i>Görvältn</i>	0	-	-	7.8	64.42	74.59	8.14	17.76	32.37	25.52	-
<i>Görvältn</i>	6.5	-	5	7.26	39.24	63.35	5.11	29.55	31.73	10.62	0.0082
<i>Görvältn</i>	6.5	-	15	7.49	43.52	75.20	5.29	29.35	31.71	11.84	0.0118
<i>Görvältn</i>	6.5	-	25	7.77	50.04	84.57	5.86	28.88	31.41	13.92	0.0162
<i>Görvältn</i>	0	-	-	7.9	65.10	74.56	8.41	17.38	31.74	23.52	-
<i>Görvältn</i>	6.5	25	-	6.69	29.96	38.73	4.13	30.80	41.86	7.88	0.0143
<i>Görvältn</i>	6.5	35	-	6.46	30.30	32.41	3.99	31.63	48.66	7.76	0.0114
<i>Görvältn</i>	6.5	45	-	6.34	29.98	27.57	3.92	31.97	52.54	7.70	0.016
<i>Sand filtrate</i>	0	-	-	6.66	30.37	44.29	4.16	17.28	58.05	8.10	-
<i>Sand filtrate</i>	3.8	-	-	6.55	24.85	33.08	3.60	25.58	55.54	6.40	0.0067
<i>Sand filtrate</i>	3.8	-	5	6.67	24.81	37.54	3.50	26.08	55.98	6.34	0.0086
<i>Sand filtrate</i>	3.8	-	15	6.89	25.78	49.83	3.62	25.51	55.62	6.60	0.0074

<i>Fyrisån</i>	0	-	-	7.72	160.95	149.60	15.46	31.54	47.07	55.68	-
<i>Fyrisån</i>	6.5	45	-	6.97	107.54	80.18	7.40	41.38	66.29	19.40	0.0233
<i>Fyrisån</i>	8.5	40	-	7.08	113.87	100.64	7.46	47.50	54.51	19.54	0.0219
<i>Fyrisån</i>	10.5	35	-	6.97	103.42	93.21	6.90	50.75	59.66	17.10	0.021
<i>Fyrisån</i>	12.5	30	-	6.92	92.98	90.11	6.43	56.16	57.74	15.06	0.0198
<i>Fyrisån</i>	14.5	-	-	7.06	88.91	100.88	6.22	64.99	42.87	14.62	0.0186
<i>Fyrisån</i>	16.5	-	-	7.02	68.46	84.14	5.20	59.64	43.08	11.34	0.0201
<i>Fyrisån</i>	18.5	-	-	6.95	64.88	79.67	5.17	64.88	44.86	10.50	0.0219
<i>Fyrisån</i>	20.5	-	5	7.05	80.08	97.97	5.67	71.13	43.78	12.64	0.024
<i>Fyrisån</i>	22.5	-	10	7.08	79.47	102.17	6.15	71.54	43.63	13.02	0.0208
<i>Fyrisån</i>	24.5	-	15	7.02	68.44	93.92	5.36	79.26	46.56	10.82	0.0274
<i>2:1 F+SF</i>	10.5	35	-	6.8	69.13	66.27	5.66	46.25	61.80	11.72	0.0181
<i>2:1 F+SF</i>	12.5	30	-	6.77	66.70	67.07	5.27	49.61	61.58	11.26	0.0178
<i>Görväln (ALG)</i>	0	-	-	8.04	65.95	75.64	8.40	17.54	32.09	23.44	-
<i>Görväln (ALG)</i>	3.8	30	-	6.7	37.14	38.35	4.64	18.78	57.35	9.72	0.0314
<i>Görväln (ALG)</i>	4.5	22	-	6.74	34.66	37.76	4.35	17.09	60.63	8.78	0.0231
<i>Görväln (ALG)</i>	5.7	9	-	6.81	31.61	40.38	4.17	17.28	59.79	8.10	0.014
<i>Görväln (ALG)</i>	9.1	-	15	6.84	27.52	46.19	3.91	16.95	67.65	7.00	0.0126
<i>Görväln (ALG)</i>	0	-	-	8.15	65.59	76.88	8.71	17.30	31.10	23.52	-
<i>Görväln (ALG)</i>	7.57	-	5	6.68	27.79	38.33	3.93	17.42	68.40	6.90	0.0067

## Appendix F: Additional verification data

The tables in this section show additional data for 4.3 *Verification of data*.

Feed water	Theoretical alkalinity	Measured alkalinity	Error [%]
R	56.37	53.44	5.49
R	38.92	39.35	1.08
R	26.72	28.30	5.57
R	38.98	35.32	10.36
R	26.78	24.42	9.67
R	49.44	52.05	5.01
R	63.63	63.35	0.45
R	75.83	75.20	0.84
R	88.03	84.57	4.10
R	41.16	38.73	6.28
R	35.06	32.41	8.19
R	28.96	27.57	5.05
SF	32.46	33.08	1.88
SF	38.56	37.54	2.71
SF	50.76	49.83	1.86
F	104.96	80.18	30.90
F	99.92	100.64	0.72
F	94.76	93.21	1.67
F	89.57	90.11	0.60
F	100.35	100.88	0.53
F	91.40	84.14	8.63
F	83.75	79.67	5.12
F	82.01	97.97	16.29
F	80.55	102.17	21.16
F	77.59	93.92	17.39
F+SF	78.39	62.41	25.61
F+SF	86.38	66.27	30.35
F+SF	78.39	67.07	16.88
R	40.52	38.35	5.66
R	40.27	37.76	6.64
R	40.49	40.38	0.28
R	41.18	46.19	10.85
R	40.74	38.33	6.29

Feed water	Theoretical UV m <sup>-1</sup>	Measured UV m <sup>-1</sup>	Error [%]
R	14.62	11.48	27.37
R	12.40	8.96	38.39
R	11.68	8.06	44.88
R	15.57	11.22	38.74
R	15.09	10.44	44.58
R	11.55	8.48	36.16

R	14.32	10.62	34.88
R	15.83	11.84	33.68
R	18.10	13.92	30.03
R	10.81	7.88	37.13
R	10.93	7.76	40.79
R	10.81	7.70	40.43
SF	7.39	6.40	15.53
SF	7.38	6.34	16.44
SF	7.67	6.60	16.14
F	23.29	19.40	20.07
F	24.59	19.54	25.84
F	22.45	17.10	31.26
F	20.29	15.06	34.70
F	19.44	14.62	32.94
F	15.12	11.34	33.35
F	14.36	10.50	36.74
F	17.58	12.64	39.11
F	17.46	13.02	34.07
F	15.12	10.82	39.72
F+SF	10.94	8.86	23.52
F+SF	10.54	11.72	10.03
F+SF	10.19	11.26	9.54
R	13.04	9.72	34.18
R	12.20	8.78	38.90
R	11.15	8.10	37.65
R	9.74	7.00	39.15
R	9.84	6.90	42.55

Theoretical [mg Al/L]	Measured [mg Al/L]	Error [%]
6.5	6.42	1.24
6.5	6.68	2.78
8.5	8.55	0.56
8.5	8.17	3.90
6.5	6.55	0.77
6.5	6.44	0.94
6.5	6.18	4.93
6.5	7.46	14.71
6.5	7.92	21.77
6.5	8.10	24.65
3.8	4.61	21.24
3.8	4.89	28.65
3.8	4.57	20.26
6.5	5.47	15.87
8.5	8.87	4.35
10.5	10.68	1.68
12.5	13.68	9.44
14.5	18.59	28.18
16.5	15.61	5.37
18.5	18.53	0.14

20.5	22.00	7.30
22.5	22.23	1.22
24.5	26.51	8.21

<b>Feed water</b>	<b>Theoretical pH</b>	<b>Measured pH</b>	<b>Error [%]</b>
R	6.52	7	6.86
R	6.4	6.73	4.90
R	6.16	6.41	3.90
R	6.32	6.62	4.53
R	6.09	6.29	3.18
R	6.7	6.99	4.15
R	7	7.26	3.58
R	7.29	7.49	2.67
R	7.58	7.77	2.45
R	6.36	6.69	4.93
R	6.21	6.46	3.87
R	6.11	6.34	3.63
SF	6.27	6.55	4.27
SF	6.4	6.67	4.05
SF	6.61	6.89	4.06
F	6.6	6.97	5.31
F	6.75	7.08	4.66
F	6.62	6.97	5.02
F	6.57	6.92	5.06
F	6.71	7.06	4.96
F	6.63	7.02	5.56
F	6.54	6.95	5.90
F	6.64	7.05	5.82
F	6.67	7.08	5.79
F	6.6	7.02	5.98
F+SF	6.55	6.95	5.76
F+SF	6.5	6.8	4.41
F+SF	6.48	6.77	4.28
R	6.37	6.7	4.93
R	6.38	6.74	5.34
R	6.41	6.81	5.87
R	6.48	6.84	5.26



