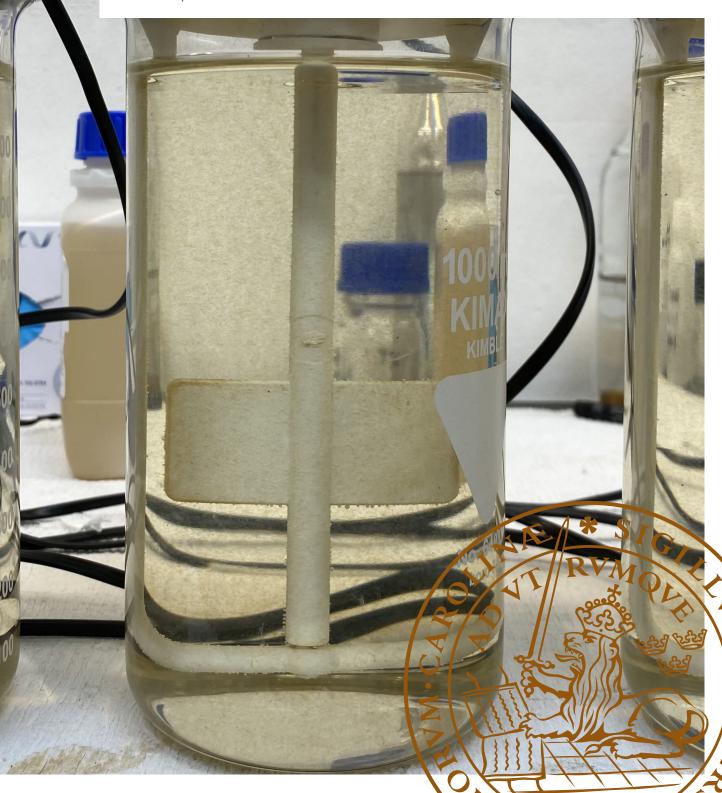
## Kemila

# Effects of recirculating water treatment sludge

DEPARTMENT OF CHEMICAL ENGINEERING | LUND UNIVERSITY SOFIA MILE | MASTER THESIS 2023



## Effects of recirculating water treatment sludge

by

## Sofia Mile

Master Thesis number: 2023-04

Water and Environmental Engineering Department of Chemical Engineering Lund University

June 2023

Supervisor: Michael Cimbritz Co-supervisor: Tobias Persson & Kristofer Hägg, Sydvatten Examiner: Åsa Davidsson

Picture on front page: Jar tests. Photo by Sofia Mile.

Postal address Box 124 SE-221 00 Lund, Sweden Web address http://www.lth.se/chemeng/ Visiting address Kemicentrum Naturvetarvägen 14 223 62 Lund, Sweden **Telephone** +46 46-222 82 85 +46 46-222 00 00

## Preface

This master thesis was conducted at Ringsjöverket (Sydvatten AB) and was carried out in collaboration with Sydvatten AB and the Department of Chemical Engineering at Lund University. I am grateful that I got the opportunity to write this thesis and that I got to deepen my knowledge of water treatment. I am also grateful for all the help I have received to be able to complete this project. Therefore, there are some people that I would like to thank.

Firstly, I would like to thank my supervisors from Sydvatten, Tobias Persson and Kristofer Hägg, for helping me with this project and for always taking their time to answer my questions and explaining different concepts and processes. Furthermore, a special thanks to the staff at Ringsjöverket for making me feel welcomed during my time there.

I would also like to thank Michael Cimbritz and Åsa Davidsson, my supervisor and examiner from the department for helpful advice and answering my questions.

Finally, I would like to thank my family and friends for not only supporting me in this project but throughout my five years at LTH.

## Summary

Drinking water is crucial in our everyday life. As with many other processes, it is important to produce water of drinking quality in a sustainable manner. This can occur by e.g., decreasing the amount of chemicals in the process. When producing drinking water, coagulation and flocculation are common processes. Briefly, coagulation is the process when negatively charged organic matter is destabilised by coagulants and during flocculation, the destabilised organic matter aggregates into flocs. These flocs settle and can be removed from the treated water. Settled flocs are called sludge and contains organic matter and residuals from coagulants, which are typically iron or aluminium salts.

This thesis was carried out in collaboration with Sydvatten AB, a drinking water producer for western Skåne and the experiments were performed at Ringsjöverket, one of the drinking water plants which Sydvatten owns. The main objective was to investigate the effects of recirculating sludge in the treatment process and if it was possible to decrease the amount of coagulant when recirculating sludge. The laboratory work was conducted by using jar tests which are common for evaluation of coagulation and flocculation. The absorbance of the treated water was measured, and the lower measured absorbance the more organic matter had been removed. The used wavelength were 254 nm and 436 nm. Similar to the large-scale process, water from lake Bolmen in Småland was used. However, instead of using ferric chloride, a polyaluminium chloride coagulant (PAX-XL60) was used.

When recirculating sludge, absorbance decreased for low coagulant dosages (3–5 mg Al/l), and for higher dosages (6–8 mg Al/l), the absorbance remained unchanged. It was therefore concluded that recirculating sludge had the greatest effect on the lower coagulant dosages. Additionally, the absorbance decreased significantly for coagulant dosage 4 mg Al/l with sludge compared to the same dosage without sludge. Furthermore, recirculating sludge with 4 mg Al/l gave absorbance results comparable to 5 mg Al/l without sludge. It was concluded that when recirculating sludge, coagulant dosage decreased with 20 % and the degree of recirculation was 23 %.

The amount of produced sludge can decrease as a result from using less coagulant which is beneficial from a sustainable perspective. Furthermore, operational costs will decrease when using less coagulant. Before the recirculation takes place in the large-scale process, further experiments are recommended. One of these is investigating how the microbial barrier in the coagulation and flocculation processes is affected when recirculating sludge. It is also recommended to examine how the temperature varies throughout an entire year and its effect on the flocculation process.

## Sammanfattning

Dricksvatten är viktigt i vårt dagliga liv. Liksom med många andra processer, är det viktigt att producera dricksvatten av god kvalitet på ett hållbart sätt. Detta kan ske genom att till exempel, minska mängden kemikalier i processen. Vid produktion av dricksvatten är koagulering och flockning två vanliga processer. Kortfattat är koagulering processen då negativt laddat organiskt material destabiliseras med hjälp av koagulanter och flockning är processen då destabiliserat organiskt material aggregeras till flockar. Dessa sjunker och kan på så vis tas bort från det behandlade vattnet. Flockarna som sjunker kallas för slam och innehåller organiskt material och rester från koagulanter, som vanligtvis är salter av järn eller aluminium.

Det här projektet utfördes i samarbete med Sydvatten AB, en dricksvattenproducent för västra Skåne och experimenten utfördes på Ringsjöverket som är ett av vattenverken som Sydvatten äger. Det huvudsakliga syftet var att undersöka effekten av recirkulation av slam i reningsprocessen och om det var möjligt att minska mängden koagulant genom recirkulation av slam. Det laborativa arbetet bestod av "jar tests" som är vanliga när koagulering och flockning ska utvärderas. Absorbansen av det behandlade vattnet mättes och ju lägre absorbans desto mer organiskt material har tagits bort. Våglängderna som användes var 254 och 436 nm. Likt processen i fullskala, användes vatten från Bolmen i Småland. I stället för att använda järnklorid, användes polyaluminium klorid (PAX-XL60) som koagulant.

När slam recirkulerades, sjönk absorbansen för låga doser av koagulant (3–5 mg Al/l) och för de högre doserna (6–8 mg Al/l) var absorbansen oförändrad. Slutsatsen drogs att recirkulation av slam har störst effekt vid låga doser av koagulant. Dessutom sjönk absorbansen signifikant för koagulantdos 4 mg Al/l med slam jämfört med samma dos utan tillsatt slam. Recirkulation av slam för dos 4 mg Al/l gav resultat för absorbans likt 5 mg Al/l utan slam. Slutsatsen drogs att när slam recirkulerades sjönk dosen koagulant med 20 % och recirkuleringsgraden var då 23 %.

Mängden slam som produceras kan minska om man använder en mindre mängd koagulant vilket är fördelaktigt ur en hållbarhetssynpunkt. Dessutom kommer rörliga kostnader att sjunka eftersom en mindre mängd koagulant behövs. Innan recirkulation kan ske i den storskaliga processen, rekommenderas ytterligare experiment. Ett av dessa är att utreda hur den mikrobiologiska barriär som koagulering och flockning står för, påverkas när slam recirkuleras. Det är också rekommenderat att undersöka hur temperaturen varierar över året och dess påverkan på flockningen.

## **Table of Contents**

1	Intro	oduction1					
	1.1	Objective					
	1.2	Disposition					
2	Background						
	2.1	Sydvatten					
	2.2	Contaminants in surface water					
	2.3	Removal of contaminants					
	2.4	Coagulation					
	2.5	Coagulants					
	2.6	Flocculation					
	2.7	Handling of sludge 11					
3	Method						
	3.1	Jar tests					
	3.2	Spectrophotometry 15					
	3.3	Dosage optimisation					
	3.4	pH optimisation					
	3.5	Sludge production					
	3.6	Recirculation of sludge 17					
4	Resi	Ilts and discussion					
	4.1	Optimisation of dosage and pH 19					
	4.2	Sludge production					
	4.3	Recirculation of sludge 22					
5	Con	Conclusion					
6	Futu	13ests13ests13ctrophotometry15age optimisation15optimisation16ge production16rculation of sludge17nd discussion19misation of dosage and pH19ge production20rculation of sludge22on31ork33es35					
7	References						
A	ppendic	es					
	Appendix A: Residual VIS-absorbance for dosage and pH optimisation						
	Appen	Appendix B: Residual VIS-absorbance for initial sludge recirculation					
Appendix C: Residual VIS-absorbance for final sludge recirculation							
	Appendix D: Populärvetenskaplig sammanfattning 4						

## **1** Introduction

Water of drinking quality is an essential part of the everyday life. Besides being important in the domestic life, water is needed in the industry as well. As the population is constantly growing, the required amount of water is increasing, and it is of interest to meet the new demand and to develop the process to a more sustainable process.

Fresh water is used when producing drinking water, and in Sweden, groundwater and surface water are used as fresh water sources. Half of the required fresh water is groundwater and artificial groundwater. When using artificial groundwater, surface water is lead through soil layers similar to how real groundwater passes through these layers. The other half is surface water which originates from water bodies such as lakes and rivers. Typically, surface water is of lower quality than groundwater and require more steps in the treatment process (Svenskt Vatten, 2016). Two of these steps are coagulation and flocculation which require coagulants. Coagulants destabilise negatively charged particles present in surface water and as a result, flocs are created which settle as sludge. Coagulants are often metal salts, which are made out of aluminium or iron (Crittenden, *et al.*, 2012).

This thesis was performed in collaboration with Sydvatten AB, a drinking water producer for western Skåne. Sydvatten owns two drinking water treatment plants (DWTP), Ringsjöverket and Vombverket. A more detailed description of the two DWTP is provided in sections 2.1.1 and 2.1.2, but in short: water from lake Bolmen in Småland is treated at Ringsjöverket where coagulation and flocculation with ferric chloride (FeCl<sub>3</sub>) are performed, followed by filtration in sand filters. Water from lake Vombsjön is treated at Vombverket, and the principal treatment is infiltration i.e., artificial groundwater (Sydvatten, 2023a; Sydvatten, 2023c).

As written previously it is of importance to develop the treatment towards a more sustainable process. One alternative is to try and decrease the amount of coagulant. Currently at Ringsjöverket, water used during backwashing of the rapid sand filters is recycled back to some of the flocculation basins. The backwashing water contains particles of sludge residues that did not successfully settle. It has been noted that these basins need a lower dosage of coagulant compared to the other basins. It is not entirely clear if this effect results from diluting incoming water with the backwashing water, or if particles which are separated from the filters affect the flocculation or a combination of both. This observed effect is the reason behind the thoughts of recirculating sludge and thus the subject of this thesis.

Furthermore, there are plans to construct pipelines in which water from lake Bolmen would be transported from Ringsjöverket to Vombverket and thus being treated in the latter. This is to increase redundancy and safety in the overall drinking water production (Sydvatten, 2020). Water from lake Bolmen has a brown-yellowish colour because of high contents of natural organic matter (NOM). At Ringsjöverket the coagulant FeCl<sub>3</sub> is used, and best removal of NOM is experienced compared to if an aluminium-based coagulant was used. However, if water from lake Bolmen were to be treated at Vombverket, pre-treatment before the infiltration would be necessary to remove NOM, but FeCl<sub>3</sub> may not be required. An aluminium-based coagulant could be sufficient due to the effective infiltration step later on. This means that the NOM content is too high for the infiltration itself, but the previous treatment does not need to be the most efficient (Persson & Hägg, 2023).

#### 1.1 Objective

The main objective was to investigate if recirculating sludge had any effect on the treatment and if it was possible to decrease the coagulant dosage when recirculating sludge. The laboratory work was performed at Ringsjöverket with water from lake Bolmen. However, the used coagulant was not FeCl<sub>3</sub>, but PAX-XL60 which is a polyaluminium chloride coagulant. The results from this thesis would thus mainly give an indication of the future of the sludge at Vombverket but also an idea for sludge treatment at Ringsjöverket, as previously described. However, at present, the coagulant for Vombverket has not been decided.

As the sludge produced at Ringsjöverket contains iron residuals and Vombverket is not currently producing any aluminium-containing sludge, the first part of the objective was to produce sludge under optimal conditions to simulate the large-scale process in lab-scale. The two parameters which were investigated to fulfil the optimal conditions were pH and coagulant dosage. Once these were known, sludge production could be conducted, and the recirculation experiments could take place. When referring to optimal conditions, it means that the sludge should contain as much NOM as possible, i.e., removing as much NOM as possible from the treated water.

The objective can be summarised into two research questions:

- What are the optimal conditions (coagulant dosage and pH) for producing sludge when using PAX-XL60 as coagulant?
- Is it possible to decrease the coagulant dosage when recirculating water treatment sludge?

#### 1.2 Disposition

This thesis begins with describing Sydvatten and briefly its two drinking water treatment plants (Ringsjöverket and Vombverket), followed by explaining why treatment with coagulants is necessary and how coagulation and flocculation work. Chapter 3 describes the experimental set-up with relevant equations and the used analytical method. This is followed by a chapter presenting the results and a discussion regarding them. The thesis ends with a conclusion and ideas for future work regarding recirculating sludge.

## 2 Background

The background is divided into two main sections. The first one shortly describes Sydvatten and how Sydvatten operates its two drinking water treatment plants. This is followed by an explanation of why treatment is needed and the mechanisms behind the removal.

#### 2.1 Sydvatten

Sydvatten AB was founded in 1966 and is today owned by 17 municipalities in Skåne. Sydvatten produces drinking water that fulfils the demand of 1 million inhabitants in western Skåne. For this to be possible, Sydvatten owns and operates two drinking water treatment plants, Ringsjöverket and Vombverket, and the tunnel transporting water from lake Bolmen to Ringsjöverket (Sydvatten, 2021). At Ringsjöverket, water from lake Bolmen is treated and lake Ringsjön is used as a reservoir for lake Bolmen. Water from lake Vombsjön is treated at Vombverket. Figure 1 displays the locations of the two DWTP, the three lakes and the 17 municipalities.



Figure 1: Map over the 17 municipalities which own Sydvatten and the locations of lakes Bolmen, Ringsjön and Vombsjön, as well as Ringsjöverket and Vombverket. Adapted and published with kind permission from Sydvatten.

#### 2.1.1 Ringsjöverket

The DWTP Ringsjöverket is located in Stehag near lake Ringsjön and was commissioned in 1963. During the first operating years, lake Ringsjön was the source of raw water but since 1987, water from lake Bolmen in Småland has been used. The supply from lake Ringsjön was not sufficient which led to the construction of the tunnel from lake Bolmen to Ringsjöverket. Water is transported through an 80 km long tunnel followed by a 25 km long pipe. Today, lake Ringsjön is used a reserve if anything were to happen with lake Bolmen or the tunnel. The production rate of drinking water at Ringsjöverket is 1400 l/s (Sydvatten, 2023a). An overview of the process is depicted in Figure 2.

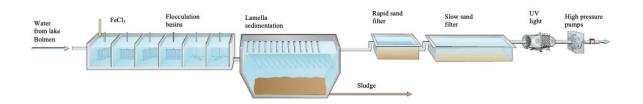
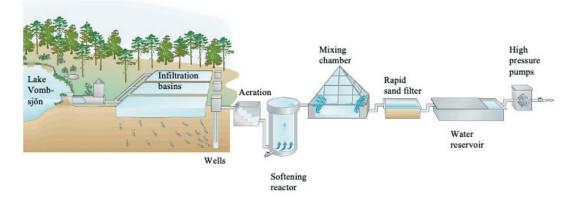


Figure 2. Overview of the treatment process at Ringsjöverket. Adapted and published with kind permission from Sydvatten.

Upon arrival at the water treatment plant, the water enters the flocculation basins and FeCl<sub>3</sub> is added. The organic matter in the water cluster together with the iron and flocs are formed. The flocs are then removed from the water in the sedimentation basins where lamellas facilitate the settling. The settled flocs are called sludge and 50 - 60 % of the sludge can be used in the biogas industry (Sydvatten, 2023b). This is followed by filtration, where rapid sand filters are used to separate the last flocs. This is followed by slow sand filters where microorganisms break down organic matter and thus decrease taste and odour. After this, water is treated with UV-light to disrupt the genetic material of microorganisms present in the water. It is a measure to prevent microorganisms from multiplying, i.e., a disinfection method. Lastly, before the water is pumped to the distribution network, chlorine is added as a disinfection agent (Sydvatten, 2023a). When treating water, having microbial barriers, i.e., removing microorganisms, is crucial. Some has been mentioned above, e.g., coagulation and flocculation, slow sand filtration, and disinfection such as UV treatment and chlorine treatment. The amount of required microbial barriers depends on the type of water and amount of microorganisms (Livsmedelsverket, 2023).

#### 2.1.2 Vombverket

Near lake Vombsjön in Veberöd, is Vombverket located. It was commissioned in 1948 and produces 1000 l/s (Sydvatten, 2023c). An overview of the process is depicted in Figure 3.



*Figure 3: Overview of the water treatment at Vombverket. Adapted and published with kind permission from Sydvatten.* 

The process begins with screens to remove mud and other large particles and is followed by division into 54 infiltration basins. The total surface area of these basins is 400 000 m<sup>2</sup>. By artificial groundwater infiltration, water is moving through sand and gravel to a natural groundwater storage. From one out of 114 wells, the water is pumped after two to three months and can undergo the next treatment step. Manganese and iron are oxidised by aerating the water and the water is then transported to softening reactors. Here, sodium hydroxide is added for removal of calcium and carbonate ions. Lime precipitates on sand and soft water is leaving the reactor. In the next step (in the mixing chamber), ferric chloride is added to remove the final lime crystals by forming flocs. These ferric-lime flocs and oxidised manganese and iron are removed in the rapid sand filters. In the last step, monochloramine is added as preventive step towards microorganisms (Sydvatten, 2023b).

#### 2.2 Contaminants in surface water

Both Ringsjöverket and Vombverket use surface water as raw water source. There is a large variety among organic and inorganic components of surface water. The organic compounds can be dissolved or suspended, and these are commonly called natural organic matter. Inorganic compounds such as mineral oxides and clay are derived from different processes in nature e.g., erosion. Removal of these components is important as they cause turbidity, and the surface of the particles may adsorb toxic components. The usual way for the removal is coagulation and flocculation followed by sedimentation (Crittenden, *et al.*, 2012).

#### 2.2.1 Natural organic matter

Decomposed molecules from animals and plants form a variety of molecules which are called natural organic matter. Depending on the origin, NOM is divided into two subcategories, aquogenic and pedogenic. Aquogenic refers to NOM originating from aquatic organisms and pedogenic to terrestrial NOM (Beckett & Ranville, 2006). Water bodies, especially surface water, contain NOM in different varieties and amounts. The characteristics depend on the pH of the water as well as origin of both organic matter and water. This means that location and time influence the composition of NOM (Sillanpää, 2014).

The components in NOM can be both hydrophilic and hydrophobic. Nearly 50 % of the total organic carbon (TOC) in water are hydrophobic acids which makes these the largest fraction of NOM. The hydrophobic acids are humic substances, and these are divided into three categories: humins, humic acids and fulvic acids (Sillanpää, 2014). Characteristics for the three categories

are conjugated double bonds, aromatic carbons, and phenols. The hydrophilic components mainly consist of nitrogenous compounds and aliphatic carbons, e.g., proteins and carboxylic acids (Matilainen, *et al.*, 2010). Due to the acidic functional groups, such as phenolic, and carboxylic groups, and pH of natural waters, NOM has a negative charge in surface water (Uy-guner-Demirel & Bekbolet, 2011). For fresh water, the pH ranges from 6.0 to 8.5 (Water Research Center, 2023).

It is desired to remove these not only for the yellow colour that may occur at high concentrations but also because these molecules react with disinfectants. In those reactions, by-products are formed that may be carcinogenic (Crittenden, *et al.*, 2012). Disinfectants can be UV, chlorination or ozonation and are used at water treatment plants as microbial barriers as mentioned above. Furthermore, as NOM is organic matter, it can be a food source for microorganisms and can cause issues in pipelines at the treatment plant or distribution pipelines (Persson, 2023).

#### 2.3 Removal of contaminants

The most common way to remove NOM is using coagulation and flocculation. In short, coagulation is the neutralisation process of charged particles and flocculation is when these neutralised particles aggregate together. By sedimentation or filtration, the large aggregates can be removed. The concepts described in sections 2.3 to 2.4 originate from Crittenden, *et al.*, 2012 if nothing else is stated.

#### 2.3.1 Surface charge

The surface charge is an important characteristic of particles in suspension as it allows the particles to continue to be suspended for a long period of time. However, particles will aggregate and settle if enough time is given but the time required is not reasonable for a sufficient drinking water production (Davis, 2019). NOM are negatively charged for most pH but below pH 3 the surface charge becomes positive. This means that the zero point of charge (ZPC) has been reached. ZPC is the pH at which the surface charge for a certain particle is zero. Below ZPC, the surface charge will be positive and above, the charge will be negative.

#### 2.3.2 Electrical double layer

Another crucial concept is the electrical double layer (EDL) and as the name imply, it consists of two layers. These two are the Helmholtz layer, and the diffuse layer and are depicted in Figure 4. For negatively charged particles, the Helmholtz layer (also called the Stern layer) consists of cations that through adsorption and electrostatic forces bind to negative charges on the particle surface. This results in an electrical field with a net negative charge in the diffusive layer, see Figure 4. Due to the net negative charge, anions are repelled, and cations are in excess. In the diffusive layer, ions are not in fixed positions and move due to diffusion and the layer ends where the electrical potential is zero.

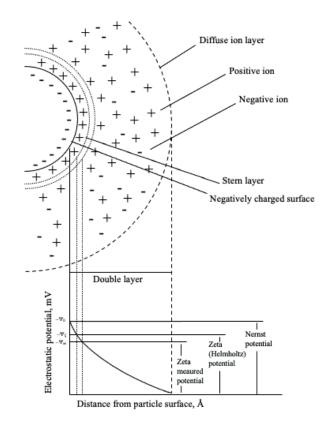


Figure 4: Electrical double layer. Adapted from Crittenden, et al., 2012.

#### 2.3.3 Zeta potential

During the movement of a particle in an electrical field, water close to the particle surface will follow and a shear plane is created. This plane lies in the diffusive layer and the measured electrical potential between the shear plane and the bulk is called zeta potential. Flocculation occurs once the zeta potential is reduced to a value below 20 mV.

#### 2.3.4 Particle stability

The balance between the attractive force (van der Waals forces) and repulsive electrostatic force affects the stability of particles. Due to electronic and magnetic resonance between two particles, van der Waals forces arise. The repulsive electrostatic forces between two particles originate from their negative surface charge. These repulsive forces, together with EDL result in an energy barrier that van der Waals forces cannot overcome on its own. For flocculation to happen, the energy barrier must be decreased, and this can occur by adding coagulants.

#### 2.4 Coagulation

By adding a coagulant, particle destabilisation occurs, and the most common mechanisms are, (1) compression of electrical double layer, (2) adsorption and charge neutralisation, (3) adsorption and interparticle bridging, and (4) enmeshment and sweep floc.

#### 2.4.1 Electrical double layer compression

By increasing the ionic strength of a solution, the double layer is compressed, i.e., the thickness is decreasing, and zeta potential is reduced as well. When the EDL is compressed, Brownian motion will result in interaction between particles and the particles can remain attached because of van der Waals forces. Due to the necessary ionic strength being larger than the limit in drinking water, this mechanism is not used in drinking water treatment.

#### 2.4.2 Adsorption and charge neutralisation

At pH 6 to 8, the majority of particles in natural waters are negatively charged. By using organic cationic polymers or hydrolysed metals salts as coagulants, charge neutralisation can take place and particle destabilisation occurs. When polymers are used, they can either function as coagulants on their own, or in a combination with inorganic coagulants to form particle bridging where the latter is most common. However, this is described in the following section. After addition of the optimal coagulant dosage, flocculation occurs. At the optimal dosage, less than half of the particle surface is covered. At higher dosages, particle stability appears once again due to reversal in surface charge.

#### 2.4.3 Adsorption and interparticle bridging

Polymer bridging is a complicated phenomenon. However, four different types of adsorption mechanisms can occur between particles and sites on polymer chains: (1) charge-charge interactions, (2) dipole interaction, (3) hydrogen bonding, and (4) van der Waals forces of attraction. Due to several sites on the polymer chain, many particles can interact with the same polymer, and the polymer creates a "bridge" between particles (Davis, 2019).

#### 2.4.4 Enmeshment and sweep floc

If coagulants contain metal, e.g., aluminium or iron, and the dosage is high, the metal ions form metal hydroxides which are insoluble and will precipitate (Davis, 2019). In these hydroxide precipitates, particles are enmeshed and can be removed. This is usually described as sweep flocculation as the precipitates sweep out the particles from the water (Duan & Gregory, 2003).

#### 2.4.5 Which mechanism occurs?

Charge neutralisation and sweep coagulation (sweep floc) are the two main processes. Charge neutralisation happens very quickly, between 0.01 and 1 seconds and it takes 1 to 7 seconds for sweep coagulation. As it is difficult to achieve the appropriate mixing for charge neutralisation, sweep coagulation is more common (Shestakova & Hansen 2023).

However, the operating mechanism also depends on coagulant dosage and colloid concentration and the relationship between these. In Figure 5, the relationship between the concentration of metal salts (Al(III) and Fe(III)) and the colloid concentration of the water, is depicted. This is for a constant pH and the appearance of the four zones may vary for different pH, but also for different coagulants. Figure 5 is shown here to visualise how the relationship can look like and that the mechanisms are involved at different conditions.

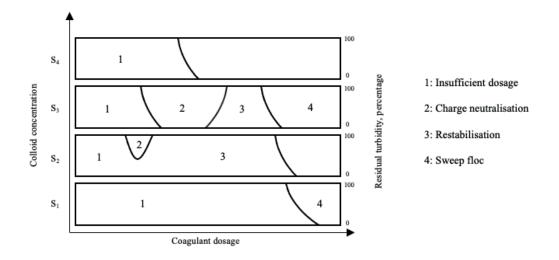


Figure 5: Residual turbidity for four different concentrations of colloids vs added coagulant. S<sub>1</sub> to S<sub>4</sub> represents different colloid concentrations (S<sub>1</sub> is the lowest and S<sub>4</sub> is the highest) and zones 1 to 4 symbolises different coagulation mechanisms. These curves are for a fixed pH. Adapted from Crittenden, et al., 2012.

At low colloid concentrations  $(S_1)$  a rather high dosage of coagulant is needed for sweep floc occur and prior to that the dosage is too low for anything to happen. At higher dosages  $(S_2)$ charge neutralisation occurs to some extent, but residual turbidity (measure of particulate matter in a solution) is still quite high. At higher coagulant dosages, restabilisation occurs and for even higher coagulant dosages, sweep floc is once again observed. For S<sub>3</sub>, charge neutralisation appears at higher coagulant dosage than for S<sub>2</sub>, but the removal of colloids is much higher. Even here, sweep floc is observed. Lastly, for S<sub>4</sub>, the zones for charge neutralisation and sweep floc blend together. This is because the needed coagulant concentration for charge neutralisation is the same as the required dosage for sweep floc to begin.

There are other parameters that indirect influence the coagulation by having a more direct effect on the amount of coagulant. These are pH, water hardness, temperature etc. When pH for instance increases, impurities such as NOM become more negatively charged and thus more coagulant is needed for destabilisation (Shestakova & Hansen, 2023).

#### 2.5 Coagulants

Coagulants can be either organic or inorganic. Cationic organic polymers have a repeating unit which can destabilise particles through charge neutralisation. If the polymers are anionic or non-ionic, a bridging mechanism is applied instead. These polymers are often used after metal salts (inorganic coagulants). Organic polymers which are positively charged are rarely used as a primary coagulant as they are not as efficient as inorganic coagulants. However, if used in combination with inorganic coagulants, the dosage of inorganic coagulants can be reduced. Anionic or non-ionic polymers can be used as flocculation aids (Crittenden, *et al.*, 2012).

Aluminium and iron salts are common inorganic coagulants. When these salts are added, dissociation to ions followed by formation of aquometal complexes such as  $Al(H_2O)_6^{3+}$  and  $Fe(H_2O)_6^{3+}$  occur. These complexes then go through various hydrolytic reactions and as a result various species, such as monomeric, polymeric, precipitates and even anionic species, are formed. A simplified reaction scheme is presented in reactions R1 and R2.  $Al(OH)(H_2O)_5^{2+}$  is monomeric,  $Al_{13}O_4(OH)_{24}^{7+}$  is polymeric and  $Al(OH)_3(s)$  and  $Al(OH)_{4^-}$  is a precipitate and anionic, respectively (Crittenden, *et.al.*, 2012).

$$Al(H_2O)_6^{3+} \leftrightarrow H^+ + Al(OH)(H_2O)_5^{2+}$$
 (R1)

$$Al(OH)(H_2O)_5^{2+} \stackrel{\uparrow H^+}{\longleftrightarrow} Al_{13}O_4(OH)_{24}^{7+} \stackrel{\uparrow H^+}{\longleftrightarrow} Al(OH)_3(s) \stackrel{\uparrow H^+}{\longleftrightarrow} Al(OH)_4^{-}$$
(R2)

As it can be seen from the reactions, protons are released in each reaction step which decreases the pH if the buffer capacity is insufficient. The buffer capacity can be explained by the amount  $OH^-$ ,  $CO_3^{2-}$  and  $HCO_3^-$  that can react with  $H^+$  and thus prevent the pH from dropping. The buffering capacity is also called the alkalinity of the water (Shestakova & Hansen, 2023). It is possible to increase the alkalinity by adding e.g., lime (Ca(OH)<sub>2</sub>) or caustic soda (NaOH) if the alkalinity is not enough for the buffering process (Crittenden, *et al.*, 2012).

At low pH values (below 4.5 for Al and 2.2 for Fe) the main species is  $Al(H_2O)_6^{3+}$  respectively  $Fe(H_2O)_6^{3+}$ . With increasing pH, the monomeric species become more abundant than previously (Pivokonsky, *et al.*, 2022). These are small and not that stable compared to polymeric species and precipitates. However, the ability for neutralisation is present as the charge per Al is high. When this species and NOM form complexes together, the complexes can be challenging to remove as they are soluble. The monomeric species may turn into polymeric species, and these destabilise NOM via charge neutralisation as well. Complexes consisting of  $Al_{13}$  are the most abundant for the polymeric complexes. The polymeric species in turn are converted to precipitates (i.e.,  $Al(OH)_3(s)$ ) and these are the most stable out of the three species. At pH 7, sweep coagulation for Al species is the main mechanism as the solubility is the lowest around neutral pH (Saxena, *et al.*, 2018). At higher pH (8.5 for Al and 10 for Fe), formation of the anionic forms occurs (Pivokonsky, *et al.*, 2022).

The temperature of the water affects the pH and thus has an effect on the coagulation and flocculation process. It has been noticed that low temperatures affected the processes negatively and have a larger effect for aluminium-based coagulants. The viscosity of water is also affected by the temperature which will influence the flocculation process (Pivokonsky, *et al.*, 2022).

As mentioned, many different complexes can be formed once a metal salt is added to water. It is also difficult to know and control which complexes are formed and this has led to pre-hydrolysed metal salts (Crittenden, *et al.*, 2012).

#### 2.5.1 Polyaluminium chloride

One example of a pre-hydrolysed metal salt is polyaluminium chloride, or PACl. It can be produced by neutralising aluminium chloride solution (AlCl<sub>3</sub>) (Duan & Gregory, 2003). When adding a base neutralisation occurs, and total neutralisation would lead to Al(OH)<sub>3</sub>. However, polymeric compounds are formed if partial neutralisation is achieved. PACl can also be formed when acid is added to Al(OH)<sub>3</sub> (Krupińska, 2020).

PACl is believed to contain large fractions of Al<sub>13</sub> and by already being partially neutralised, the decrease in pH is lower (Duan & Gregory, 2003). Other benefits with PACl are that the flocculation process is more rapid and enhanced performance at lower temperatures compared to coagulants which have not been pre-hydrolysed (Wei, *et al.*, 2015). The coagulation mechanism of PACl is not entirely clear yet, but charge neutralisation (due to Al<sub>13</sub> having high positive charge) and sweep floc are believed to occur (Duan & Gregory, 2003).

When producing these coagulants, the raw material is aluminium hydrate which is produced from bauxite (an aluminium ore) and hydrochloric acid (Kemira Oyj, 2011; Ruys, 2019a). According to a report from International Aluminium Institute (IAI), bauxite mining can be considered as footprint neutral when it comes to land area by having the same area for mine rehabilitation as for mining. Mine rehabilitation refers to returning the ecosystem to its original state before the mining started (International Aluminium Institute, 2008). However, the ore is still a finite resource (Ruys, 2019b) which may raise concerns about the overall sustainability of these coagulants.

#### 2.6 Flocculation

The theory behind flocculation is based on observations from Smoluchowski and Langelier. Smoluchowski noticed that when particles collide with fluid molecules, Brownian motion arises which in turn creates collisions between particles. Langelier observed that particle collisions occur due to velocity gradients when stirring the fluid. Microscale flocculation or perikinetic flocculation is when Brownian motion is the principal way of aggregation. Small flocs, from 1 to 100  $\mu$ m, are formed after a few seconds. Macroscale flocculation or orthokinetic flocculation occurs when mixing is used. Mixing may not only bring particles together but also break up flocs. A steady state between formation and breakup of flocs occurs after a period of mixing (Davis, 2019).

Floc growth can also depend on different settling velocities of particles. The settling velocity is dependent on the size and density of a particle and due to gravitational forces, larger particles settle faster. The different settling velocities are the reason for flocs at the different densities and/or sizes to collide. This is called differential settling and is a mechanism which is present during the settling phase in a heterogenous solution, such as a variety in density and size (Crittenden, *et al.*, 2012).

Due to attractive intermolecular forces such as hydrogen bonds or van der Waals forces, particles can bind together and create flocs. These attractive forces are limited by distance and coagulants are added to ensure collision and formations of flocs (Pivokonsky, *et al.*, 2022). When adding coagulant, rapid mixing is needed to ensure coagulation (Crittenden, *et al.*, 2012). During flocculation however, the mixing is at a lower intensity as flocs can break when shear forces are too high (Shestakova & Hansen, 2023).

#### 2.7 Handling of sludge

After the flocculation process, flocs will settle and create a sludge. Depending on the chosen disposal method, different methods of treatment to remove water prior to the disposal might be necessary. The first step is typically thickening which can remove a large amount of water in a fast manner. This is followed by a dewatering process (where more water is removed), which can either be mechanical or nonmechanical. In the mechanical process, water is removed by force by using some kind of equipment. In the nonmechanical way, sludge is spread out which results in that water is either draining or evaporating (Davis, 2019).

There are different ways to dispose the sludge where the most common ones are: letting it enter the sewage system, transferring it back to the water source or landfilling. Sending it to wastewater treatment plants (WWTP) has both its advantages and disadvantages. The dosage of chemicals at the WWTP can be decreased as the sludge adsorb some phosphate. However, sludge production in the WWTP increases and affects the disposal and the disposal cost of the WWTP. Transferring it back to a water source can affect the ecosystem around the outlet where the sludge has been released. The ecosystem can be negatively affected as sludge can build up and make the water turbid (Shestakova & Hansen, 2023). Landfilling is a convenient way, however with potentially stricter legislations and lack of places for landfilling, more sustainable ways of handling sludge are needed (Nguyen, *et al.*, 2022).

#### 2.7.1 Recovery of coagulant from sludge

One idea could be to recover coagulants from the sludge. Alkaline and acid treatments have been investigated for recovery of coagulants from water treatment sludge. However, the related costs for the recovery, the effectiveness of the process and quality of coagulants are factors yet to be determined (Ahmad, *et al.*, 2016). At pH between 1 and 3, around 80 % of the solid aluminium hydroxide can be dissolved and reused. During alkaline treatment, aluminium oxide can be recovered. Between pH values 11.4 and 11.8 with NaOH, 80 % can be recovered and with the less expensive Ca(OH)<sub>2</sub>, 30 % can be recovered. The disadvantage with using either of these treatments is that they are nonselective which can result in an increased amount of impurities in the final product (Nguyen, *et al.*, 2022). Besides, at low pH, NOM dissolve which can lead to disinfection by-products in later treatment steps (Keeley, *et al.*, 2012). These by-products may be carcinogenic as previously mentioned.

Another method of recovery is using membrane processes and an example of this is the Donnan membrane process, which is an ion exchange membrane. Advantages such as an improved quality and high recovery rate are observed compared to acidic treatments. However, long diffusion time is needed and while it can be reduced by increasing the membrane area, the cost of the membrane increases. Costs related to recovery of the membrane and disposal are other factors that affect the operational cost and must also be considered (Nguyen, *et al.*, 2022). Other membrane processes can also be used for coagulant recovery. Keeley *et al.* (2012) compared three membrane processes (Donnan membrane, electrodialysis and ultrafiltration) and concluded that external prices and demands regarding performance affect the operational costs and thus affect the feasibility of the recovery. It was also concluded that when acid treatment was incorporated in the process, the operational costs decreased with nearly 50 % (Keeley, *et al.*, 2012).

#### 2.7.2 Sludge at Sydvatten

The sludge that Sydvatten produces at Ringsjöverket contains iron residuals as the coagulant is FeCl<sub>3</sub> as previously mentioned. The sludge is put through a gravity thickener at the treatment plant where the dry content is increased from 0.5-0.8 % to 2 % followed by transportation without any pumping required to Rönneholms mosse (circa 5 km from Ringsjöverket) to the dewatering facility. In the dewatering process, water is pressed out from the sludge and the dry content has increased to 15-18 %. One part of the sludge is deposited on Rönneholms mosse and the other part is collected by biogas plants. The sludge itself is not environmentally hazard-ous but needs to be placed somewhere and it is of importance that it is placed near the water treatment plant to reduce costs and energy related to transportation (Sweco, 2022).

When producing biogas, bacteria such as methanogens digest organic material in anaerobic conditions. At these conditions, hydrogen sulphide is produced from sulphur containing substrates which affects the performances of the bacteria in a negative way. Added sludge which is rich in iron can bind the hydrogen sulphide and thus decrease the risk of disturbances in the biogas production (Persson, *et al.*, 2021).

## 3 Method

The idea was to simulate the large-scale process of coagulation and flocculation on lab-scale by using jar tests. Jar tests are commonly performed to evaluate parameters that affect the co-agulation and flocculation. The way these were performed in this thesis is described in a later paragraph (3.1). When performing the sludge recirculation experiments, the objective was to add different volumes of sludge and coagulant dosages to examine if the sludge had any effect on the treatment and what that effect would be. All tests were performed at Ringsjöverket using raw water from lake Bolmen, however a polyaluminium chloride coagulant from Kemira (PAX-XL60) was used instead of FeCl<sub>3</sub> which is used at Ringsjöverket when treating water from lake Bolmen.

As previously mentioned, the main reason for using an aluminium-based coagulant was because the results may provide an indication for future treatment at Vombverket. There are plans to construct a pipe from Ringsjöverket to Vombverket which will transport water from lake Bolmen to increase the redundancy and safety (Sydvatten, 2020). When water from lake Bolmen is treated at Vombverket, an aluminium-based coagulant may be an alternative for sufficient removal of NOM as the next treatment step will be infiltration which provides high levels of purity. However, the results could also potentially indicate a future role for the sludge produced at Ringsjöverket (Persson & Hägg, 2023). Another reason for using PAX-XL60 is that FeCl<sub>3</sub> requires a smaller range of possible pH in the solution than PAX-XL60 which makes it a bit trickier to use in jar tests.

Due to the usage of PAX-XL60, it was required to produce the sludge as Vombverket is not currently producing any aluminium sludge and the sludge from Ringsjöverket contains iron residuals. To simulate the actual process, sludge under optimal conditions had to be produced which led to that the experiments started with investigating at which pH and coagulant dosage optimal conditions were achieved. Once, these were known sludge could be produced and the recirculation experiments could begin.

#### 3.1 Jar tests

The jar tests were conducted with a flocculator (Flocculator 2000, Kemira). The flocculator simulated the different processes (coagulation, flocculation, and sedimentation) by changing the intensity of mixing after addition of coagulant. The same set-up was used throughout the project, with one litre beakers made out of glass, and the following procedure was used:

- 1. One litre of water was added to each of the beakers and either 0.1 M HCl or 0.4 M NaOH was added, depending on coagulant dosage and target pH for the specific beaker.
- 2. The flocculator started the rapid mixing (400 rpm) for 30 s and when 5 s remained, PAX-XL60 was added. This was followed by flocculation with slow mixing (50 rpm) for 20 min and measurement of pH. The next step was the 30 min sedimentation with no mixing.
- 3. Depending on the aim of the current experiment, which could either be analysing the effect of the coagulation/flocculation process or producing sludge for future experiments one of the following occurred:
  - a. With a 60 ml syringe, samples were taken from each beaker for measurements of absorbance. The samples were taken from the same depth in each baker for comparable results.

b. Sludge from the bottom of each beaker was transferred into a large flask.In Figures 6 and 7, the set-up and flocculator are depicted.



Figure 6: The set-up with three beakers and stirrers from Kemira connected to Flocculator 2000 from Kemira.

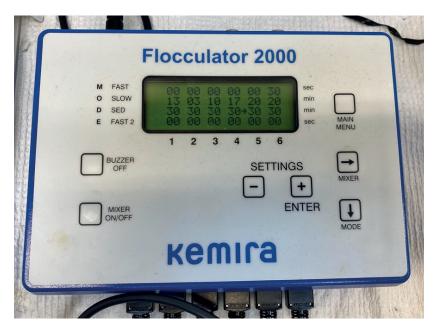


Figure 7: Flocculator 2000 from Kemira used in the experiments. The display shows a snapshot of an experiment, where beakers 1-5 are at different stages of the slow stirring and 6 has not been started yet.

Throughout the experiments, a 1 litre measuring glass was used for measuring the volume of water going into the beakers. PAX-XL60 was added by using an adjustable pipette and the tip was carefully wiped due to a substantial volume of coagulant got stuck on the outside of the tip. If this had not been done, a larger and an unknown dosage of coagulant could have been added

to the water. The equations Eq.1 and Eq.2 were used to calculate the volume of PAX-XL60 from a decided dosage.

$$\frac{\text{Dosage Al}}{\text{Fraction of Al in PAX-XL60}} = \text{Dosage PAX} - \text{XL60}$$
(Eq.1)

$$\frac{\text{Dosage PAX-XL60}}{\text{Density of PAX-XL60}} = V$$
(Eq.2)

Dosage Al and PAX-XL60 is in g Al/m<sup>3</sup> and g/m<sup>3</sup>, respectively. Fraction of Al in PAX-XL60 is 0.075 and the density of PAX-XL60 is 1.31 g/cm<sup>3</sup>. This resulted in a volume (V) in  $\mu$ l.

The effect of the coagulation and flocculation processes was analysed by measuring the absorbance in a spectrophotometer (DR 5000, HACH Lange). Light in both the visible and UV-range was sent through the cuvette and the absorbance was measured.

#### 3.2 Spectrophotometry

Absorbance measurements are commonly used when characterising NOM. Using light within the UV-Vis spectrum is a method that is simple and provide results quickly. NOM consist of a variety of aromatic structures and conjugated double bonds which absorb light in the UV range. Although different chromophores (part of a molecule which absorb light) absorb light at different wavelengths (Matilainen, *et al.*, 2011), 254 nm is commonly used for humic substances and 436 nm is used for the yellow colour representation (Abbt-Braun, *et al.*, 2004). A simplified sketch of spectrophotometry is shown in Figure 8.

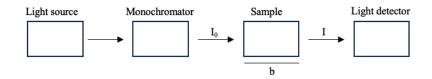


Figure 8: Overview of spectrophotometry. Adapted from Harris, 2015.

The wavelength from the light source which should pass through the sample is selected in the monochromator. The sample (with path length b) absorb light and the intensity before and after (I<sub>0</sub> and I, respectively) the sample are measured. From these measured values, the absorbance (A) can be calculated using equation Eq.3 (Harris, 2015). Typically, absorbance is unitless.

$$A = \log \frac{l_0}{L}$$
(Eq.3)

When evaluating the absorbance results, a low measured absorbance means that few functional groups have been detected. This means that given the conditions (coagulant dosage and volume of added sludge) as much as possible of NOM has been removed from the water and thus the sludge contains as much NOM as possible.

#### 3.3 Dosage optimisation

The coagulant dosage was the first parameter to be determined by using jar tests. The dosage varied from 1 to 10 mg Al/l. It was decided that pH should be close to 6 as most aluminium coagulants work well around this value.

Once PAX-XL60 was added to the water, pH would decrease. This meant that depending on the pH of the raw water and the dosage of coagulant, different volumes of HCl or NaOH would be required to reach the desired pH. In general, dosages 1 to 3 mg Al/l needed HCl, and dosages 5 to 10 mg Al/l required NaOH whereas dosage 4 mg Al/l did not need neither acid nor base. pH values between 5.9 and 6.1 were approved as it was difficult to reach exactly pH 6 each time. The absorbance was measured for evaluation.

#### 3.4 pH optimisation

Once the optimal dosage was set, the pH optimisation was next to be evaluated by using jar tests. The aim was to reach different pH values between 4 and 8 using the optimal dosage determined as described above. To reach the different pH values, the same solutions of HCl and NaOH were used as during the dosage optimisation. Even here, the absorbance was measured for evaluation.

#### 3.5 Sludge production

Once the optimal conditions, i.e., coagulant dosage, and pH, were known, the next step could proceed, which was creating sludge. This part was divided into two steps, where the first one was to determine how much sludge one beaker can produced during the optimal conditions. The second step was to produce as much sludge as possible and create a stock solution of sludge which could be used during the recirculation trials later on. In both of the steps, pH was measured to ensure that the appropriate pH was reached.

In the first step, three beakers were used and when the sedimentation time had expired, as much of the clear water as possible was removed without losing any sludge. To minimise the risk of losing sludge, a pipetboy was used to remove excess water. A total solids (TS) analysis was performed on the content from the three beakers. The sludge from each beaker was transferred to an aluminium form and was placed in an oven at 107 °C overnight. By measuring the weight of the empty form and the weight of the form and sample after the required time in the oven, the mass of sludge produced in one beaker could be determined, according to equation Eq.4. A mean value of the three  $m_{sludge,beaker}$  was calculated to give a more reliable value of the mass. All masses were in g.

$$m_{sludge,beaker} = m_{sample+form\ after\ oven} - m_{form}$$
(Eq.4)

The second step (which occurred at several different occasions) used the same optimal conditions as determined earlier and thus the same procedure as described above. Furthermore, for each occasion, raw water was retrieved from the same moment in time. As much as possible of the clear water phase was poured off and the remaining sludge solution from each beaker was poured into a flask, which was called stock solution. Three samples of 50 ml each was retrieved from the flask for a TS-analysis by using a 50 ml measuring glass. The TS-analysis was performed to determine the sludge concentration in the stock solution. The flask was turned upsidedown to create a homogeneous solution before the samples were taken as sludge would sink to the bottom with time. Figure 9 displays the difference between sedimented sludge and a more homogeneous solution.

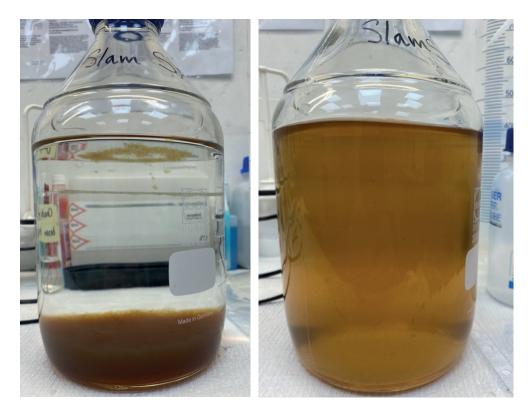


Figure 9: Difference between heterogenous and a more homogenous sludge solution.

The concentration of the sludge solution of each sample was determined using the equation Eq.4 to calculate  $m_{sludge,sample}$  ( $m_{sludge,sample}$  is  $m_{sludge,beaker}$  in this case), followed by equation Eq.5.

$$c_{sludge,sample} = \frac{m_{sludge,sample}}{V}$$
(Eq.5)

The volume (V) is 50 ml and the sludge concentration in the flask ( $c_{stock}$ ) is the same as the mean value of the three  $c_{sludge,sample}$  which is in g/ml.

#### 3.6 Recirculation of sludge

When the sludge concentration was determined, the experiments with sludge recirculation could proceed. Each beaker was filled with water and sludge solution so that the total volume would be 1 litre. The stock solution was once again turned upside-down to get a homogenous solution and by using measuring glass, a known volume could be poured into the beakers. This was followed by coagulation, flocculation, sedimentation and measuring of absorbance as described above. Combinations of different coagulant dosage and volume of recirculated sludge were tested. The added volume of recirculated sludge to each beaker was determined by the degree of recirculation (x), according to equation Eq.6 and Eq.7. In other words, the volume of added sludge corresponds to a weight percentage of sludge that one beaker could produce.

$$x * m_{sludge, beaker} = m_{recirculated sludge}$$
 (Eq.6)

$$\frac{m_{\text{recirculated sludge}}}{c_{\text{stock}}} = V_{\text{recirculated sludge}}$$
(Eq.7)

#### 4 Results and discussion

The results are divided into the categories described in the method chapter and are presented as well as discussed. For all graphs presented in this section, the corresponding VIS graphs are presented in the appendices A to C.

#### 4.1 Optimisation of dosage and pH

The residual absorbance results from the dosage optimisation with PAX-XL60 together with absorbance for untreated water are presented in Figure 10.

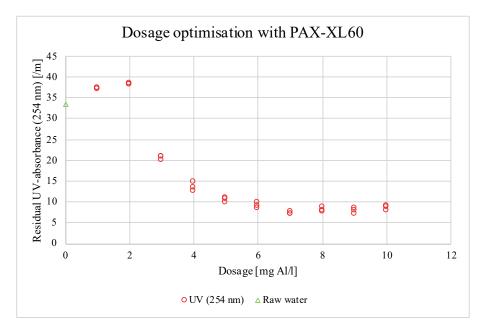
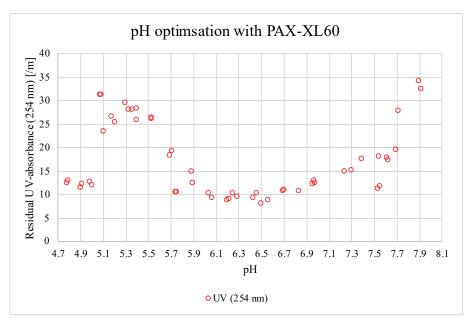


Figure 10: Residual UV-absorbance at 254 nm, for dosages of coagulant PAX-XL60 ranging between 1 and 10 mg Al/l at pH between 5.9 and 6.1. Absorbance for raw water is plotted as the green triangle.

At the lower dosages (1 and 2 mg Al/l), the absorbance is higher than for the untreated water and there is a slight increase at 2 mg Al/l compared to 1 mg Al/l. For dosages 3 to 5 mg Al/l the absorbance is decreasing and the residual absorbance for dosages 6 to 10 mg Al/l is at the lowest and most similar to each other.

The similar results in residual absorbance for dosages 6 to 10 mg Al/l suggested that removal of NOM would be rather similar in this dosage range. It was desired to use a low coagulant dosage to minimise the usage and cost of chemicals and therefore it was decided to use 7 mg Al/l as the optimal dosage. It can be argued that 6 mg Al/l would also work, however its residual absorbance was slightly higher than for 7 mg Al/l and the residual absorbance had a greater impact on the decision than the amount of coagulant. The residual absorbance indicates how much NOM is removed and a low value means that more has been removed compared to a higher value. Since it was desired to remove as much as possible (to get a sludge that contains as much NOM as possible), the residual absorbance had the greater influence on the dosage decision. It was desired to get a sludge with as much NOM residuals as possible to get an optimal sludge for the recirculation and thus to imitate the large-scale process at Ringsjöverket.

The most likely reason for the high residual absorbance for 1 and 2 mg Al/l (and higher than the untreated water) is that the coagulation was insufficient due to the dosages were too low, and it only meant that more particles was added to the water. It was also noted during the flocculation for these dosages that the flocs took longer time to form and were smaller compared to the other dosages which validates that the coagulation was not enough to remove NOM.



The absorbance results from the pH optimisation are presented in Figure 11.

*Figure 11: Residual UV-absorbance at 254 nm, for pH values varying between 4.8 to 7.9 with a dosage of coagulant PAX-XL60 of 7 mg Al/l.* 

According to Figure 11, a pH value between 6 and 7 is suitable for the dosage of 7 mg Al/l. pH values lower and higher than this range result in a residual absorbance that starts to increase towards the absorbance of the raw water, which was around 36 /m for these pH experiments.

Even though pH values between 6 and 7 gave similar results in residual absorbance it was decided that the optimal pH would range between 6 and 6.5. The reason for this decrease in range was because there was an observed increase in residual absorbance at pH 6.7 which continues to 6.9 which indicates that the removal of NOM is slightly lower than for pH 6 to 6.5. Similarly, to the dosage optimisation, it crucial to remove as much NOM as possible.

A decrease in residual absorbance below pH 5.2 can be observed. Furthermore, at pH 5 and lower, the residual absorbance decreased to levels similar to pH 6 to 7. This was unexpected and this effect was noticeable at different occasions. Although PAX-XL60 is a pre-hydrolysed coagulant and thus more control over the complexes is provided, it still goes through hydrolysation reactions, and it might be that at this pH a certain type of complexes was formed that contributed to a more successful flocculation than for the slightly higher pH.

#### 4.2 Sludge production

During the sludge production, the coagulant dosage was 7 mg Al/l and pH varied between 6.0 and 6.2. The amount of sludge that was produced in one beaker at these conditions was determined by TS-analysis. The results are shown in Table 1 below, together with the TS-content of sludge after the lamella sedimentation in the large-scale process at Ringsjöverket.

Table 1: Mass of dry sludge [g] produced in one beaker and the TS-content in one beaker during optimal conditions. TS-content of sludge after the lamella sedimentation in the large-scale process is also presented for comparison.

	Beaker 1	Beaker 2	Beaker 3	
Dry sludge [g]	0.038	0.037	0.033	
<b>TS in %</b> [g/g]	0.0038	0.0037	0.0033	
Large scale TS in %	0.0031			

The amount of sludge that was produced in one beaker is thus the mean value from the masses in Table 1 and is 0.036 g. The TS-content of the produced sludge was calculated by dividing the mass of dry sludge with the mass of the content in the beaker which was approximated to 1000 g. This resulted in a TS-content of 0.0036 % (mean value from the values in Table 1) and in large-scale production the corresponding TS-content is 0.0031 %. However, that sludge contains residues from FeCl<sub>3</sub> and not PAX-XL60, but it indicates that the sludge produced on labscale is comparable with the large-scale sludge.

Sludge was created at four different occasions and within each occasion the same raw water was used, i.e., the water was taken from the same point in time. This was to minimise potential parameters affecting the result of the recirculation. For each sludge production, a TS-analysis was performed from samples (of 50 ml) from the stock solutions for calculations regarding the sludge concentration. The dry weight of sludge from these samples and the corresponding concentration of sludge are presented in Table 2. The concentration of sludge in the stock solution is the mean value from the concentration of sludge from the corresponding samples.

Table 2: Results from sludge production, where three samples (of 50 ml each) for each sludge production have been analysed with TS-analysis. Dry weight of sludge [mg] and concentration of dry sludge [mg/l] for each sample are presented. The concentration of dry sludge for each stock solution [mg/l] is a mean value from the concentration of sludge from the three corresponding samples and these values are presented as well.

Date of pro- duced sludge	Sample no.	Dry weight of sludge [mg]	<b>Concentration of dry sludge</b> [mg/l]	Concentration of dry sludge in stock solution [mg/l]
	1	12.9	258	249
2023-03-08	2	12.4	248	
	3	12.0	240	
	1	13.2	264	
2023-03-13	2	13.5	270	265
	3	13.1	262	
	1	14.4	288	
2023-03-27	2	14.7	294	290
	3	14.7	288	
	1	14.2	284	
2023-04-03	2	14.3	286	285
	3	14.2	284	

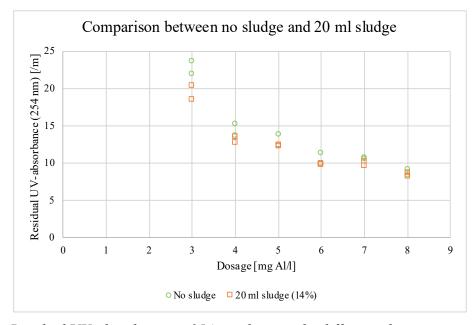
From Table 2, it can be observed that the concentration of dry sludge in the stock solution ( $c_{stock}$ ) does not change drastically between the different times when sludge was produced. However, for the first two occasions the concentration is slightly lower than for the last two. The reason for this might be that the technique when pouring off the water after sedimentation improved over time, which resulted in that more water could be poured off or less sludge was lost or a combination of both.

#### 4.3 Recirculation of sludge

The results regarding the sludge recirculation are divided into two categories: experiments to examine if any effect could be discovered and experiments when comparing two different co-agulant dosages and using sludge for the lower dosage. The results from these are presented below. For each performed experiment, the same raw water was used when comparing not using sludge with recirculating sludge in order to minimise parameters that could affect the results.

#### 4.3.1 Examining possible effects when recirculating sludge

The initial experiments with recirculating sludge are presented below, in Figure 12. Here, 20 ml sludge was added to 980 ml raw water. The used sludge volume corresponds to 14 wt% of sludge that can be produced from one beaker, i.e., the degree of recirculation is 14 %.



*Figure 12: Residual UV-absorbance at 254 nm showing the difference between using no sludge (green rings) and using 20 ml sludge (orange squares) for coagulant dosages between 3 and 8 mg Al/l. Sludge was produced on the 8<sup>th</sup> of March.* 

Figure 12 shows that the residual absorbance is lower when sludge has been added for coagulant dosages 3 to 6 mg Al/l. For the other two dosages, no change in residual absorbance can be observed.

In Figure 13, 30 ml sludge was used with 970 ml raw water. The volume sludge corresponds to a degree of recirculation of 22 %.

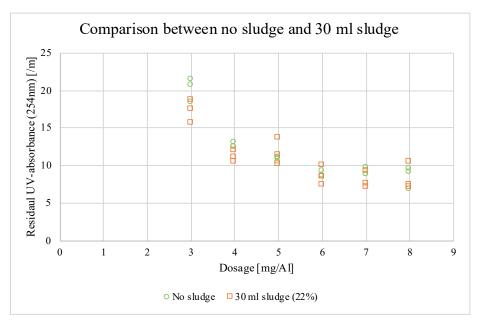


Figure 13: Residual UV-absorbance at 254 nm showing the difference between using no sludge (green rings) and using 30 ml sludge (orange squares) for coagulant dosages between 3 and 8 mg Al/l. Sludge was produced on the 13<sup>th</sup> of March.

Residual absorbance decreased when sludge was used for dosages 3 and 4 mg Al/l and for the other higher dosages, the decrease was not noticeable.

It can be observed that for a few measurements in Figure 13 the residual absorbance was higher when sludge was added compared to when only coagulant was used (e.g., at coagulant dosage 5 mg Al/l). This could be a result of something went wrong during the coagulation or flocculation. The coagulant could have been added after that standard time (which was when five seconds remained of the rapid stirring) and thus not had the same time to be distributed in beaker and encountered therefore less NOM. However, nothing which was out of the ordinary procedure was noted during the experiment.

During the sludge recirculation, a volume of sludge was added to first beaker, then the second and lastly the third. After this, pH adjuster (HCl or NaOH) was added, and the mixing started for beaker 1 which was followed by the adding of coagulant. This was then repeated for beaker 2 and 3. This meant that the sludge was in beaker 3 for a longer period of time than in beaker 1 before the coagulation started. It was performed in this way to ensure that the same homogenous sludge solution was added to each beaker (within the same coagulant dosage) as the sludge settled rather quickly in the larger flask (the stock solution). If one beaker was dealt with at the time, the sludge would settle a bit and the solution had to be turned upside down again and it was believed that it could have resulted in slightly different sludge solutions which might could have affected the results. By adding the sludge in the three beakers in the beginning this could be avoided. With this procedure in mind, it was noted that the residual absorbance was slightly higher for the first beaker compared to the other two. Moreover, for some coagulant dosages, the third beaker had a lower residual absorbance than the second beaker (especially for 5, 6 and 7 mg Al/l). This observation raised the concern if the time the sludge was in the beaker affected the flocculation and was therefore observed in the following experiments.

From these two initial trials, indication was given that the sludge affected the lower dosages of coagulant, i.e., 3-5 mg Al/l, as the absorbance for these dosages decreased, whereas for the higher dosages the difference in absorbance between using and not using sludge was not

particularly noticeable. This indicates that the sludge facilitates the flocculation for the lower dosages somehow, perhaps by providing flocs which the new neutralised NOM can aggregate onto. Since the sludge already contains destabilised NOM, it is more likely that the flocculation is enhanced rather than the coagulation.

The next set of experiments were conducted to investigate the impact of a low and high recirculation volumes, 10 ml respectively 60 ml. The results are presented in Figure 14. The volumes correspond to a degree of recirculation of 8 % and 48 %, respectively. The same procedure was used for adding the sludge as described previously and the time was noted to see if it had any effect on the flocculation.

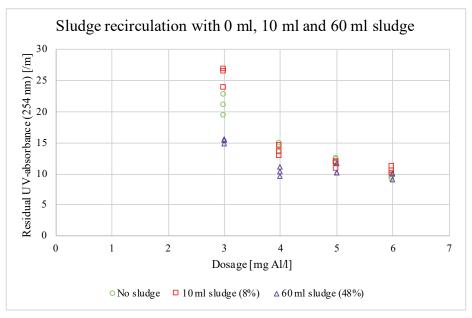


Figure 14: Residual UV-absorbance at 254 nm showing the difference between recirculating 0 ml (green rings) 10 ml (red squares) and 60 ml (purple triangles) sludge for coagulant dosage between 3 and 6 mg Al/l. Sludge was produced on the  $27^{th}$  of March.

The results show that once again the sludge has the greatest impact at the lower coagulant dosages, especially for 3 and 4 mg Al/l. This trend can be observed for 60 ml sludge for both coagulant dosages. However, for 10 ml sludge, the reverse effect was discovered for dosage 3 mg Al/l, but for 4 mg Al/l, the absorbance was comparable to or slightly lower than using no sludge. For dosages 5 and 6 mg Al/l no apparent differences between the three experiments were visible. Furthermore, coagulant dosage 3 mg Al/l together with 60 ml sludge resulted in absorbance measurements similar to coagulant dosage 4 mg Al/l. When using 60 ml sludge with 4 mg Al/l, the residual absorbance was lower than the residual absorbance for 5 mg Al/l without sludge. This indicates that using sludge and a lower coagulant dosage would result in similar absorbance result as a higher coagulant dosage but without using sludge.

The increase in absorbance at a low dosage of coagulant (3 mg Al/l) and low volume of added sludge (10 ml) suggests that the sludge may have a negative effect on the flocculation. This effect was unexpected and nothing in particular during the experiments were noted. It could be that the volume was too low and not providing enough flocs for the "enhanced" flocculation. As the coagulant dosage is rather low, it might be possible that the sludge only increased the concentration of organic matter which resulted in the higher residual absorbance. It could also be a coincidence. However, it was not investigated any further as no improvement (i.e., decreased absorbance) was noticed for the other dosages with this sludge volume. The trend with

using sludge at a low degree of recirculation and not noticing any difference was not entirely unexpected. As mentioned previously, Ringsjöverket currently recirculates backwashing water from the rapid sand filters and have noticed that when this water is split even over four flocculation basins the coagulant dosage is not affected. However, when the same volume is divided over two basins, the required coagulant dosage decreases. This suggests that if the amount of backwashing water is too low, no effect is observed and that it needs to be over a certain level to notice any effect. It might be possible that this applies for the recirculated sludge volume as well. It was therefore decided that recirculating 10 ml sludge (which corresponds to 8 wt%) would not be considered further in this thesis.

Once again three beakers were used for each coagulant dosage and sludge volume. The pattern where beaker 1 (which contained sludge for the shortest period of time) had the highest absorbance and the third beaker had the lowest absorbance was not noticed here. On the contrary, the number of the beaker with highest absorbance varied between the different coagulant dosages and sludge volumes and no obvious pattern was noticed.

There might be a possibility that the decrease in absorbance is a result from dilution of the raw water with the sludge solution as a smaller volume of raw water is used, instead of the sludge actually affecting the flocculation. The dilution effect was compared with the observed effect (decrease in absorbance). The observed effect was calculated by comparing the mean values of residual absorbance without and with sludge. The observed effect was only calculated for the critical dosage. The critical dosage was the coagulant dosage which gave the smallest decrease in absorbance, and it was 5 mg Al/l. Recirculation with 10 ml sludge was not included here as no improvement were observed. The dilution and observed effects are presented in Table 3.

	20 ml sludge	30 ml sludge	60 ml sludge	
Mean UV-absorbance				
5 mg Al/l [/m]	13.0	10.8	12.2	
5 mg Al/l (sludge) [/m]	12.3	10.4	10.8	
Effect				
Observed effect [%]	$\frac{13.0 - 12.3}{13.0} \times 100 = 5.4$	$\frac{10.8 - 10.4}{10.8} \times 100 = 4.9$	$\frac{12.2 - 10.8}{12.2} \times 100$ = 11.9	
Dilution effect [%]	$\frac{20}{980} \times 100 = 2.0$	$\frac{30}{970} \times 100 = 3.1$	$\frac{60}{940} \times 100 = 6.4$	

*Table 3: Comparison between dilution effect and observed effect for when recirculating 20ml, 30 ml and 60 ml sludge. Observed effect was calculated for the critical dosage which was 5 mg Al/l.* 

The improvement (i.e., decrease in absorbance) did not solely dependent on the dilution in these experiments, as the observed effect was higher than the dilution effect. However, the difference

is not very high, and it can depend on that only three jars were used for each sludge and coagulant dosage, which means the fluctuations in the measurements affect the mean value greatly. For the next set of experiments more jars were used to minimise this and provide a more reliable result.

#### 4.3.2 Examining if a lower coagulant dosage is possible when using sludge

The next experiment was conducted to compare the residual absorbance between coagulant dosage 4 mg Al/l, 4 mg Al/l with 60 ml sludge and 5 mg Al/l, see Figure 15, due to the results presented in Figure 14. For this, 15 beakers were used for each test compared to previous experiments to get a more accurate overview of the effect and to be able to use statistical tools in Excel. The sludge volume of 60 ml corresponds to a degree of recirculation of 47 %.

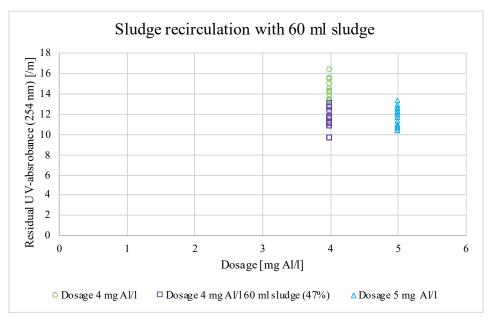


Figure 15: Residual UV-absorbance at 254 nm showing effect of 60 ml sludge at 4 mg Al/l (purples squares) compared to 4 and 5 mg Al/l with no added sludge (green rings and turquoise triangles, respectively). The sludge was produced on the  $3^{rd}$  of April.

The results show that adding 60 ml sludge to 940 ml water at coagulant dosage at 4 mg Al/l decreased the residual UV-absorbance compared to using the same dosage but with no sludge. A t-test for a confidence interval of 95 % was conducted to determine if this difference was significant (if so, p should be 0.05 or lower), and p is 8.406\*10^-8 which implies that the difference is significant and thus do not depend on coincidence. Furthermore, the absorbance at 4 mg Al/l with sludge are comparable with the values with 5 mg Al/l. This indicates that it might be possible to save 20 % coagulant dosage when 60 ml sludge is used, and it corresponds to a degree of recirculation of 47 %.

The final set of experiments were performed in the same manner as previously but with 30 ml added sludge. The results are presented in Figure 16 below. The sludge volume of 30 ml corresponds to a degree of recirculation of 24 %. Once again, 15 beakers were used for the same reason as described above.

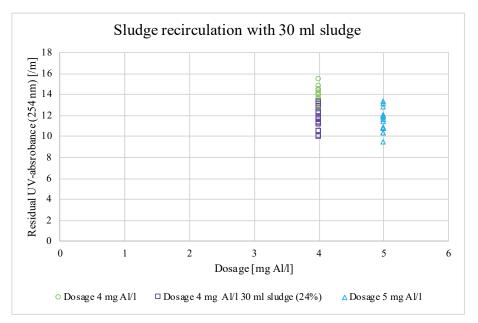


Figure 16: Residual UV-absorbance at 254 nm showing effect of 30 ml sludge at 4 mg Al/l (purple squares) compared to 4 and 5 mg Al/l with no added sludge (green rings and turquoise triangles, respectively). The sludge was produced on the  $3^{rd}$  of April.

The results from Figure 16 show that there is a significant difference between 4 mg Al/l and 4 mg Al/l with 30 ml sludge, as p was  $2.665*10^{-7}$  according to t-test performed in Excel. Even here, the residual absorbance for 4 mg Al/l with sludge is similar to the absorbance for 5 mg Al/l. These results indicates once again that sludge affected the flocculation and that it is possible to recirculate 24 % sludge instead of 47 % sludge.

Another way to examine the observed effect of the recirculated sludge is to calculate how much the absorbance decreases (between 4 mg Al/l and 4 mg Al/l with sludge) by using average values according to Table 4. As a smaller volume of raw water is used in the sludge recirculation it might be possible that the recirculated sludge dilutes the raw water and thus be responsible for the decrease in absorbance. By calculating the dilution effect (performed in Table 4) and compare it with the decrease in absorbance (the observed effect), it would be possible to understand if the dilution was the reason for the improvement or not.

	30 ml sludge	60 ml sludge		
Mean UV-absorbance				
4 Al mg/l [/m]	13.9	14.1		
4 Al mg/l (sludge) [/m]	11.6	11.4		
5 Al mg/l [/m]	11.5	11.7		
Effect				
Observed effect [%]	$\frac{13.9 - 11.6}{13.9} \times 100 = 16.5$	$\frac{14.1 - 11.4}{14.1} \times 100 = 19.1$		
Dilution effect [%]	$\frac{30}{970} \times 100 = 3.1$	$\frac{60}{940} \times 100 = 6.4$		

*Table 4: Comparison between recirculating 30 ml (24 wt%) and 60 ml (47 wt%) sludge.* 

For both of the two cases in Table 4 it can be seen that the dilution effect is 3.1 % and 6.4 % for recirculating 30 ml and 60 ml respectively, whereas the decrease in absorbance (observed effect) is 16.5 % and 19.1 % respectively. This indicates that the sludge actually influences the flocculation and the decrease in residual absorbance is not solely a result from dilution of the raw water. In other words, these lab-scale experiments have indicated that recirculating sludge has an effect and can enhance the flocculation. It has been noticed the coagulant dosage could be decreased with around 20 % even if the dilution effect has been considered and still reach the same absorbance results.

Despite using 15 beakers per sludge volume, three at the time were performed as previously described. The reason for this was to continue to investigate if the time the sludge spent in the beakers before the coagulation took place, had any effect. The pattern that was observed in the initial sludge recirculation experiment with 30 ml was not noticed here for either of the sludge volumes. This suggests that the time had no effect and that the pattern observed earlier possibly depended on coincidence.

#### **4.3.3** Potential savings in operational costs

The results showed that recirculating sludge could decrease the coagulant dosage. By decreasing the coagulant dosage, the operational costs connected to the coagulation will decrease as a result. To provide an estimation of the potential savings, costs from Ringsjöverket are used.

With an annual cost of approximately 6.8 million SEK (from 2022) for the FeCl<sub>3</sub> at Ringsjöverket, operational costs may be decreased with roughly 1.4 million SEK when decreasing the coagulant dosage with 20 % when recirculating sludge. This decrease would also affect the required amount of NaOH and the corresponding operational cost. NaOH is needed above a certain coagulant dosage (40 g/m<sup>3</sup>) to maintain pH and above 40 g/m<sup>3</sup>, the relationship between required dosage of NaOH and the dosage of FeCl<sub>3</sub> is linear. A decrease in coagulant dosage with 20 % would result in a decrease of 57 % of NaOH which would mean that the operational cost would decrease with 1.4 million SEK, as the annual cost of NaOH for 2022 was 3.3 million SEK. This means that 2.8 million SEK would be saved in operational costs regarding chemicals for the flocculation process. Other costs, such as capital costs for new pipes and pumps have not been investigated as that did not fall under the aim of this thesis as it is not a feasibility study.

The potential savings at Vombverket cannot be evaluated as coagulation and flocculation are not existing processes yet. Nevertheless, these results have provided some insights of the possible future of the sludge when water from lake Bolmen is treated at Vombverket.

## 4.3.4 Factors to be investigated further

If sludge recirculation were to be implemented in the large-scale process several benefits would happen such as less coagulant would be required for the treatment and as a result, less sludge would be produced and therefore needed to be handled. This would both affect the economics and the sustainability of the process.

However, these results are only an indication of the usage of sludge and there are several factors yet to be investigated. One of these is how or if the characteristics of the sludge change when it is being recycled and how the microbiological barrier is affected. It is important that there are sufficient microbial barriers in the treatment. The coagulation/flocculation step is one of them and it is important that this barrier is not altered with to still be able to ensure good quality of the drinking water. If it is tempered with, it must be compensated later in the process which perhaps would not be feasible. At the end of the day, the quality of the drinking water is more important than saving money. Another factor is the temperature which affects the viscosity and the flocculation. The experiments were performed in a room with a heater due to cold temperatures in the room. The pH meter also measured the temperature and the temperature of the water varied from 13 to 18 °C. The water will not have this temperature throughout the year and therefore it is of importance to investigate if the effect can be observed at lower temperatures as well.

Furthermore, if this were implemented at Ringsjöverket the iron content in the sludge leaving the plant needs to be examined as well. Since a smaller dosage of coagulant has been used, the produced sludge would contain a larger fraction of organic matter and smaller fraction of iron compared to the sludge which is produced now. This might result in an increase of the need for sludge at the biogas plants as the iron content would be lower. This in turn, could possibly lead to more transportation of sludge which may not be beneficial for the biogas plants in terms of cost and storage. This needs to be investigated to see if it would be possible to send the "new" sludge to the biogas plants or if other ways of handling it must be examined or if the only option is landfilling. This, together with results from the discussed factors above and the decrease in operational costs would determine the if recirculating sludge at Ringsjöverket is possible.

# **5** Conclusion

The aim was to investigate if there were any effects when water treatment sludge is recirculated in the flocculation process. This was performed at Ringsjöverket using water from lake Bolmen in Småland together with PAX-XL60 from Kemira as the coagulant. Sludge was produced under optimal conditions which were investigated prior to the sludge production. Coagulant dosage and pH were examined to establish these optimal conditions, and these were 7 mg Al/l and between 6 and 6.5, respectively, which answers the first research question.

When recirculating sludge using jar-tests (degree of recirculation was 22 %), residual UV-absorbance decreased for low dosages of coagulant (3-5 mg Al/l) compared to when using the same coagulant dosage without any sludge. For higher dosages (6-8 mg Al/l), the residual absorbance did not decrease. This indicates that the sludge has the greatest effect for the lower coagulant dosages.

Additional results show that residual absorbance decreased significantly when using 4 mg Al/l with sludge (degree of recirculation was 23 %) compared to when using the same dosage without any sludge. The dilution effect was around 3 % and the observed effect (decrease in residual absorbance) was around 17 %. This suggests that the decrease was not entirely a result of diluting the raw water with sludge and that the sludge enhanced the flocculation. Furthermore, using 4 mg Al/l with sludge resulted in residual absorbance similar to using 5 mg Al/l without sludge. This indicates that it is possible to lower the coagulant dosage by 20 % when recirculating sludge where the degree of recirculation is 23 %, which answers the second research question.

With this decrease in coagulant dosage and recirculation of sludge, less sludge needs to be handled. Additionally, when decreasing coagulant dosage, the required amount of pH adjuster (NaOH in this case) will decrease as well, and the operational costs connected to coagulation and flocculation will decrease. To put things in perspective, the operational costs could decrease with 2.8 million SEK at Ringsjöverket if sludge were recirculated at 23 % and coagulant dosage decreased with 20 %.

In other words, this thesis has indicated that sludge can be used as a resource in water treatment plants. This would be beneficial for both the economics and the sustainability of the process. However, before these benefits can be experienced, there are factors which need to be investigated. These are discussed in the next chapter, Future work.

# 6 Future work

Before recirculating sludge in the large-scale process, some factors need to be investigated. These has been mentioned in the discussion section previously.

One of these is to investigate how recirculating sludge affect the microbial barrier in the flocculation process. This could not be covered in this thesis but if using flow cytometry, microorganisms can be detected, and the process can be evaluated if it is feasible from a microbial barrier perspective. As mentioned previously, it is crucial to have microbial barriers to ensure that drinking water of good quality is produced, and this is more important than decreasing operational costs and compromising the quality.

Another factor to investigate is how temperature affects the flocculation when sludge is recirculated. It would be interesting to see how the sludge affects the flocculation in colder temperatures as the temperature affects the viscosity which affects the flocculation. The temperatures in the performed experiments varied between 13 and 18 °C and these temperatures will not be experienced throughout an entire year.

Furthermore, it is of interest to investigate other alternative ways of handling sludge for comparison, both economically and sustainably.

# 7 References

Abbt-Braun, G., Lankes, U. & Frimmel, F. H., 2004. Structural characterization of aquatic humic substances – The need for a multiple method approach. *Aquatic Sciences*, 66, pp. 151-170. <u>https://doi.org/10.1007/s00027-004-0711-z</u>

Ahmad, T., Ahmad, K. & Alam, Km, 2016. Sustainable management of water treatment sludge through 3'R' concept. *Journal of Cleaner Production*, 124, pp. 1-13. https://doi.org/10.1016/j.jclepro.2016.02.073

Beckett, R. & Ranville, J., 2006. Chapter 17 – Natural organic matter. In G. Newcombe & D. Dixon, ed. 2006. *Interface Science in Drinking Water Treatment: Theory and Application*. Cambridge: Academic Press, pp. 299-315. ISBN: 978-0-12-088380-6

Crittenden, J. C., Rhodes Trussell, R., Hand, D. W., Howe, K. J. & Tchobanoglous, G., 2012. *MWH's Water Treatment: Principles and Design*. 3<sup>rd</sup> ed. Hoboken: John Wiley & Sons. ISBN: 9781118131473

Davis, M. L., 2019. *Water and Wastewater ENGINEERING: Design Principles and Practice*. 2<sup>nd</sup> ed. New York: McGraw-Hill Education. ISBN: 978-1-260-13227-4

Duan, J. & Gregory, J., 2003. Coagulation by hydrolysing metal salts. *Advances in Colloid and Interface Science*, 100-102, 475-502. <u>https://doi.org/10.1016/S0001-8686(02)00067-2</u>

Harris, D. C., 2015. *Quantitative Chemical Analysis*. 9<sup>th</sup> ed. New York: W. H. Freeman and Company. ISBN: 978-1-4641-3538-5

International Aluminium Institute, 2008. *Fourth sustainable bauxite mining report*. [pdf] London: International Aluminium Institute. Available at: <u>https://bauxite.world-aluminium.org/fileadmin/\_migrated/content\_uploads/IV\_Sustainable\_Bx\_\_Mining\_Report.pdf</u> [Accessed at 2023-04-27]

Keeley, J., Jarvis, P. & Judd, S. J., 2012. An economic assessment of coagulant recovery from water treatment residuals. *Desalination*, 287, pp. 132-137. <u>https://doi.org/10.1016/j.de-sal.2011.09.013</u>

Kemira Oyj, 2011 *Kemira Oyj: Production of polyaluminium chloride restarted in Krems, Austria.* [press release] 17 October 2011. Available at: <u>https://www.kemira.com/company/media/newsroom/releases/kemira-oyj-production-of-</u> <u>polyaluminium-chloride-restarted-in-krems-austria/</u> [Accessed at 2023-04-27]

Krupińska, I., 2020. Aluminium Drinking Water Treatment Residuals and Their Toxic Impact on Human Health. *Molecules*, 25(3), 641. <u>https://doi.org/10.3390/molecules25030641</u>

Livsmedelsverket, 2023. *Mikrobiologiska säkerhetsbarriärer*. [online] Available at: <u>https://kontrollwiki.livsmedelsverket.se/artikel/339/mikrobiologiska-sakerhetsbarriarer</u> [Accessed at 2023-05-26]

Matilainen, A., Gjessing, E. T., Lahtinen, T., Hed, L., Bhatngagar, A. & Sillanpää, M., 2011. An overview of the methods used in the characterisation of natural organic matter (NOM) in relation to drinking water treatment. *Chemosphere*, 83(11), pp. 1431-1442. https://doi.org/10.1016/j.chemosphere.2011.01.018

Matilainen, A., Vepsäläinen, M. & Sillanpää, M., 2010. Natural organic matter removal by coagulation during drinking water treatment: A review. *Advances in Colloid and Interface Science*, 159(2), pp. 189-197. <u>https://doi.org/10.1016/j.cis.2010.06.007</u>

Nguyen, M. D., Thomas, M., Surapaneni, A., Moon, E. M. & Milne, N. A., 2022. Beneficial reuse of water treatment sludge in the context of circular economy. *Environmental Technology* & *Innovation*, 28, 102651. <u>https://doi.org/10.1016/j.eti.2022.102651</u>

Persson, T., 2023. Disussion about NOM. [conversion] (Personal communication, 2023-05-24)

Persson, T. & Hägg, K., 2023. *Discussion on using PAX-XL60*. [conversion] (Personal communication, 2023-05-12)

Persson, T., Persson, K. M. & Åström, J., 2021. Ferric Oxide-Containing Waterworks Sludge Reduces Emissions of Hydrogen Sulfide in Biogas Plants and the Needs for Virgin Chemicals. *Sustainability*, 13(13), 7416. <u>https://doi.org/10.3390/su13137416</u>

Pivokonsky, M., Novotná, K., Čermáková, L. & Petříček, R., 2022. Jar Tests for Water Treatment Optimisation: How to Perform Jar Tests – a handbook. London: IWA Publishing. ISBN: 9781789062694

Ruys, A., 2019a. 2 – Bauxite: The principal aluminum ore. In: L. Overned, ed. 2019. *Alumina Ceramics: Biomedical and Clinical Applications*. Sawston: Woodhead Publishing. ISBN: 978-0-08-102443-0

Ruys, A., 2019b. 3 - Refining of alumina: The Bayer process. In: L. Overend, ed. 2019. *Alumina Ceramics: Biomedical and Clinical Applications*. Sawston: Woodhead Publishing. ISBN: 978-0-08-102443-0

Saxena, K., Brighu, U. & Choudhary, A., 2018. Parameters affecting enhanced coagulation: a review. *Environmental Technology Reviews*, 7(1), pp. 156-176. https://doi.org/10.1080/21622515.2018.1478456

Shestakova, M. & Hansen, B., 2020. *About water treatment*. Helsinki: Kemira Oyj. ISBN: 978-951-97173-9-5

Sillanpää, M., 2014. Chapter 1 – General Introduction. In: M. Sillanpää, ed, 2015. *Natural Organic Matter in Water: Characterization and Treatment Methods*. Oxford: Butterworth-Heinemann, pp.1-15. ISBN: 978-0-12-801503-2

Svenskt Vatten, 2016. *Produktion av dricksvatten*. [online] Available at: <u>https://www.svenskt-vatten.se/fakta-om-vatten/dricksvattenfakta/produktion-av-dricksvatten/</u> [Accessed at 2023-01-24]

Sweco, 2022. *Utredning hantering vattenverksslam*. [pdf] Malmö: Sweco Sverige AB. Available at: <u>https://vattenbokhandeln.svensktvatten.se/wp-content/uploads/2023/01/Rapport\_utred-ning-hantering-vattenverksslam\_220518.pdf</u> [Accessed at 2023-05-03]

Sydvatten, 2020. *Bolmentunneln*. [online] Available at: <u>https://sydvatten.se/var-verksam-het/bolmentunneln/</u> [Accessed at 2023-05-05]

Sydvatten, 2021. *Delägarkommuner*. [online] Available at: <u>https://sydvatten.se/om-sydvat-ten/delagarkommuner/</u> [Accessed at 2023-02-23]

Sydvatten, 2023a. *Ringsjöverket*. [online] Available at: <u>https://sydvatten.se/var-verksam-het/vattenverk/ringsjoverket/</u> [Accessed at 2023-05-05]

Sydvatten, 2023b. *Sydvatten – collaborating for public welfare*. [pdf] Sydvatten. Available at <u>https://sydvatten.se/app/uploads/2023/04/Verksprocessr\_eng\_fo%CC%88r-hemsi-dan\_2023.pdf</u> [Accessed at 2023-01-24]

Sydvatten, 2023c. *Vombverket*. [online] Available at: <u>https://sydvatten.se/var-verksamhet/vat-tenverk/vombverket/</u> [Accessed at 2023-04-20]

Uyguner-Demirel, C. S. & Bekbolet, M., 2011. Significance of analytical parameters for the understanding of natural organic matter in relation to photocatalytic oxidation. *Chemosphere*, 84(8), pp. 1009-1031. <u>https://doi.org/10.1016/j.chemosphere.2011.05.003</u>

Water Research Center, 2023. *The pH of Water*. [online] Available at: <u>https://www.knowyourh2o.com/indoor-4/the-ph-of-water</u> [Accessed at 2023-01-26]

Wei, N., Zhang, Z., Liu, D., Wu, Y., Wang, J & Wang, Q., 2015. Coagulation behavior of polyaluminum chloride: Effects of pH and coagulant dosage. *Chinese Journal of Chemical Engineering*, 23(6), pp. 1041-1046. <u>https://doi.org/10.1016/j.cjche.2015.02.003</u>

# Appendices

## Appendix A: Residual VIS-absorbance for dosage and pH optimisation

In Figures 17 and 18, the residual VIS-absorbance for the dosage and pH optimisation, respectively, are presented.

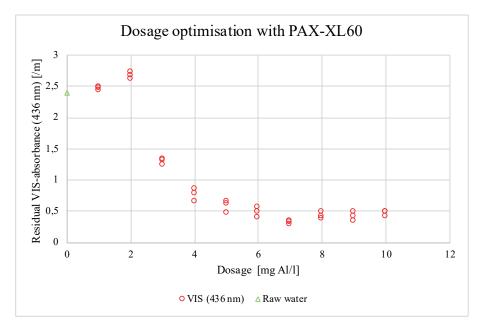
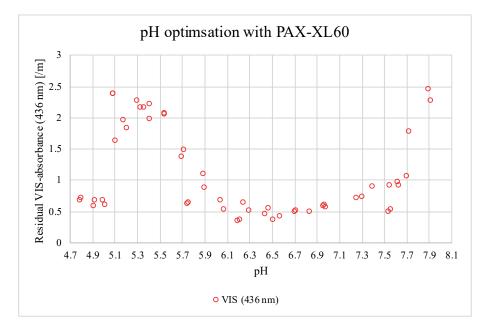


Figure 17: Residual VIS-absorbance at 436 nm, for dosages of coagulant PAX-XL60 ranging between 1 and 10 mg Al/l at pH value between 5.9 and 6.1. Absorbance for untreated or raw water is plotted as the green triangle.



*Figure 18: Residual VIS-absorbance at 436 nm, for pH values varying between 4.8 to 7.9 with a dosage of coagulant PAX-XL60 of 7 mg Al/l.* 

### **Appendix B: Residual VIS-absorbance for initial sludge recirculation**

Residual VIS-absorbance from the initial sludge recirculation experiments, where 20 ml and 30 ml sludge were used, are presented in Figures 19 and 20. These volumes correspond to 14 wt% and 22 wt%, respectively.

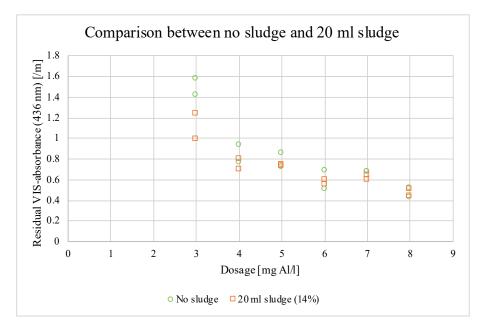


Figure 19: Residual VIS-absorbance at 436 nm showing the difference between using no sludge (green rings) and using 20 ml sludge (orange squares) for coagulant dosages between 3 and 8 mg Al/l. Sludge was produced on the 8<sup>th</sup> of March.

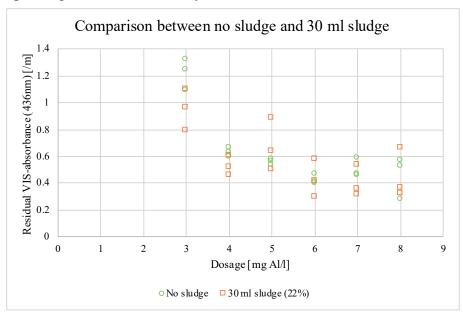


Figure 20: Residual VIS-absorbance at 436 nm showing the difference between using no sludge (green rings) and using 30 ml sludge (orange squares) for coagulant dosages between 3 and 8 mg Al/l. Sludge was produced on the 13<sup>th</sup> of March.

In Figure 21, residual VIS-absorbance is presented when 10 ml (8 wt%) and 60 ml (48 wt%) sludge were used to investigate the effect of using low and high volumes of sludge.

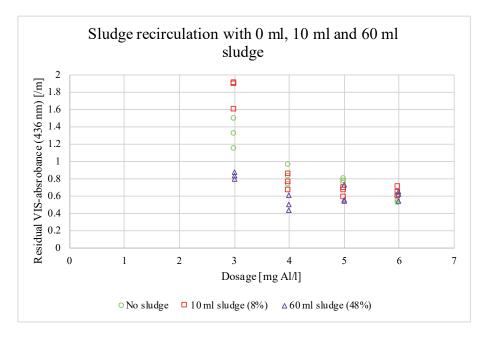


Figure 21: Residual VIS-absorbance at 436 nm showing the difference between recirculating 0 ml (green rings) 10 ml (red squares) and 60 ml (purple triangles) sludge for coagulant dosage between 3 and 6 mg Al/l. The sludge was produced on the  $27^{th}$  of March.

## Appendix C: Residual VIS-absorbance for final sludge recirculation

In this set of experiments, 4 and 5 mg Al/l was the chosen coagulant dosage and sludge (60 ml) was added to the lower dosage. The residual VIS-absorbance results are presented in Figure 22.

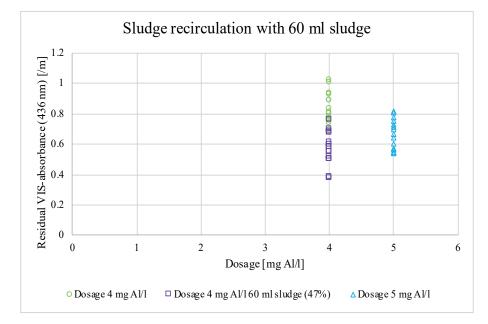


Figure 22: Residual VIS-absorbance at 436 nm showing effect of 60 ml sludge at 4 mg Al/l (purple squares) compared to 4 and 5 mg Al/l with no added sludge (green rings and turquoise triangles, respectively). The sludge was produced on the  $3^{rd}$  of April.

In Figure 23, residual VIS-absorbance is presented when 30 ml sludge was recirculated with coagulant dosage 4 mg Al/l and compared to when sludge was not added to dosages 4 and 5 mg Al/l.

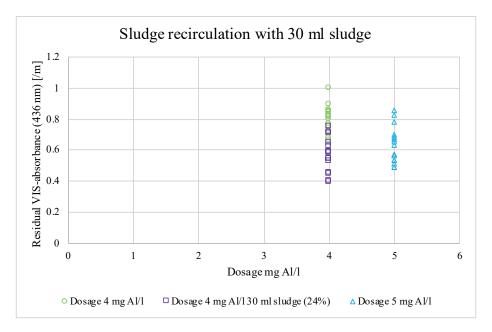


Figure 23: Residual VIS-absorbance at 436 nm showing effect of 30 ml sludge at 4 mg Al/l (purple squares) compared to 4 and 5 mg Al/l with no added sludge (green rings and turquoise triangles, respectively). The sludge was produced on the  $3^{rd}$  of April.

## Appendix D: Populärvetenskaplig sammanfattning

## Kan slam från vattenrening återanvändas hos vatten-verken för en mer hållbar dricksvattenproduktion?

# I takt med att befolkningen ökar, ökar även behovet av dricksvatten. Hur ska man möta det här behovet på ett hållbart sätt? Jo, genom att återanvända slam – en restprodukt från dricksvattenproduktionen.

I Sverige används både ytvatten från sjöar och grundvatten för att tillgodose befolkningens behov av dricksvatten. Då ytvatten generellt sätt är av lägre kvalitet än grundvatten behövs det fler reningssteg innan det är godkänt för konsumtion. Ett vanligt tecken på att ytvatten är av lägre kvalitet är att vattnet har en gul-brun färg. Det beror på att vattnet innehåller organiskt material och det är något som vattenverken behöver ta bort i reningen.

Kemisk fällning är ett vanligt reningssteg hos många vattenverk. Kemisk fällning innebär att kemikalier, till exempel olika metallsalter, tillsätts i stora reningsbassänger och klumpar ihop det organiska materialet. Dessa klumpar kan sedan sjunka till botten och kan på så vis avlägsnas från vattnet. Detta kallas slam och är en restprodukt som måste tas hand om. Deponi i naturen nära vattenverket är en vanlig hantering av slam. Slammet innehåller inte några miljöfarliga ämnen men tar upp plats i naturen och för att främja en mer hållbar produktion av dricksvatten är det av intresse att hitta ett nyttigt ändamål för slammet. För att reningen ska bli ännu mer hållbar är det också intressant att undersöka om det går att minska mängden metallsalter eftersom metallerna oftast utvinns av malm som är en ändlig resurs.

Den här studien har gjorts i samarbete med Sydvatten AB för att undersöka om det går att recirkulera slammet i processen i hopp om att kunna minska kemikaliedosen som behövs för reningen. Försöken har gjorts på Ringsjöverket, som är ett av vattenverken som Sydvatten äger. Försöken har gjorts så att de ska efterlikna den storskaliga processen genom att återskapa den kemiska fällningen i olika bägare. Olika mängden kemikalier och volymer av slam har analyserats för att se om slammet har någon önskvärd effekt på fällningen. Resultaten från denna studie har visat att en del av slammet kan användas i reningsprocessen och mängden kemikalier kan sjunka med 20 %.

Flera fördelar kan uppmärksammas genom att recirkulera en del av det slam som produceras i verken i reningsprocessen. Genom att recirkulera slam kan både mängden kemikalier och producerat slam minskas. Detta gynnar processen ur ett miljömässigt och ekonomiskt perspektiv eftersom mindre slam kommer att behövas hanteras och en mindre mängd kemikalier behövs.

Innan slamåteranvändning kan införas i fullskala på vattenverken behöver en del försök göras för att säkerställa att resten av produktionen inte störs av slammet och att den önskvärda kvaliteten hos vattnet fortfarande uppnås i de senare reningsstegen. I slutändan är det viktigaste ändamålet att producera dricksvatten av god kvalitet och inte att spara pengar. Resultaten från studien har dock indikerat att slammet kan ses som en resurs för vattenverken och kan bidra till en mer hållbar reningsprocess.



LUND UNIVERSITY Faculty of Engineering Department of Chemical Engineering