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Municipal wastewater treatment by microsieving, microfiltration and forward osmosis

Concepts and potentials

HEY, TOBIAS

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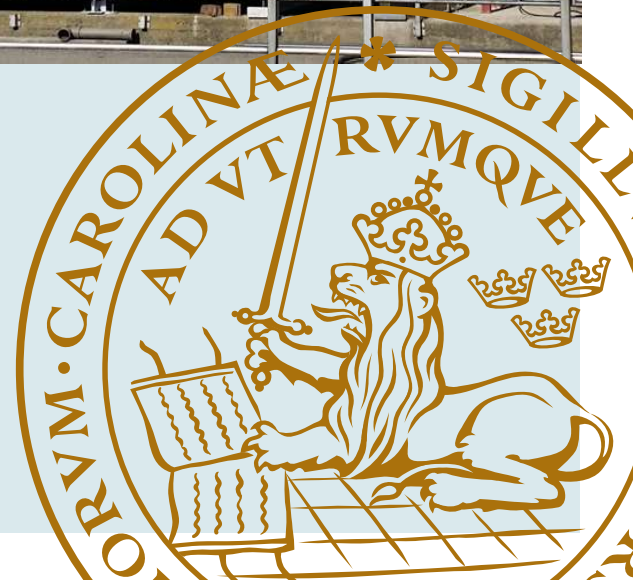
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FACULTY OF ENGINEERING | LUND UNIVERSITY



Municipal wastewater treatment by microsieving, microfiltration and forward osmosis

Concepts and potentials

Tobias Hey



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DOCTORAL THESIS

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| Title and subtitle Municipal wastewater treatment by microsieving, microfiltration and forward osmosis – Concept and potentials | | |
| Abstract <p>Conventional wastewater treatment plants are designed for treating manmade wastewater (e.g., from households and industries) and to protect the environment (e.g., receiving water bodies) and humans from adverse effects.</p> <p>The objective of this work was to investigate the feasibility of treating municipal wastewater without a biological treatment step by applying different separation processes, such as microsieving, microfiltration and forward osmosis. The scope of this work was to treat municipal wastewater with a lower area demand while meeting the Swedish wastewater discharge requirements and allowing for the integration of the new separation techniques with existing full-scale wastewater treatment plants. To achieve these goals, pilot-plant and bench scale studies were conducted using raw municipal wastewater on-site at a full-scale wastewater treatment plant.</p> <p>Two different treatment concepts were identified to be feasible for municipal wastewater treatment based on the experimental findings. The first concept comprised coagulation and anionic flocculation before microsieving with subsequent microfiltration. The second concept only included microsieving and forward osmosis. Both concepts were evaluated for their specific electricity, energy and area demands, including sludge treatment, and were compared with five existing conventional wastewater treatment plants.</p> <p>Both concepts complied with the Swedish wastewater discharge demands for only small- and medium-sized wastewater treatment plants because up to only 65% of the nitrogen was retained. Nevertheless, both concepts achieved high retentions, with $\geq 96\%$ for biochemical oxygen demand, $\geq 94\%$ for chemical oxygen demand, and $\geq 99\%$ for total phosphorus. Furthermore, the evaluation of both concepts showed that the specific electricity demand was 30% lower than the average specific electricity demand for 105 traditional Swedish wastewater treatment plants with population sizes of 1 500-10 000. In addition, the specific area demand could be reduced by at least 73% for existing wastewater treatment plants supporting the same population or a population of equivalent magnitude. Moreover, the results indicated that the new method had positive effects on electricity and energy due to the increased biogas potential compared to conventional wastewater treatment.</p> | | |
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Municipal wastewater treatment by microsieving, microfiltration and forward osmosis

Concepts and Potentials

Tobias Hey



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*“Where I am going you cannot follow me now,
but you will follow afterward.”*

(John 13:36)

Preface

During my PhD studies, I considered municipal wastewater treatment in two parts.

In the first part, I conducted full-scale in-line primary sludge hydrolysis experiments at the Klagshamn wastewater treatment plant. The objective of this study was to investigate the generation of an internal carbon source resulting from volatile fatty acid production for total nitrogen removal. The production of internal carbon sources at the Klagshamn wastewater treatment plant potentially reduced the demand for an external carbon source and reduced costs by 50%. Hence, carbon dioxide emissions from the production of an external carbon source were reduced. This part of my PhD-study has led to five publications. Four of these papers were included in my technical licentiate dissertation, which was defended in June 2013.

Jönsson K., **Hey T.**, Norlander H. and Nyberg U., 2009. Impact on gas potential of primary sludge hydrolysis for internal carbon source production. Proceeding of the 2nd IWA Specialized Conference Nutrient Management In Wastewater Treatment Processes, 6-9 September 2009, Krakow, Poland. ISBN: 9781843395775, pp. 459-466.

Hey T., Jönsson K. and la Cour Jansen J., 2012. Full-scale in-line hydrolysis and simulation for potential energy and resource savings in activated sludge - a case study. *Environmental Technology* 33(15), 1819-1825.

Hey T., Jönsson K. and la Cour Jansen, J, 2012. Calibration of a dynamic model for prediction of the potential of combined in-line hydrolysis with predenitrification at a full scale plant. *SNE* 22(3-4), 115-120.

Hey T., Sandström D., Ibrahim V. and Jönsson K., 2013. Evaluating 5 and 8 pH-point titrations for measuring VFA in full-scale primary sludge hydrolysate. *Water SA* 39(1), 17-22.

Hey T. (2013) Carbon utilisation for extended nitrogen removal and resource savings. [Technical licentiate dissertation]. Lund University, 2013.

Ibrahim V., **Hey T.** and Jönsson K. 2014. Determining Short Chain Fatty Acids in Sewage Sludge Hydrolysate: A Comparison of Three Analytical Methods and Investigation of Sample Storage Effects. *Journal of Environmental Sciences* 26(4), 926-933.

In the second part, wastewater treatment in terms of resource savings remained an area of the focus to determine if municipal wastewater can be treated to comply with Swedish wastewater discharge demands without a biological treatment step. A pilot-scale plant was built on site at the Källby WWTP in Lund, where real raw municipal wastewater was treated by using microsieving, microfiltration and forward osmosis.

Microfiltration experiments were conducted at the pilot scale by applying only the feed's hydrostatic pressure as the trans-membrane pressure. This low trans-membrane pressure was applied to obtain low electricity demand and low fouling propensities in comparison to the higher trans-membrane pressures found in direct membrane filtration.

Forward osmosis experiments were performed using two different forward osmosis membranes. One membrane was a biomimetic membrane and had not been tested earlier for municipal wastewater treatment purposes. Hence, this work presents a novel technology for treating municipal wastewater. In addition to the use of an artificial draw solution, seawater was tested when using the forward osmosis membrane.

Two treatment concepts were selected and evaluated for specific electricity, energy and area demands and compared with conventional wastewater treatment.

Acknowledgements

A PhD thesis usually ends with the outcome of the study, but after more than eight years of my industrial PhD, I can already conclude at the beginning of writing my thesis that it has been a truly life-changing experience for me.

However, this long journey has only been possible with financial support from VA SYD, Sweden Water Research, VINNOVA, Region Skåne Biogas and Swedish Water and Wastewater Association through via VA-teknik Södra.

Nevertheless, during this long journey I have received guidance and support that have made my journey easier in many ways from several people. Specifically, I am deeply grateful for the following:

First, I would like to express my sincere gratitude to Karin Jönsson and Jes la Cour Jansen. I was privileged to have ‘doctor parents’ that shared their great passion about wastewater treatment and who ensured that I remained on the track of science and they served as a firm rock when the sea got stormy.

Åsa Davidsson and Michael Cimbritz for their support and many discussions about particles, separations and the conversion of everything to biogas. For your warm companionship and help in many ways.

Henrik Aspegren, Ulf Nyberg, Monika Erlandsson and Liselotte Stålhandske for taking care of administration issues and ensuring that I could fulfil my industrial PhD, which was not an easy task.

Janne Väänänen for your companionship. Without you, my office at Källby would not have been filled with great and fruitful discussions, including explicit coffee research and BBQ sessions.

Niada Bajraktari, for our long discussions and your support considering forward osmosis on the phone, in Denmark, or in Sweden when you were in need of Swedish cheese and cinnamon buns. However, I am most thankful for your friendship, our abilities to cheer each other up, and for sharing our laughter and tears.

Jörg Vogel, my fellow country man, for sharing knowledge and ideas and for setting me up with forward-osmosis ‘stuff’. Your great support when things did and did not work out, and the on-site visits in Sweden.

Nicolas Heinen and Claus Hélix-Nielsen for giving me the opportunity to collaborate with Alfa Laval and Aquaporin, respectively and for providing discussions about membrane processes.

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Misagh Mottaghi: we started as roommates and continue as friends. Thank you very much for the great and deep discussions about everything excepting wastewater treatment and for all the good and addictive Persian pastries.

A pilot-plant is like a small child, the moment you turn your back something happens. Because I am not a superman, I received a lot of help from Anders Arkell, Daniel Henriksen, Jan Svensson, Lars-Erik Svensson, Lilla Harrie ‘Boys’ (LHP), Malin Göthrick, Mattias Jönsson, Per Härnäs, Per-Åke Rask and Peter Nilsson to build up the pilot plant and make it run.

Furthermore, since I used microsieve filters, microfiltration and forward osmosis membranes, I received a lot of help and support from Carles Pellicer-Nàcher and Pille Kängsepp from Hydrotech, Dongxu Chen, Emmanuel Joncquez and Jessica Bengtsson from Alfa Laval, Oliver Geschke and Sylvie Braekevelt from Aquaporin.

I am very thankful to the VA SYD laboratory team for analysing and discussing the results of my samples and for understanding when the pilot plant had its defiant moments that resulted in no samples even when everything was scheduled.

I am very grateful to my colleagues from VA SYD, VA-teknik Södra and at the Department of Chemical Engineering at Lund University for always providing me with a warm welcome.

As I was part of the project ‘The warm and clean city’, I would like to say “Thank you!” to my partners and the reference group for sharing their activities and thoughts in this project, which provided me with further insights and allowed me to think more outside of the box.

Hans Thullner and the City of Gärtringen for accepting me as an apprentice when I had no knowledge of wastewater treatment. Hans Thullner for your warm personality, enthusiasm and passion about wastewater treatment and for laying the very first brick in my wastewater career.

Kenth Hansson for your support in many ways and for always looking after me and my family.

Stefan Raap. Good friends are like stars, you do not always see them, but you know they are always there. Thank you for always being there.

My lovely family in Sweden, Germany and the Philippines. For your love and support and for filling every day with wonderful life.

Abstract

Conventional wastewater treatment plants are designed for treating manmade wastewater (e.g., from households and industries) and to protect the environment (e.g., receiving water bodies) and humans from adverse effects.

The objective of this work was to investigate the feasibility of treating municipal wastewater without a biological treatment step by applying different separation processes, such as microsieving, microfiltration and forward osmosis. The scope of this work was to treat municipal wastewater with a lower area demand while meeting the Swedish wastewater discharge requirements and allowing for the integration of the new separation techniques with existing full-scale wastewater treatment plants. To achieve these goals, pilot-plant and bench scale studies were conducted using raw municipal wastewater on-site at a full-scale wastewater treatment plant.

Two different treatment concepts were identified to be feasible for municipal wastewater treatment based on the experimental findings. The first concept comprised coagulation and anionic flocculation before microsieving with subsequent microfiltration. The second concept only included microsieving and forward osmosis. Both concepts were evaluated for their specific electricity, energy and area demands, including sludge treatment, and were compared with five existing conventional wastewater treatment plants.

Both concepts complied with the Swedish wastewater discharge demands for only small- and medium-sized wastewater treatment plants because up to only 65% of the nitrogen was retained. Nevertheless, both concepts achieved high retentions, with $\geq 96\%$ for biochemical oxygen demand, $\geq 94\%$ for chemical oxygen demand, and $\geq 99\%$ for total phosphorus. Furthermore, the evaluation of both concepts showed that the specific electricity demand was 30% lower than the average specific electricity demand for 105 traditional Swedish wastewater treatment plants with population sizes of 1 500-10 000. In addition, the specific area demand could be reduced by at least 73% for existing wastewater treatment plants supporting the same population or a population of equivalent magnitude. Moreover, the results indicated that the new method had positive effects on electricity and energy due to the increased biogas potential compared to conventional wastewater treatment.

Populärvetenskaplig sammanfattning

Från 1950-talet fram till millennieskiftet ökade världens befolkning från tre till sex miljarder och 2038 förväntas vi vara 9 miljarder människor. Redan idag lever mer än 50 % av alla människor i städer och med nuvarande urbaniseringstakt kommer två tredjedelar att bo i städer år 2050. I och med den kraftiga urbaniseringen kommer också kraven på avloppsrening att öka för att motverka negativa effekter på miljö och hälsa, t.ex. i form av algblomning i vattendrag.

I Sverige behandlas avloppsvatten i princip genom mekanisk, biologisk och kemisk rening. Därigenom reduceras våra utsläpp av organiska och syreförbrukande ämnen samt näringsämnen i form av kväve och fosfor.

I det mekaniska steget tas allt bort som vi slänger i våra toaletter t.ex. toapapper, dambindor, kondomer och t.o.m. kläder som annars skulle orsaka störningar i reningsprocesserna. I det biologiska reningssteget finns mikroorganismer, t.ex. bakterier, som har förmåga att omvandla kol till koldioxid och kväve till kvävgas. För att kunna göra det så behöver bakterierna syre som tillsätts med hjälp av energikrävande blåsmaskiner. Det biologiska steget kräver därför mycket energi men också plats för att bakterierna ska kunna göra sitt jobb. Det tredje reningssteget är den kemiska reningen där olika kemikalier används för att avskilja fosfor. Efter att avloppsvattnet har gått igenom alla tre reningssteg, där innehållet av kol, kväve och fosfor kraftigt har reducerats, så släpps det renade vattnet ut i ett vattendrag, men trots det är jobbet inte riktigt klart. I alla tre reningssteg genereras slam. Detta behandlas ofta i en s.k. rötkammare där olika sorters bakterier finns med förmågan att omvandla slammets innehåll av kol till biogas. Biogasen kan användas för att producera elektricitet, värme och till och med fordonsgas. Denna används för att driva stadsbussarna i många städer.

I och med att mer avloppsvatten kommer att genereras i städerna så kan följden bli att kapaciteten för befintliga avlopprensingsreningsverk behöver utökas. Samtidigt förväntas skärpta reningskrav med tanke på andra föroreningar som finns i avloppsvatten, t. ex. olika läkemedelsrester, och ytterligare reningssteg behövs därför. Konsekvenserna av den ökade urbaniseringen och de strängare utsläppskraven är därför att både mer plats och energi sannolikt kommer att behövas.

I vilken utsträckning är det möjligt att rena avloppsvatten utan den plats- och energikrävande biologiska reningen? En möjlighet är att använda olika fysikaliska tekniker som kan avskilja ämnen, från en storlek som är större än ett hårstrå till en storlek som man inte ens kan se i ett mikroskop. Dessa tekniker kan liknas vid flera silar i rad med allt mindre öppningar. De olika siltyperna som testats i detta projekt utgör exempel på mikrosilning, mikrofiltrering och framåtriktad-osmos (på engelska *forward-osmosis*). Mikrosilning blockerar partiklar som är större än 100 μm t.ex. sand. Mikrofiltrering har öppningar på 0.2 μm vilket är mindre än de allra flesta bakterier och framåt-osmos avskiljer t.o.m. virus. Framåtriktad-osmosen har särskilda öppningar s.k. akvaporiner som härmar vattentransporten i våra celler. Framåtriktad-osmosen drivs med hjälp av en koncentrationsskillnad. På den ena sidan finns avloppsvatten som har en låg salthalt och på den andra sidan av silen finns havsvatten som har en högre salthalt än avloppsvattnet. Vattenmolekyler i avloppsvattnet vandrar sedan genom dessa akvaporiner till sidan med havsvatten för att jämna ut saltkoncentrationen. Naturen strävar efter jämvikt! Medan vattenmolekyler lämnar sidan med avloppsvatten ökar koncentrationen av olika ämnen, t.ex. fosfor, på denna sida av membranet. På det här viset stannar t.ex. kol och fosfor i avloppsvattnet medan det rena vattnet transporteras till membranets andra sida.

Det är fullt möjligt att rena avloppsvatten på mekanisk väg. Tekniken är i första hand intressant för små- och medelstora reningsverk som har utsläppskrav för kol och fosfor. Kväve avskiljs däremot inte i tillräckligt hög grad, vilket oftast är nödvändigt för stora reningsverk. Samtidigt kan mer biogas produceras, så mycket att anläggningens hela elektricitetsbehov kan täckas. Samma mängd avloppsvatten kan behandlas på minst halva ytan jämfört med befintliga avloppsreningsverk. Förhoppningen är att arbetet ska kunna bidra till en mer effektiv avloppsvattenrening där både energi och plats kan sparas.

List of papers

This thesis comprises the following original papers in chronological order, which are referred to in the text by their Roman numerals I-IV.

- Paper I** **Hey T.**, Väänänen J., Heinen N., la Cour Jansen J., and Jönsson K. Potential of combining mechanical and physicochemical municipal wastewater pre-treatment with direct membrane filtration. *Environmental Technology*. DOI: 10.1080/09593330.2016.1186746
- Paper II** **Hey T.**, Zarebska A., Bajraktari N., Vogel J., Hélix-Nielsen C., la Cour Jansen J. and Jönsson K. Influence of mechanical pre-treatments on non-biological treatment of municipal wastewater by forward osmosis. *Environmental Technology*. DOI: 10.1080/09593330.2016.1256440
- Paper III** **Hey T.**, Bajraktari N., Vogel J., Hélix-Nielsen C., la Cour Jansen J. and Jönsson K. The effects of physicochemical pre-treatment of municipal wastewater on forward osmosis. *Environmental Technology*. DOI: 10.1080/09593330.2016.1246616
- Paper IV** **Hey T.**, Bajraktari N., Davidsson Å., Vogel J., Hélix-Nielsen C., la Cour Jansen J. and Jönsson K. Evaluation of direct membrane filtration and direct forward osmosis as concepts for compact and energy-positive municipal wastewater treatment. (*Submitted*)

My contributions to the papers

- Paper I** I designed and supervised the construction and installation of the pilot plant, including the operational parameters. I designed and conducted all experiments and evaluated and interpreted the data resulting from the work. I drafted and wrote the manuscript and received comments from my co-authors.
- Paper II** I designed and conducted all experiments, excluding the membrane fouling experiments. I evaluated and interpreted the data obtained from the experiments and wrote the manuscript, excluding the section regarding membrane fouling. I drafted and wrote the manuscript and received comments from my co-authors.
- Paper III** I designed and conducted all experiments. I evaluated and interpreted the data received from all experiments. I drafted and wrote the manuscript and received comments from my co-authors.
- Paper IV** I evaluated the selected process configurations. The biogas experiments were conducted and evaluated by my co-author. I drafted and wrote the manuscript with my co-authors and received comments.

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1 Introduction

Most of the wastewater treatment plants in Sweden were built between the 1960s and 1970s and use mechanical, biological and chemical treatment processes to obtain discharge that meets the national guidelines regarding carbon reduction (Swedish Environmental Protection Agency, 2016).

In 1991, the European Council Directive (Directive 91/271/EEC) was introduced, which harmonised the wastewater discharge guidelines of European countries for regulating wastewater discharge regarding carbon, nitrogen and phosphorus. The discharge demands depend on the size of the wastewater treatment plant expressed as population equivalent (PE) and the ecological sensitivity of the receiving water body.

Currently, more than 95% of the sewage in Sweden is treated by 411 municipal wastewater treatment plants. Of these wastewater treatment plants, more than half have sizes ranging from 2 000-10 000 PE (Swedish Environmental Protection Agency, 2016). Furthermore, the wastewater loads of these wastewater treatment plants have increased over the past decades and is continuing to increase due to population growth. Hence, some of these wastewater treatment plants need to extend the biological treatment step to accommodate current and future wastewater loads and discharge demands. Moreover, the present discharge limits are expected to become stricter in the near future, and 45 priority substances and 8 other pollutants are targeted (Directive 2015/495/EC), which could require additional treatment steps.

Until today, the biological treatment step is the most important and common process used at wastewater treatment plants for reduced discharge of carbon and nitrogen to comply with the wastewater directive (Directive 91/271/EEC). However, to accommodate for future wastewater loads and comply with stricter discharge demands, retrofitting and extension of the biological treatment step are inevitable. Hence, increased demands for electricity, resources, (e.g., in the form of chemicals), and area are expected where available area may already be a problem. Furthermore, there is a trend to consider wastewater as a viable resource that contains water, nutrients (Lutchmiah *et al.*, 2011) and energy in the form of organic carbon (Lateef *et al.*, 2013). On the other hand, municipal wastewater also contains harmful pollutants, e.g., pathogens, heavy metals and endocrine disruptive compounds.

Membrane separation can be used to enable increased resource recovery and to treat wastewater in a more compact way (van Loosdrecht and Brdjanovic, 2014). Furthermore, the direct use of membrane separation for raw municipal wastewater for wastewater reclamation purposes (e.g., irrigation purposes) is trending (Ravazzini *et al.*, 2008). However, few large-scale studies have been conducted for long-term operation focused on complying with wastewater discharge demands. Furthermore, few studies have included evaluations of specific electricity, energy and area demands.

1.1 Aims and objectives

The work presented in this thesis is focused on the use of different separation techniques, such as microsieving, microfiltration and forward osmosis, in different configurations to treat municipal wastewater without a biological treatment step. Furthermore, coagulation and flocculation were applied in different combinations with different separation techniques.

The main goal in this work was to provide proof of-concept results for treating municipal wastewater mechanically and physicochemically to fulfil Swedish wastewater discharge demands.

The specific objectives of this study are presented as follows:

- To test different treatment concepts applied to raw municipal wastewater.
- To operate microfiltration at low trans-membrane pressure and without backflushing.
- To test forward-osmosis with seawater as natural draw solutions for municipal wastewater treatment.
- To investigate the effects of coagulation and flocculation before microsieving and membrane filtration, i.e., microfiltration and forward osmosis.
- To identify and evaluate suitable treatment configuration(s) that could be integrated at full-scale wastewater treatment plants.

1.2 Outline of this thesis

Direct membrane filtration concept using different wastewater types, i.e., raw municipal wastewater and primary settler effluent for different reuse purposes is described in **Chapter 2**. Thereafter, in **Chapter 3**, the forward osmosis process is briefly described from the municipal wastewater treatment perspective.

Chapter 4 describes the methodologies used at the full-scale wastewater treatment plant Källby in Lund using real municipal wastewater to apply different mechanical and physicochemical treatment configurations.

The results of this work regarding the application of direct membrane filtration (**Paper I**) and forward osmosis (**Paper II, III, IV**) are presented and discussed in **Chapter 5**. Most of the work was focused on applying different treatment configurations, such as the mechanical and physicochemical pretreatment of raw municipal wastewater before microfiltration (**Paper I**) and/or forward osmosis (**Paper II, III, IV**). Furthermore, the water flux and retention of common wastewater parameters, such as carbon, phosphorus and nitrogen, are presented and compared with the present wastewater discharge demands. In addition, two treatment concepts are selected that comply with the Swedish wastewater discharge demands and are evaluated at the full scale regarding their specific electricity, energy and area demands. Furthermore, biogas production experiments were conducted with the separated sludge(s) and evaluated for specific electricity and energy production (**Paper IV**).

In **Chapter 6**, conclusions are drawn regarding the feasibility of the tested configurations for municipal wastewater treatment based on the results and discussions. **Chapter 7** provides some suggestions for future work, and further implementation of the proposed methods for future wastewater treatment are provided at the end of the thesis.

2 Direct Membrane Filtration

2.1 Background

Since the introduction of a membrane with high capacity and good separation characteristics by Loeb & Sourirajan (1959), membrane separation has taken a step toward commercialised separation-processes. The continuous development of membranes has led to different separation processes, i.e., microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse-osmosis (RO), which are hydraulic pressure-driven membrane processes. The aforementioned membranes have mainly been applied in industrial applications, e.g., microfiltration for cell-harvesting and ultrafiltration for the recycling of dyes and latex in the waste process stream. The application of membranes for municipal wastewater treatment remains scarce.

However, in the late 1970s, Sachs *et al.* (1976) and Higgins (1978) reported that treated wastewater effluents have potential for reuse as irrigation due to increasing global water shortages. Nevertheless, concerns have risen regarding the contamination of groundwater with heavy metals when wastewater effluent is used and spread (Higgins, 1978). Bhattacharyya *et al.* (1978) and Christensen and Plauman (1981) investigated the feasibility of using ultrafiltration for synthetic and real shower wastewater, synthetic laundry wastewater, industrial wastewater and secondary settler effluent for water reuse. These studies showed high (>90%) organic carbon and particle retention. Moreover, Christensen and Plauman (1981) reported a specific energy requirement of $4.6 \text{ kWh} \cdot \text{m}^{-3}$ for ultrafiltration.

Eventually, Kolega *et al.* (1991) used microfiltration for primary and secondary treated effluents, which improved the effluent quality, and for disinfection. Microfiltration of the primary effluent resulted in reductions of 61-89% for biochemical oxygen demand (BOD), 100% for suspended solids (SS), and 42-75% for total phosphorus (TP), respectively. In addition, harmful pathogens, e.g., total and faecal coliform contents, and faecal streptococci organisms were not found in the microfiltration permeate. Moreover, in the same study, up to 50% of the heavy metals Cu, Pb, Zn, Cr, and Sn were retained.

2.2 Sewer mining concept

In 1996, Butler and McCormick (1996) reported on the ACTEW water mining project in Australia, which used raw sewage from a main sewer to delay the construction of new dams for potable water by re-using municipal wastewater. The raw sewage underwent primary treatment, e.g., screening, chemically assisted primary treatment and biological treatment, before microfiltration. The microfiltration permeate was intended to be used for irrigation and at recreational areas. In the same study, the descriptions of ‘main interception sewage treatment’ and ‘sewer mining’ were coined, in which the latter one is internationally and widely accepted (Marks, 2006; Brown *et al.*, 2010; Lutchmiah *et al.*, 2011; Xie *et al.*, 2013; González-Viar *et al.*, 2016).

An additional wastewater re-use concept was presented by Abdessemed *et al.* (1999), who treated raw wastewater by using screening and filtering through a deep bed filtration system before ultrafiltration. The ultrafiltration permeate was intended for drinking water but did not comply the World Health Organisation’s standards for drinking water. Nevertheless, the ultrafiltration permeate was recommended for irrigation purposes because ammonium nitrogen was present and it was free of bacteria, e.g., free of total coliforms.

2.3 Direct membrane filtration

In the Netherlands, a research program was conducted with the goal of developing new and more sustainable municipal wastewater treatment methods based only on mechanical or physicochemical treatment processes. The initial aim of this research was based on the fact that part of the COD fraction in raw municipal wastewater consists of particulate organic matter, i.e., the particulate COD fraction, which can be removed by membrane-filtration and utilised for biogas production (van Nieuwenhuijzen *et al.*, 2000). Thus, the organic load to the biological treatment decreases, which allows for the design of a more compact biological treatment stage (van Nieuwenhuijzen *et al.*, 2000; 2001; 2002). Hence, ultrafiltration was applied to raw wastewater from four different municipal wastewater treatment plants and was described as direct ultrafiltration. Ultrafiltration with raw wastewater resulted in retentions of 50% for BOD, 65% for COD, 30% for TP, and 20% for total nitrogen (TN), respectively. Thus, the ultrafiltration permeate could be applied for irrigation purposes to supply viable nutrients for different types of crops. Ravazzini (2008) studied direct application of ultrafiltration for raw wastewater after screening and for primary treated wastewater for irrigation purpose (Ravazzini *et al.*, 2005).

This treatment step was evaluated as a potential pretreatment step prior to reverse osmosis (Rulkens *et al.*, 2005). Their study showed that high-quality water, i.e., water enriched with valuable nutrients and organics, can be produced in a single treatment step. Hence, the produced ultrafiltration permeates from both types of treated feed materials, i.e., raw wastewater and primary settler effluent, were particle free and showed COD retentions of 63% and 58%, respectively. During ultrafiltration, 10% and 20% of the TN and TP were retained. van Nieuwenhuijzen (2002) and Ravazzini *et al.* (2008) both described the direct application of membrane filtration for raw wastewater, which was referred to as **Direct Membrane Filtration (DMF)**. The term DMF is used throughout this thesis. A schematic showing the direct membrane filtration concept applied for raw municipal wastewater is depicted in Figure 1.

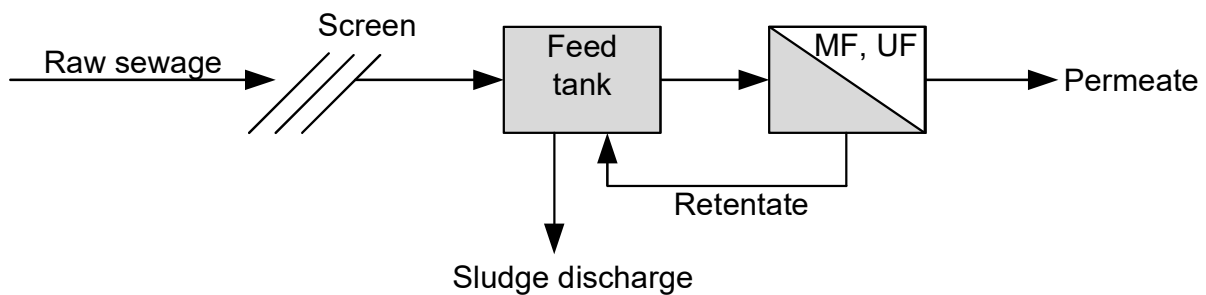


Figure 1.

Schematic process layout of the direct membrane filtration (DMF) of raw sewage (adapted from Ravazzini *et al.* 2011, with permission from IWA Publishing, London).

Furthermore, Lateef *et al.* (2013) investigated the DMF concept for carbon and nutrient utilisation and stated that 75% of organic matter can be recovered and used for energy purposes. Moreover, the DMF concept was evaluated as an attractive option for compact wastewater treatment that enables nitrogen and phosphorus recovery and energy production. In addition, DMF could be installed and performed within the space of currently working systems.

2.4 Direct Membrane Separation

In the study conducted by Ahn and Song (1999), microfiltration of septic tank effluent containing raw domestic wastewater from apartments and dormitories was conducted for water reuse, e.g., toilet flushing, sprinkling irrigation and landscaping. The microfiltration experiment was conducted continuously for 120 days and the results showed that >99% of the suspended solids (SS) were retained.

Approximately 93% of the chemical oxygen demand (COD) and BOD were retained, which was partially attributed to biodegradation in the septic tank. Nevertheless, the microfiltration-permeate qualities met the Korean standards for water reuse. In addition, Ahn *et al.* (2001) used microfiltration of low-strength wastewater for water reuse and described the principle as **Direct Membrane Separation (DMS)**. Microfiltration retained more COD, BOD and SS than total nitrogen (only minor amount of nitrogen were retained). Beside the DMS-concept, Ahn *et al.* (1998; 2000) also used microfiltration and ultrafiltration for greywater from a resort, which included hotel buildings and a shopping-mall, and achieved permeates with qualities similar to low-strength wastewater (Ahn *et al.*, 2001).

In addition to DMF (Ravazzini *et al.*, 2005) and DMS (Ahn *et al.*, 2001), Jin *et al.* (2015) introduced an additional description for the application of microfiltration for raw sewage: **Direct Sewage Microfiltration (DSM)**.

2.5. Addition of chemicals before membrane filtration

To improve COD retention, the following chemicals (e.g., coagulants and flocculants) were added before microfiltration: Ironchloride (FeCl_3) (Abdessemed and Nezzal, 2002; Ravazzini, 2011), polyaluminiumchloride (PACl) (Ravazzini, 2011; Delgado Diaz *et al.*, 2012), cationic flocculants (Ravazzini, 2011) and a mixture of PACl and cationic flocculant (Ravazzini, 2011). Furthermore, Jin *et al.* (2015) added PACl directly into a membrane reactor and described the process as **Hybrid Coagulation Microfiltration (HCM)**. The studies showed that coagulation significantly improved COD reduction by up to 95% (Abdessemed and Nezzal, 2002; Delgado Diaz *et al.*, 2012; Jin *et al.*, 2015). Furthermore, Abdessemed and Nezzal (2002) reported that the permeate flux significantly improved compared to chemical untreated raw municipal wastewater. Ravazzini *et al.* (2011) stated that coagulants have few effects on total organic carbon (TOC) removal but substantially increase the filterability of the wastewater.

Table 1 provides an overview of previously conducted studies regarding the application of microfiltration and ultrafiltration for different types of wastewater feed, i.e., raw municipal wastewater (Raw MWW), primary settler effluent (Primary Eff.), greywater and septic tank effluent (Septic tank). However, secondary effluent (Christensen and Plauman, 1981; Kolega *et al.*, 1991; Tchobanoglous *et al.*, 1998; Delgado Diaz *et al.*, 2012) and tertiary effluents (Tchobanoglous *et al.*, 1998) were also used in some of the studies but are not within the scope of this thesis.

Table 1.

Overview of the conducted studies performing direct membrane filtration on different wastewater types, i.e. raw municipal wastewater (Raw MWW), primary settler effluent (Primary Eff.), greywater (Greywater) and septic tank effluent (Septic tank).

| Feed | Membrane | | Duration | Reference |
|--------------|----------|----------------------|-----------|---|
| | Type | Area | | |
| Raw MWW | UF | (-) | 400 min | Abdessemed <i>et al.</i> , 1999 |
| Raw MWW | UF | 0.17 m ² | 7 hours | van Nieuwenhuijzen <i>et al.</i> , 2000; 2002 |
| Raw MWW | MF | 4 m ² | 120 days | Ahn <i>et al.</i> , 2001 |
| Raw MWW | UF | 0.073 m ² | 120 min | Ravazzini <i>et al.</i> , 2005 |
| Raw MWW | MF | 0.13 m ² | >200 min | Lateef <i>et al.</i> , 2013 |
| Raw MWW | MF | 0.33 m ² | 100 hours | Jin <i>et al.</i> , 2015 |
| Raw MWW | MF | 1.025 m ² | > 6 days | Paper I |
| Primary Eff. | MF | (-) | 6 months | Kolega <i>et al.</i> , 1991 |
| Primary Eff. | MF | 34.6 m ² | >1 year | Juby <i>et al.</i> , 2000; 2001; 2013 |
| Primary Eff. | UF | (-) | 200 min | Abdessemed and Nezzal, 2002 |
| Primary Eff. | MF | 34.4 m ² | >1 year | Sethi and Juby, 2002 |
| Primary Eff. | UF | 0.073 m ² | 120 min | Ravazzini <i>et al.</i> , 2005 |
| Primary Eff. | UF | 0.093 m ² | 100 min | Delgado Diaz <i>et al.</i> , 2012 |
| Greywater | MF, UF | 0.08 m ² | 12 hours | Ahn <i>et al.</i> , 1998 |
| Greywater | MF, UF | 0.3 m ² | 77 days | Ahn and Song, 2000 |
| Greywater | UF, NF | 0.014 m ² | (-) | Ramon <i>et al.</i> , 2004 |
| Septic tank | MF | 20 m ² | 120 days | Ahn and Song, 1999 |

- : The value was not reported in the study.

The listed studies in Table 1 were mainly conducted at bench scale and over short durations. However, in the studies of Juby *et al.* (2000; 2001; 2013) and Sethi and Juby (2002), vast amounts of raw municipal wastewater were treated at the pilot scale. Nevertheless, no description of transition to full-scale application has been found.

2.6 Integrated Membrane Anaerobic Stabilization

Beside the studies of van Nieuwenhuijzen *et al.* (2000; 2002), Ravazzini *et al.* (2005; 2008; 2011), Ahn *et al.* (1998; 1999; 2000; 2001) and Jin *et al.* (2015), Juby *et al.* (2000) conducted pilot-scale experiments at the Orange County Sanitation District in Southern California, USA. This study addressed future challenges such as shortages of land, the high costs associated with additional secondary wastewater treatment facilities, and the disposal of biosolids.

In comparison with DMF (Ravazzini *et al.*, 2005) and DMS (Ahn *et al.*, 2001), microfiltration was applied on primary settler effluent, and the produced microfiltration permeate was subsequently treated with reverse osmosis for (soluble) carbon rejection (Juby *et al.*, 2000; 2001; 2013). The generated primary sludge and the microfiltration retentate were anaerobically digested in a conventional anaerobic digester for biogas production. Furthermore, the generated reverse-osmosis retentate, containing high amounts of (soluble) carbon, was utilised in a high rate upflow anaerobic sludge blanket reactor (UASBR) for biogas utilisation. This process concept was introduced as the **Integrated Membrane Anaerobic Stabilization (IMANS™)** process and is shown in Figure 2 (Juby *et al.*, 2000; 2001; 2003; 2013; Sethi and Juby, 2002). The produced reverse-osmosis permeate contained low BOD, COD, TOC and ammonium concentrations, and evaluating the concept was found to offer significant benefits for both wastewater treatment and operational costs (Juby *et al.*, 2001).

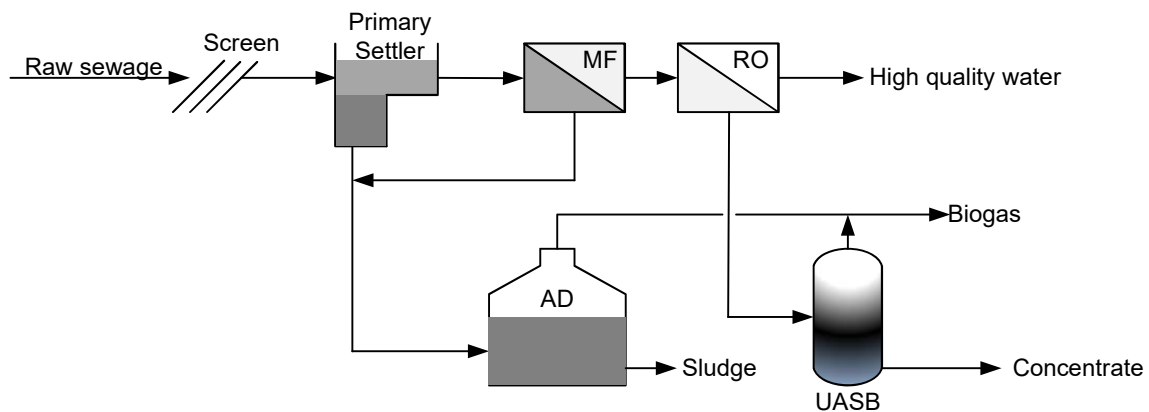


Figure 2.

Simplified schematic drawing of the IMANS™ process (adapted from Juby *et al.*, 2000, reprinted with permission from Proceeding of WEFTEC® 2000, the 73rd Annual Water Environment Federation Technical Exhibition and Conference, Anaheim, California, October 14_18, 2000. Copyright © 2000 Water Environment Federation, Alexandria, Virginia).

Following the aforementioned studies listed in Table 1 that used different membrane filtration process types (e.g., microfiltration, ultrafiltration and nanofiltration) for different wastewater types (i.e., raw municipal wastewater (Raw MWW), primary settler effluent (Primary Eff.), greywater and septic tank effluent), Table 2 provides an overview of the reported BOD, COD, TN and TP retention according to the employed membrane process. Furthermore, the studies that used coagulants before microfiltration (e.g., FeCl₃ or PACl) are marked with an asterisk.

Table 2.

Overview of the reported retentions (%) of BOD, COD, TOC, TN and TP of microfiltration, ultrafiltration and/or nanofiltration applied on raw municipal wastewater, primary settler effluent, greywater and septic effluent.

| Feed | BOD | CODt | TOC | TN | TP | Reference |
|--------------------------|--------|---------|--------|--------|--------|---|
| Raw municipal wastewater | 95% | 98% | 71% | 36% | | Ahn <i>et al.</i> , 2001 |
| | | 70-80% | | | | Lateef <i>et al.</i> , 2013 |
| | | 95%* | | | | Jin <i>et al.</i> , 2015* |
| Primary settler effluent | 61-89% | | | | 42-75% | Kolega <i>et al.</i> , 1991 |
| | 44% | 49% | | | | Juby <i>et al.</i> , 2000 |
| | 48% | 50% | | | | Juby <i>et al.</i> , 2001 |
| | 47% | 49% | | | | Sethi and Juby, 2002 |
| | 57% | 65% | 8% | 0% | | Juby <i>et al.</i> , 2013 |
| Greywater | | 54-61% | 87-89% | | | Ahn <i>et al.</i> , 1998 |
| | | 80% | | | | Ahn and Song, 2000 |
| Septic tank effluent | | 93% | 93% | 66% | | Ahn and Song, 1999 |
| Raw municipal wastewater | 94-99% | 97% | | | | Abdessemed <i>et al.</i> , 1999 |
| | | 69% | | 17% | 36% | van Nieuwenhuijzen <i>et al.</i> , 2000 |
| | 47-52% | 51-65% | | 13-22% | 18-53% | van Nieuwenhuijzen, 2002 |
| Primary effluent | | 37% | | 9% | 20% | Ravazzini <i>et al.</i> , 2005 |
| | | 88%* | | | | Abdessemed and Nezzal, 2002* |
| | | 42% | | 0% | 20% | Ravazzini <i>et al.</i> , 2005 |
| Greywater | | 81-95%* | | | | Delgado Diaz <i>et al.</i> , 2012* |
| | | 59-60% | 68-86% | | | Ahn <i>et al.</i> , 1998 |
| | | 80-84% | | | | Ahn and Song, 2000 |
| NF Greywater | | 45-49% | | | | Ramon <i>et al.</i> , 2004 |
| | | 93% | 83% | | | Ramon <i>et al.</i> , 2004 |

*Chemicals were used as coagulants, e.g., FeCl₃, PACl.

3 Forward osmosis

3.1 Background

In comparison to the membrane processes driven by hydraulic pressure (ΔP), e.g., microfiltration or reverse osmosis (RO), the forward osmosis (FO) membrane processes are driven by an osmotic pressure gradient ($\Delta\pi$) across a dense semipermeable membrane (see Figure 3). This osmotic pressure gradient is generated when a feed solution (e.g., municipal wastewater) with a low osmotic pressure and a draw solution (e.g., seawater) with a higher osmotic pressure are used. The ‘forward osmosis’ phenomenon occurs as water begins to migrate from the feed solution to the draw solution to reach osmotic equilibrium. While water migrates from the feed solution to the draw solution, the osmotic pressure increases in the feed solution because the (salt) concentration increases and decreases in the draw solution due to (water) dilution. Since osmotic pressure is the driving force and no hydraulic pressure is applied, forward osmosis is potentially a low-energy technology solution (Lutchmiah *et al.*, 2014).

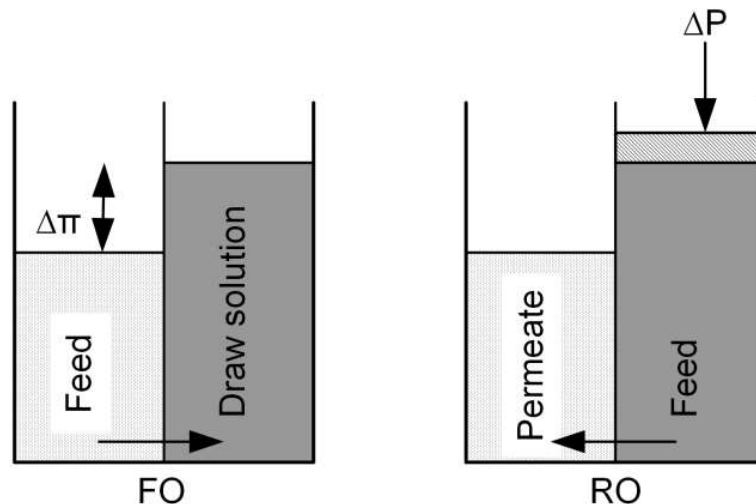


Figure 3.

Principles of forward osmosis (FO) and reverse osmosis (RO) processes. The black arrows in both figures (left and right) represent the transport of water through a semipermeable membrane. In forward osmosis (left), water transport is driven by the osmotic pressure gradient ($\Delta\pi$), and in reverse osmosis (right), water transport is driven by the exerting pressure that drives the water from the concentrated feed to the diluted permeate (adapted from Wicaksana *et al.*, 2012, with permission from Springer Science+Business Media).

Historically, the membrane forward osmosis process was first published in the middle of the 1970s. Kravath and Davis (1975) proposed using a hypertonic glucose solution for seawater desalination to generate potable water, which could be applied to supply water in emergencies on lifeboats. Kessler and Moody (1976) provided theoretical and practical demonstrations of the use of a forward osmosis extractor to transfer fresh water from seawater into a solution containing low-molecular-weight nutrients. In the same year, Moody and Kessler (1976) suggested using forward osmosis for agricultural water reclamation, e.g., the extraction of brackish water for use as fertiliser and irrigation (fertigation). However, the forward osmosis membrane process was overlooked for nearly 30 years and rediscovered in 2004 by McCutcheon and Elimelech (2004), who suggested that forward osmosis could be used in applications such as wastewater treatment (van der Bruggen and Luis, 2015). Since then, the number of forward osmosis publications has increased rapidly, but only a few of these studies (~7%) have considered complex water (Lutchmiah *et al.*, 2014), e.g., wastewater, centrates from anaerobic digesters, drilling wastewater, landfill leachate, and wastewaters from food and beverage industries (Coday *et al.*, 2014).

Nevertheless, the number of publications considering municipal wastewater treatment has steadily increased because forward osmosis has the following potential advantages over current membrane technologies: (1) high rejection of particles, pathogens and emerging substances, (2) more energy-efficient than reverse osmosis due to the lack of high hydraulic pressure, (3) no need for extensive pretreatment for forward osmosis, (4) excellent operation in terms of durability and water quality for highly polluted waters, (5) flexibility and applicability due to scalability, (6) reduced fouling propensities and simple cleaning, and (7) convenient application for upconcentration purposes (Lutchmiah *et al.*, 2014). Despite the aforementioned advantages, full-scale applications using the forward osmosis process are not widely used; because the osmotic pressure gradient is the driving force, a sufficient draw solution must always be available. Therefore, many studies of alternative draw solutions, which have become the Holy Grail of forward osmosis, have been conducted (van der Bruggen and Luis, 2015).

3.2 Forward osmosis for municipal wastewater treatment

Based on the ‘sewer mining concept’, forward osmosis was used for water recovery and energy production (in the form of biogas) from real primary-settled wastewater (Lutchmiah *et al.*, 2011). This study showed that water can be recovered to a certain extent, but not completely, due to internal concentration polarization (ICP) and fouling.

Nevertheless, stable water fluxes $>4.3 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ were obtained and membrane cleaning was easily performed because the fouling layer was thin and loose. Lutchmiah *et al.* (2011) indicated that sewage is a resource for water, energy and nutrients and is not waste.

Xie *et al.* (2013a) applied forward osmosis in combination with membrane distillation (FO-MD) to raw sewage (after screening) for small-scale decentralised sewer mining. The study showed that water can be recovered by 80% and that high removals (91-98%) of trace organic contaminants can be achieved.

In the study of Zhang *et al.* (2014), forward osmosis was directly applied to raw sewage, which concentrated the COD by 308% within 22 hours. The concentrated COD could be utilised for low-energy wastewater treatment and could increase energy production.

Wang *et al.* (2016) used low-strength primary settled wastewater at the pilot scale for nutrient and energy recovery. Forward osmosis was conducted for 50 days and resulted in high solute rejections of 99.8% and 99.7% for COD and TP, respectively. However, lower solute rejections were achieved for ammonium (48.1%) and TN (67.8%). Wang *et al.* (2016) also reported that a critical and subcritical concentration factor of 5 should be used to achieve a cost-efficient treatment during long-term operation. This pilot-scale study demonstrated the promise of using forward osmosis for concentrating low-strength municipal wastewater but also showed the limitations of this method regarding nitrogen rejection.

Apart from water and nutrient recovery, the forward osmosis process has been applied for the following different applications considering wastewater treatment: rejection of trace organic contaminants (Hancock *et al.*, 2011; Linares *et al.*, 2011; Jin *et al.*, 2012; Xie *et al.*, 2012a & b; Alturki *et al.*, 2013; Xie *et al.*, 2013a & b; Liu *et al.*, 2015; Madsen *et al.*, 2015) and heavy metals (Valladares Linares *et al.*, 2013; Cui *et al.*, 2014), dewatering potentials and simultaneous nutrient recovery (Nguyen *et al.*, 2013). Furthermore, forward osmosis was also applied for membrane bioreactor systems either using activated sludge (Luo *et al.*, 2015) or in an anaerobic digester (Ding *et al.*, 2014).

Table 3 provides a list of studies that applied different types of wastewater and involved the use of forward-osmosis membranes for different purposes. The table also shows the used feed and draw solutions, forward osmosis-membrane types and membrane areas (cm^2). Studies applying effluents from secondary- (Cath *et al.*, 2010; Linares *et al.*, 2011) and tertiary settlers (Cath *et al.*, 2010) and industrial wastewater (Coday *et al.*, 2014) were not included because the focus was on raw municipal wastewater. However, since urine and centrate from anaerobic digesters (AD-centrate) contain high amounts of (soluble) nitrogen, nitrogen rejection was included based on the total nitrogen discharge demand.

Table 3.

Overview of conducted forward osmosis experiments with different wastewater types, i.e., synthetic wastewater, raw municipal wastewater, primary settler effluent, anaerobic-digester centrate, and urine.

| Feed | Draw | Membrane | | System | Purpose | Reference |
|-----------------------------|----------------------------|----------|-----------------------|--------|---------------------------|--------------------------------|
| | | Type | Area | | | |
| Synthetic wastewater | 100 g·L ⁻¹ NaCl | CTA | 139 cm ² | FO-RO | Water recovery | Cath <i>et al.</i> , 2005a & b |
| | 4% NaCl | CTA | 250 cm ² | FO | Water-recovery | Valladres <i>et al.</i> , 2013 |
| | 3.5 wt% NaCl | CTA, TFC | 38.5 cm ² | FO | Nutrient recovery | Xue <i>et al.</i> , 2015 |
| Raw municipal wastewater | 1.5 M NaCl | CTA | (-) | FO-MD | Water recovery | Xie <i>et al.</i> , 2013a |
| | 3.5 wt% NaCl | CTA | 64.6 cm ² | FO | Low-energy WWT | Zhang <i>et al.</i> , 2014a |
| | 1 and 2 M NaCl | CTA | 40 cm ² | FO | Removal of TrOCs | Liu <i>et al.</i> , 2015 |
| | 0.5-3 M NaCl, DI | CTA, TFC | 20.0 cm ² | FO | Cleaning protocol | Wang <i>et al.</i> , 2015 |
| | 2 M NaCl, Seaw. | TFC | 140 cm ² | FO | Wastewater treatment | Paper II, III, IV |
| Primary settler effluent | 0.5-4.5 M NaCl | CTA | 124 cm ² | FO | Water recovery | Lutchmiah <i>et al.</i> 2011 |
| | Mixture (60 bar) | CTA | 125 cm ² | FO | WW-up concentration | Ansari <i>et al.</i> , 2016a |
| | 0.5 M NaCl | CTA | 0.3 m ² | FO | Water & nutrient recovery | Wang <i>et al.</i> , 2016 |
| Anaerobic-digester centrate | 70 g·L ⁻¹ NaCl | CTA | 139 cm ² | FO-RO | Nutrient recovery | Holloway <i>et al.</i> , 2007 |
| | 1.5 M MgCl ₂ | CTA | (-) | FO-MD | Water & nutrient recovery | Xie <i>et al.</i> , 2014 |
| | Seawater | CTA | 123.5 cm ² | FO | Nutrient recovery | Ansari <i>et al.</i> , 2016b |
| Urine | 0.5-2 M NaCl | CTA | 42 cm ² | FO | Nutrient recovery | Zhang <i>et al.</i> , 2014b |

(-): The value was not reported in the study. CTA: Cellulose-tri-acetate, DI: Deionized water, MD: Membrane distillation, RO: Reverse osmosis, TFC: Thin film composite, TrOCs: Trace organic contaminants, wt%: Weight percentage, WW: Wastewater, WWT: Wastewater treatment.

Considering the identified studies that applied forward osmosis to different types of wastewater, i.e., synthetic wastewater, raw wastewater, primary settler effluent, anaerobic digested centrate and urine, Table 4 provides an overview of the reported solute rejections of COD, TOC, TN and TP.

Table 4.

Overview of the solute rejections (%) for COD, TOC, TN and TP of the different wastewater types, e.g., synthetic wastewater, raw wastewater, primary settler effluent, anaerobic digester centrate and urine, on forward osmosis.

| Feed | COD | TOC | TN | TP | Reference |
|-----------------------------|------|------|--------|--------|---------------------------------|
| Synthetic wastewater | 99% | | 56-59% | 99% | Valladares <i>et al.</i> , 2013 |
| | | | 0% | >90% | Xue <i>et al.</i> , 2015 |
| Raw municipal wastewater | | >95% | >95% | | Xie <i>et al.</i> , 2013 |
| | 72% | | | | Zhang <i>et al.</i> 2014a |
| Primary settler effluent | >99% | | | | Ansari <i>et al.</i> , 2016a |
| | >99% | | 68% | >99% | Wang <i>et al.</i> , 2016 |
| Anaerobic digester centrate | | | 85-97% | >99% | Holloway <i>et al.</i> , 2007 |
| | | | >90% | >97% | Xie <i>et al.</i> , 2014 |
| | | | | >95% | Ansari <i>et al.</i> , 2016b |
| Urine | | | 31-91% | 97-99% | Zhang <i>et al.</i> , 2014b |

3.3 Biomimetic membranes

Aquaporins are responsible for maintaining the osmotic gradient in the cell and for the transport of water in all living organisms (King *et al.*, 2004; Beitz and Agre, 2009). Furthermore, aquaporins are a group of transmembrane proteins that are highly selective for molecules and have been shown to effectively transport up to 10^9 molecules per second through the aquaporin water channel (Jensen *et al.*, 2006).

In recent years development of forward osmosis membranes has moved in the direction of biomimetic membranes that incorporate functional biomolecules in a synthetic membrane matrix. Zhao *et al.* (2012) proposed a new type of biomimetic membrane with functional aquaporin water channels encapsulated in vesicles and embedded in the active layer of a thin film composite membrane.

Biomimetic forward osmosis membranes with integrated aquaporin channels are a new type of membrane technology that has a high potential for development and application in many different fields. Since the aquaporin membrane (Aquaporin Inside™, Aquaporin A/S, Copenhagen, Denmark) is a new membrane, few studies of its use for different processes have been conducted, e.g., the membrane has been used for wastewaters from the textile industry (Petrinić and Hélix-Nielsen, 2014), in a biorefinery process (Kalafatakis *et al.*, 2015), for the removal of trace organics from water (Madsen *et al.*, 2015), for reduction of dimethylsilanediol (DMSD) in

the International Space Station's water recycling system (Tommerup *et al.*, 2016), and for crystallisation of Na_2CO_3 for CO_2 capture (Ye *et al.*, 2016).

However, no studies of using the Aquaporin Inside™ membrane (AIM) for municipal wastewater to study its performance in terms of water flux and solute rejection have been conducted. Therefore, in this thesis, the AIM is tested with integrated aquaporin channels for wastewater treatment applications.

4 Methodologies

4.1 Pilot-scale plant at the Källby WWTP

A pilot-scale plant (see Figure 4) was built at the wastewater treatment plant Källby in Lund, Sweden, using real municipal wastewater to attain practical experiences. Municipal wastewater from the sand trap outlet was pumped at a constant flow rate to the pilot-scale plant and used as feed for all conducted experiments (**Paper I-IV**). The experiments conducted at the pilot scale were conducted using coagulation, flocculation, microsieving (MS) and microfiltration (MF) (**Paper I**). Forward osmosis (FO) was conducted separately at the bench scale (**Paper II-IV**) because the AIM forward-osmosis membrane was in the pre-commercial stage, as stated by Coday *et al.* (2014) and Perry *et al.* (2015). The pilot-scale plant was constructed so that different process configurations could be operated, e.g., before microfiltration or forward osmosis.

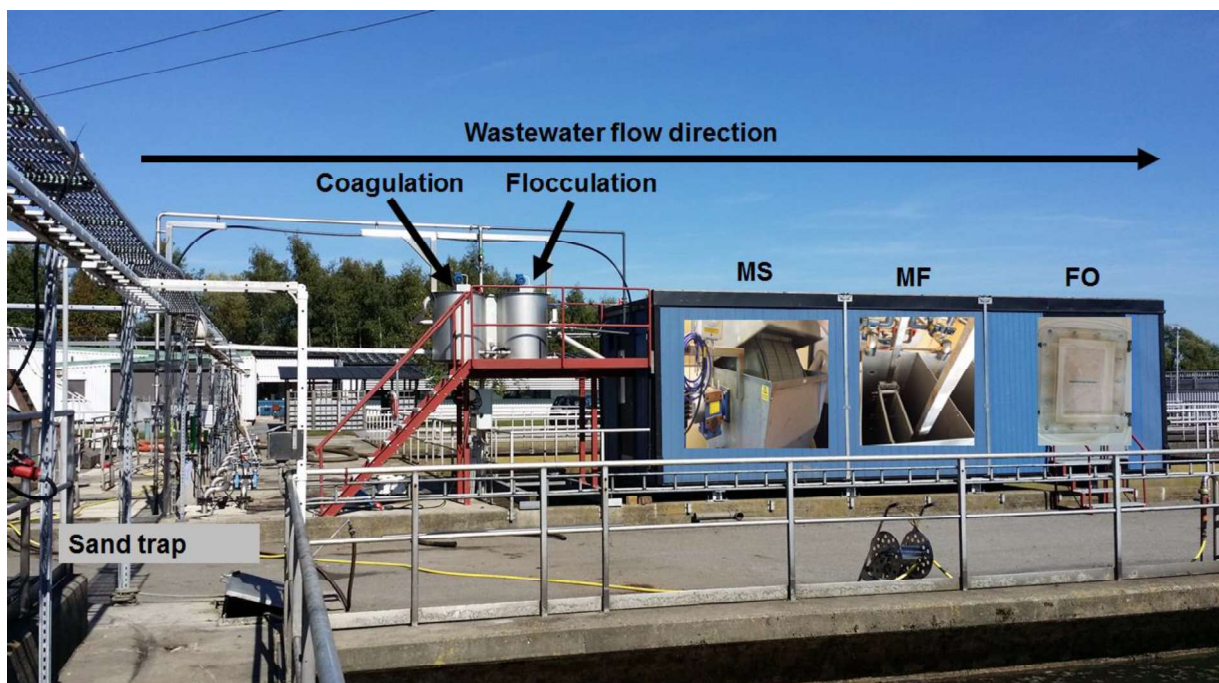


Figure 4. Pilot-scale plant at the Källby wastewater treatment plant in Lund, Sweden. The feed to the pilot-scale plant is pumped from the sand trap outlet and can undergo coagulation, flocculation, microsieving (MS) and microfiltration (MF) in different combinations. Forward osmosis (FO) was operated in bench scale inside the pilot-scale plant container.

4.1.1 Coagulation and flocculation

Coagulation chemicals, e.g., FeCl_3 or PACl , are commonly used for phosphate precipitation at municipal wastewater treatment plants in pre-, simultaneous-, or post-precipitation processes. By adding a coagulate such as PACl to wastewater, Al^{3+} dissociates and promotes phosphorus precipitation by forming AlPO_4 . Furthermore, colloids can coagulate in the presence of the coagulant, leading to the formation of flocs. Together, the coagulant and anionic flocculant can increase the binding strength of the generated macrofloc's by linking the negatively charged polymer group to the positively charged sites in the flocs, making the floc more compact (Gillberg *et al.*, 2003). The addition of only cationic flocculants is also described as cationic polymer coagulation (Kvinnesland and Ødegaard, 2004). Such polymers create effects on macrofloc formation similar to other coagulants. However, cationic polymer coagulants affect phosphorus precipitation less due to the absence of metal salts (Väänänen *et al.*, 2016).

In this study, polyaluminium chloride (PACl) was used for coagulation and anionic or cationic polymers were used for flocculation (**Paper I, III, IV**). The chemicals were dosed flow-proportionally to the raw municipal wastewater to obtain final concentrations of $15 \text{ mg}\cdot\text{L}^{-1}$ (PACl), $3 \text{ mg}\cdot\text{L}^{-1}$ (anionic flocculant) and $4 \text{ mg}\cdot\text{L}^{-1}$ (cationic flocculant) before entering the appropriate mixing tank. Continuous stirring was conducted in the mixing tanks to ensure sufficient mixing with velocity gradients (G-values) of $\sim 100\text{-}200 \text{ s}^{-1}$ (coagulation) and $\sim 80\text{-}150 \text{ s}^{-1}$ (flocculation), respectively. The total hydraulic retention time for both coagulation and flocculation to ensure sufficient mixing was at least 6 min. The coagulation and flocculation mixing tanks of the pilot scale plant are depicted in Figure 5.

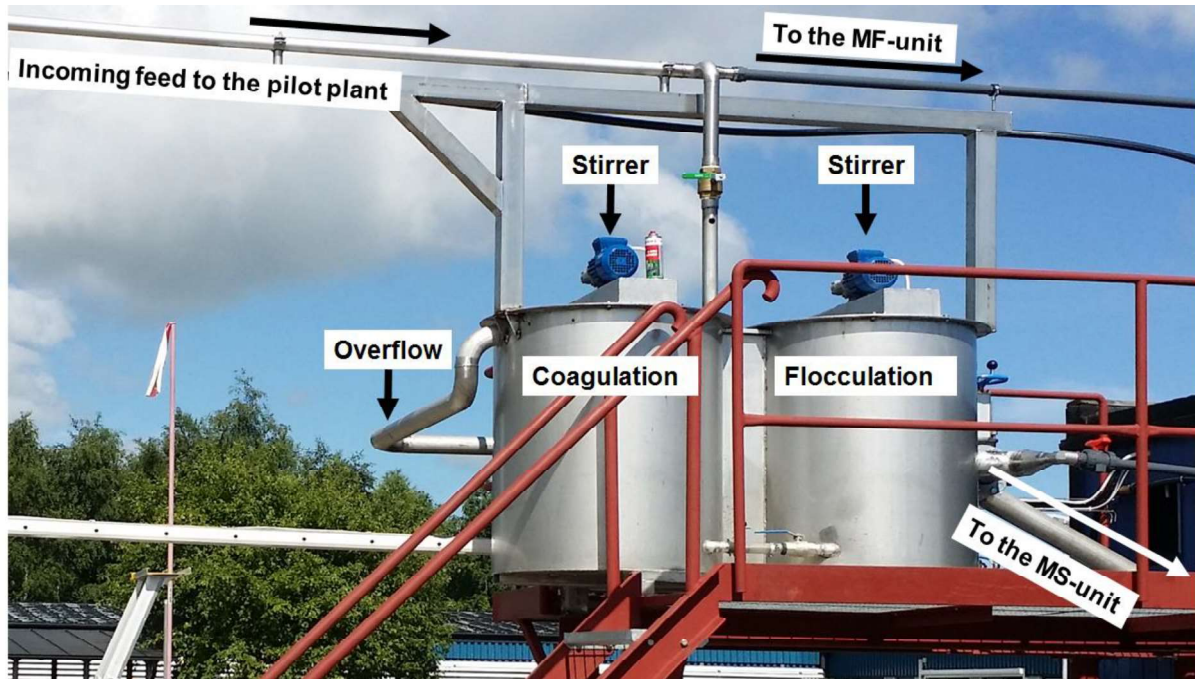


Figure 5. Coagulation and flocculation mixing tanks including stirrers outside the pilot plant.

4.1.2 Microsieving

As an alternative to primary clarifiers and to reduce the specific area demand for primary treatment, microsieving has been shown to reduce SS by 50% without the addition of any chemicals (Rusten and Ødegaard, 2006). In the study of Väänänen *et al.* (2016), wastewater from the sand trap outlet was directly passed through a drum rotating microsieve with a sieve pore size of 100 μm . As the sieve pore size (μm) decreased, the SS retention (%) increased as follows: 100 μm - 43%, 40 μm - 45% and 30 μm - 66%.

Among the conducted direct membrane filtration studies, only a few studies applied mechanical pretreatment method(s) before microfiltration or ultrafiltration. Nevertheless, Ahn *et al.* (1998; 1999; 2000; 2001) used a 500 μm sieve, Juby *et al.* (2000) used a 200 μm strainer, Sethi and Juby (2002) used a two stage pretreatment method with 1190 mm and 600 mm openings, and Juby (2013) applied a 350 μm strainer before microfiltration.

Abdessemed *et al.* (1999) applied sandfiltration, and Ravazzini *et al.* (2005) used a 560 μm sieve before ultrafiltration. However, the importance of the employed pretreatment method before membrane filtration, e.g., microfiltration or ultrafiltration, was not addressed.

In this work, (**Paper I-IV**), a drum rotating microsieve with a sieve pore size of 100 μm (see Figure 6) was used for mechanical pretreatment before microfiltration and forward osmosis to observe the operational performance in terms of permeate flux, operational stability and permeate quality.

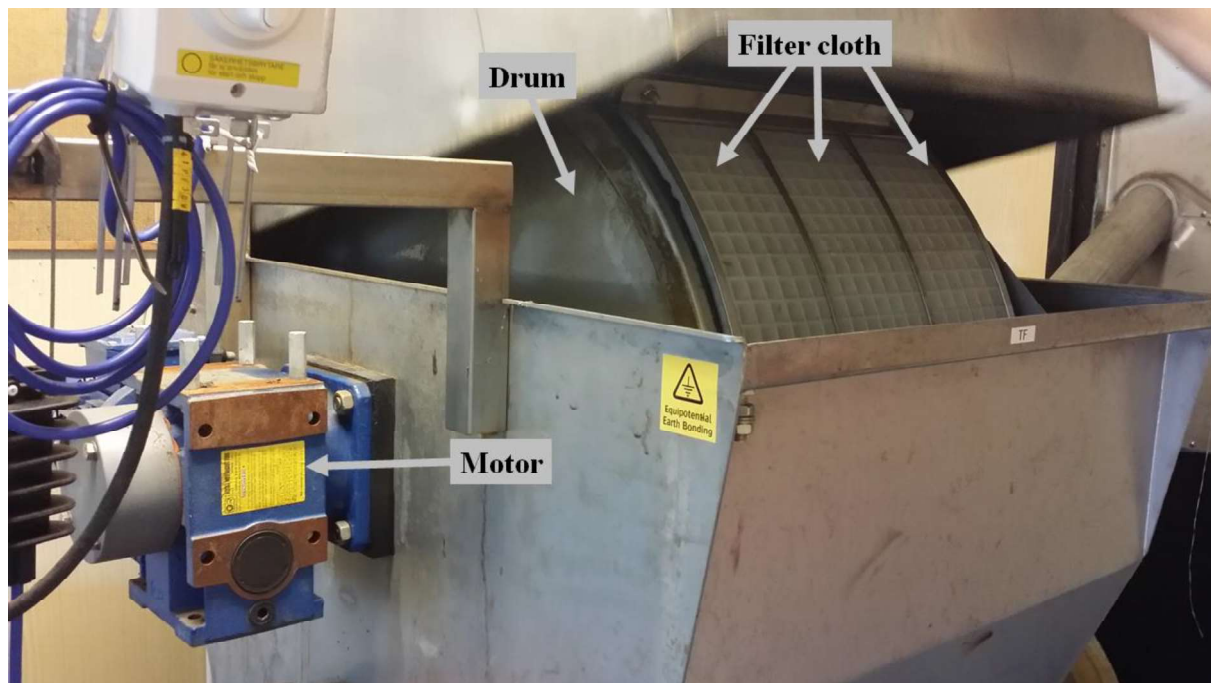


Figure 6. A rotating drum microsieve with a sieve pore size of 100 μm (Hydrotech HDF2005, Veolia Water Technologies AB, Sweden) that was operated inside the pilot plant.

4.1.3 Combination of coagulation, flocculation and microsieving

Combined coagulation/flocculation with microsieving for the removal of SS, COD, BOD and TP of raw municipal wastewater was studied by Ljunggren *et al.* (2007), Remy *et al.*, 2014 and Väänänen *et al.*, 2016. These studies showed average retentions of 86-95%, 70-95%, 50% and >95% for SS, COD, BOD and TP, respectively. However, Ljunggren *et al.* (2007) used FeCl_3 for coagulation, while Remy *et al.* (2014) and Väänänen *et al.* (2016) used PACl.

Furthermore, Ljunggren *et al.* (2007) and Väänänen *et al.* (2016) investigated the use of cationic coagulation and achieved SS reductions of 80-90%. In addition, Väänänen *et al.* (2016) achieved retentions of 70-90% for COD and 50-90% for TP. However, despite the high TP retention (50-90%) by the microsieve, the final TP concentration in the microsieve filtrate corresponded to 2-5 $\text{mg}\cdot\text{L}^{-1}$.

Nevertheless, because large amounts of carbon (SS, COD) and phosphorus (TP) can be removed by coagulation/flocculation in combination with microsieving, the produced microsieve filtrate was considered to have a positive effect on the

microfiltration process regarding permeate flux, permeate quality and, consequently, wastewater discharge demands. Hence, both combinations, i.e., coagulation/flocculation and cationic coagulation with microsieving, were selected to be tested at the pilot scale before microfiltration.

4.1.4 Microfiltration

The driving force for microfiltration is a hydraulic pressure gradient across the membrane and is also described as the trans-membrane pressure (TMP). In previous studies that employed microfiltration for direct membrane filtration, TMPs of 30-45 kPa (Lateef *et al.*, 2013), 40-70 kPa (Jin *et al.* 2015) and up to 120 kPa (Ahn *et al.*, 1998; 1999; 2000; 2001) have been reported. However, in most studies, the applied trans-membrane pressure increased during the experiments rather than remaining constant because a constant permeate flux was desired (Juby *et al.*, 2013; Jin *et al.* 2015).

In this study, microfiltration (**Paper I**) was conducted using a commercially available microfiltration membrane (MFP2) from Alfa Laval (Alfa Laval A/S, Nakskov, Denmark). Microfiltration was performed using a constant trans-membrane pressure of 3 kPa (0.03 bar) that was generated by the hydrostatic pressure of the feed above the microfiltration membranes and controlled by an online pressure sensor installed at the bottom of the tank. Five flat sheet microfiltration membranes (0.2 μm nominal pore size) were used to give a total active membrane area of 1.025 m^2 . Air scouring (e.g., for cake layer removal (Vigneswaran *et al.*, 2011)), with a standard air demand (SAD_m) of 0.69 $\text{Nm}^3 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ was supplied by an aerator during microfiltration operation and membrane cleaning. Permeate production was interrupted every 10 min to perform 2 min of membrane relaxation in the same manner as reported by Ahn and Song (1999) and van Nieuwenhuijzen (2002). Furthermore, no backflushing was applied during the microfiltration, as described in the study of Abdessemed *et al.* (1999). The backflush omittance was chosen to observe the permeate flux at a constant trans-membrane pressure. Each microfiltration experiment was conducted at the pilot scale for more than 6 days.

The produced permeate flux was measured constantly with a flow meter and recorded every 6th second by a central computerised system and corrected to a standard temperature of 15°C, as suggested by van Nieuwenhuijzen (2002). Furthermore, the bottom valve of the membrane tank was controlled over time to ensure a hydraulic retention time of less than 6 min inside the membrane tank to minimise biological activity (**Paper I**). The retention (%) of each wastewater compound by the microsieve or microfiltration device was calculated as follows:

$$\text{Retention (\%)} = \frac{(c_{in} - c_{out})}{c_{in}} \cdot 100 \text{ (Equation 1)}$$

where c_{in} ($\text{mg}\cdot\text{L}^{-1}$) represents the concentration (e.g., CODt) in the raw municipal wastewater and c_{out} ($\text{mg}\cdot\text{L}^{-1}$) represents the concentration in the microsieve filtrate or the microfiltration permeate.

Membrane cleaning was performed using citric acid and hydrogen peroxide to remove inorganic and organic matter, respectively. Figure 7 shows the microfiltration unit used for microfiltration permeate production in **Paper I-IV**.

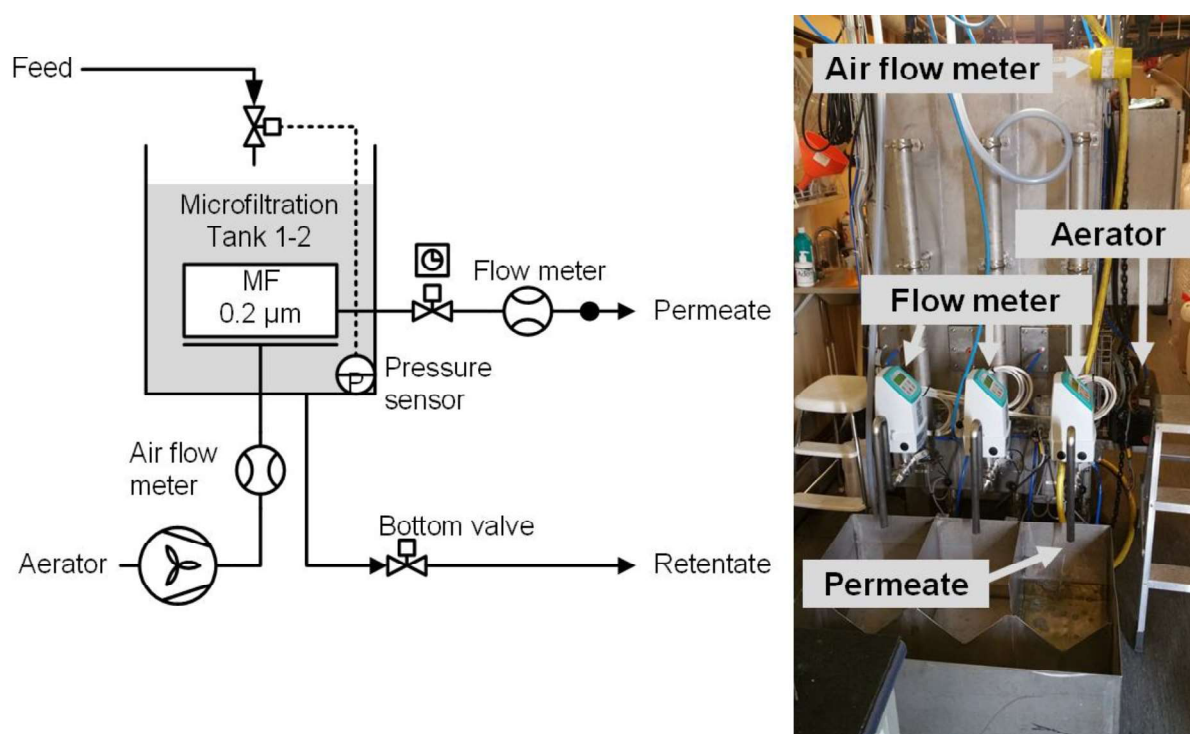


Figure 7. Schematic layout of one microfiltration unit (left) and the microfiltration pilot plant inside the pilot plant (right). Three membrane tanks were available for operation, but only two tanks were used, and one tank was used as a backup tank. All tanks can be operated independently and in parallel.

4.1.5 Combination of coagulation and membrane filtration

Jin *et al.* (2015) directly added PACl to a microfiltration tank containing raw wastewater and achieved 75% COD retention. This method was described as hybrid coagulation microfiltration (HCM).

Furthermore, coagulation of primary effluent with either FeCl_3 (Abdessemed and Nezzal, 2002) or AlCl_3 (Delgado Diaz *et al.*, 2012) was applied before ultrafiltration, respectively. The studies showed that COD was reduced by 86% (FeCl_3) and 81-95% (AlCl_3). Moreover, Abdessemed and Nezzal (2002) reported that the permeate flux was significantly improved by using a coagulant.

AlCl_3 was chosen for coagulation in the pilot-scale study because AlCl_3 was used in the study conducted by Väänänen *et al.* (2016) and achieved high carbon retentions when microsieving was used for raw municipal wastewater at the Källby wastewater treatment plant. Furthermore, Delgado Diaz *et al.* (2012) reported higher COD retentions with PACl than with FeCl_3 (86%, Abdessemed and Nezzal, 2002).

4.1.6 Forward osmosis membrane

Forward osmosis experiments (**Paper II, III**) were conducted using two different flat-sheet, thin-film-composite (TFC), forward-osmosis membranes. One membrane was obtained from Hydration Technologies Inc. (HTI, Albany, OR, USA), and the other membrane was obtained from Aquaporin A/S (Copenhagen, Denmark). Compared with the HTI membrane, the Aquaporin Inside™ (AIM) membrane is a relatively new membrane, and few applications have been tested. The AIM membrane is a modified TFC membrane with an active layer (AL) containing aquaporin proteins reconstituted in spherical vesicles and encapsulated by a polyamide thin film supported by polyethersulfone (Yip *et al.*, 2010; Zhao *et al.*, 2012). The AIM membrane can also be described as a biomimetic membrane.

Forward osmosis experiments were performed in bench-scale studies using a rectangular membrane module with two identical compartments measuring 175 mm (length) by 80 mm (width) and 1.3 mm (height) (see Figure 8). The active membrane area was 140 cm², and the active layer (AL) faced the feed solution (FS), which was described as the AL-FS mode. Counter-current circulation of the feed and draw solutions was applied using variable micro gear pumps to generate cross-flow velocity according to the procedure of Aquaporin A/S, Denmark.

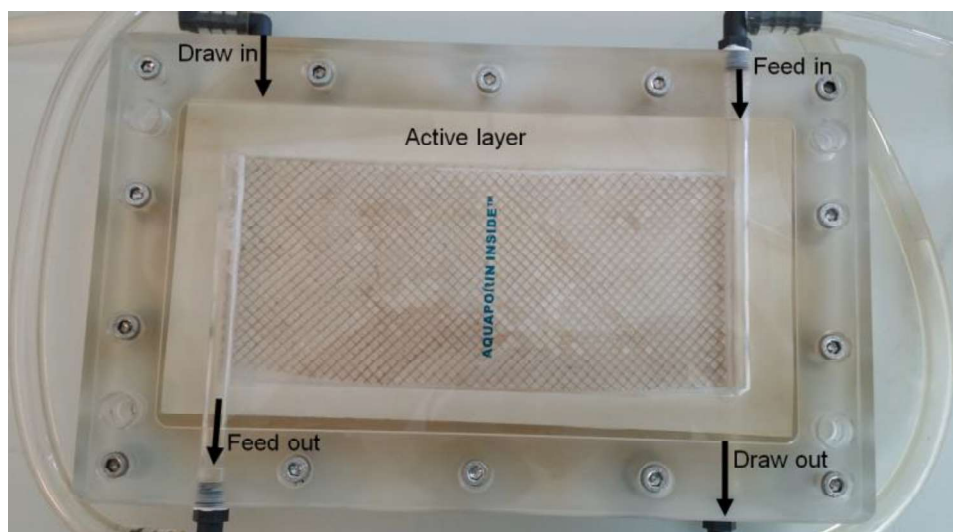


Figure 8. Aquaporin Inside™ membrane inside a membrane-module used for the bench-scale experiments. The feed solution, e.g., raw municipal wastewater, was applied on the active layer, and the draw solution, i.e., 2 M NaCl or Öresund seawater, was applied on the support layer.

4.1.7 Forward osmosis bench-scale setup

Each forward osmosis experiment was conducted as follows (**Paper II, III and IV**): 2 L of each feed and draw solution was placed in 5 L reservoirs, and the draw solution was placed on an electronic balance (see Figure 9). During the forward-osmosis experiment, the mass change of the draw solution was recorded every 5 min while cross-flow was applied to monitor the water flux course. The duration of the experiment was at least 4 hours, and the dilution factor of the draw solution, i.e., the volume ratio of the draw solution after and before the forward osmosis experiment, was set to 1.4. This choice was made to compare the conducted forward-osmosis experiments and membrane performances in terms of water flux and solute rejection.

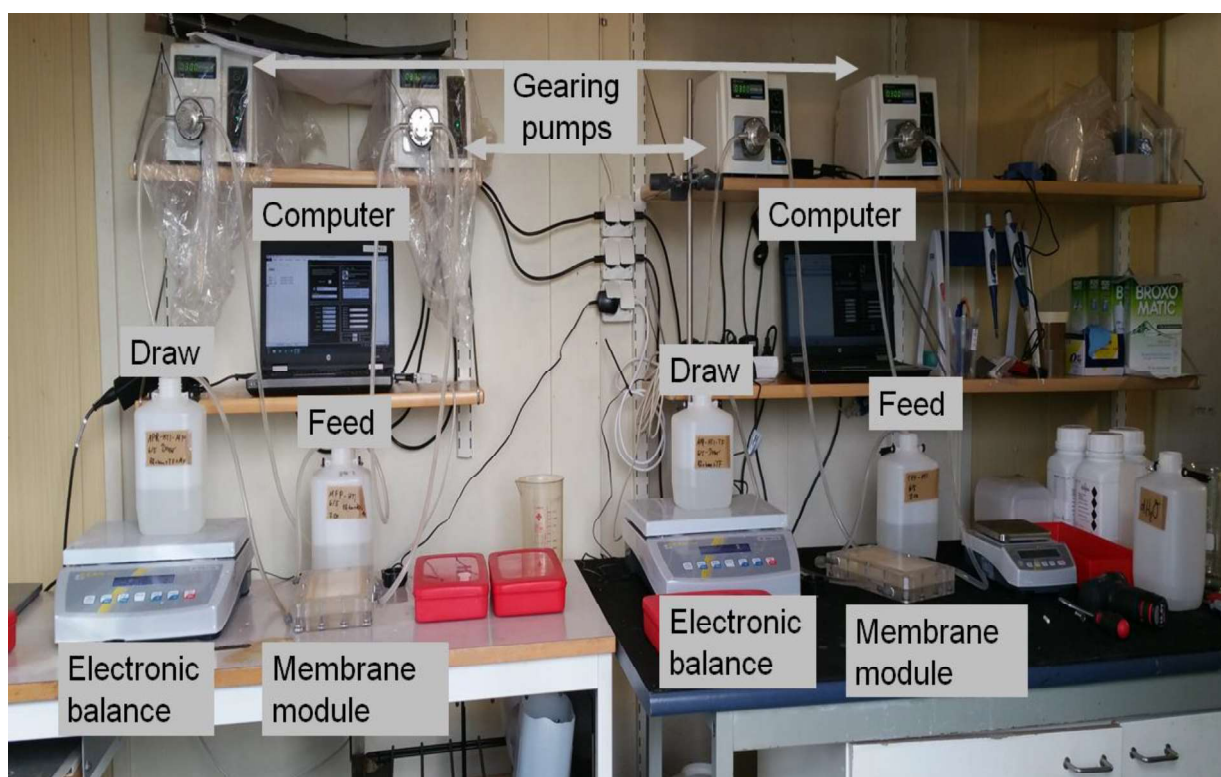


Figure 9. Dual bench scale setup for forward osmosis experiments (**Paper II, III, IV**) inside the pilot-plant container. The draw solution was placed on an electronic balance connected to a computer. Gearing pumps were used to generate cross-flow in the membrane module.

The effects of mechanical (**Paper II**) and physicochemical (**Paper III**) pretreatment in terms of water flux and solute rejection preceding forward osmosis have not been widely reported in the literature. Nonetheless, Coday *et al.* (2014) suggested using filtration, e.g., microfiltration, ultrafiltration, and/or coagulation/flocculation, for pretreatment before forward osmosis to reduce premature membrane fouling when treating domestic wastewater.

Hence, in this work, different mechanical (**Paper II**) and physicochemical (**Paper III**) pretreatment configurations have been tested before forward osmosis. For mechanical pretreatment, i.e., microsieving and microfiltration, the produced microsieve filtrate and microfiltration permeate have been used. In addition, raw municipal wastewater was used as a reference in terms of water flux and solute rejection.

To generate an osmotic pressure difference across the forward-osmosis membrane, 2 M NaCl was used as the model draw solution (**Paper II** and **Paper III**), which was the same molarity found in the study of Widjojo *et al.* (2011). However, in comparison to the studies using raw municipal wastewater, the molarity of the draw solution (2 M NaCl) was slightly higher because the Aquaporin InsideTM membrane was tested empirically for its feasibility for wastewater application.

Furthermore, because forward-osmosis experiments were conducted in southern Sweden, Öresund seawater taken from Lomma Bay (55°40'44.8"N 13°03'29.6"E) was used as the nearest and most natural draw solution for possible full-scale application (**Paper IV** and this thesis).

To evaluate and compare the conducted forward osmosis experiments, the solute rejection (R , %) and final concentration ($\text{mg}\cdot\text{L}^{-1}$) in the forward osmosis permeate (C_{Permeate}) were calculated as follows (**Paper II and III**):

$$R = 1 - \frac{V_{\text{Draw}(t=\text{End})} \cdot c_{\text{Draw}(t=\text{End})} - V_{\text{Draw}(t=0)} \cdot c_{\text{Draw}(t=0)}}{\frac{V_{\text{Permeate}}}{c_{\text{Feed}(t=0)} + c_{\text{Feed}(t=\text{End})}}} \cdot 100\% \text{ (Equation 2)}$$

where $V_{\text{Draw}(t=\text{End})}$ and $c_{\text{Draw}(t=\text{end})}$ ($\text{mg}\cdot\text{L}^{-1}$) are the final volume (L) and concentration ($\text{mg}\cdot\text{L}^{-1}$) in the draw solution at the end of the forward osmosis experiment, respectively. $V_{\text{Draw}(t=0)}$ and $c_{\text{Draw}(t=0)}$ ($\text{mg}\cdot\text{L}^{-1}$) are the initial volume (L) and initial concentration ($\text{mg}\cdot\text{L}^{-1}$) in the draw solution at the beginning of the forward osmosis experiment. The permeate volume V_{Permeate} (L) is the difference between the final and initial draw solution volumes. Furthermore, $c_{\text{Feed}(t=0)}$ ($\text{mg}\cdot\text{L}^{-1}$) is the initial concentration and $c_{\text{Feed}(t=\text{End})}$ ($\text{mg}\cdot\text{L}^{-1}$) is the final concentration in the feed solution, respectively. The final concentration in the permeate (C_{Permeate} , $\text{mg}\cdot\text{L}^{-1}$) was calculated as follows:

$$C_{\text{(Permeate)}} = \frac{V_{\text{Draw}(t=\text{End})} \cdot c_{\text{Draw}(t=\text{End})} - V_{\text{Draw}(t=0)} \cdot c_{\text{Draw}(t=0)}}{V_{\text{Permeate}}} \text{ (Equation 3)}$$

4.2 Direct membrane filtration experiments

4.2.1 Mechanical pretreatment

To investigate the importance of mechanical pretreatment, e.g., microsieving, the initial experiment (**Paper I**) was conducted in parallel by directly microfiltering raw municipal wastewater and microsieving the filtrate (see Figure 10). Furthermore, no studies were found that continuously operated microfiltration for treating raw municipal wastewater at a constant trans-membrane pressure of 3 kPa without backflushing. Thus, the main focus of this experiment was on the operational behaviour with and without microsieving in terms of the permeate flux course and operational stability of the municipal wastewater treatment system.

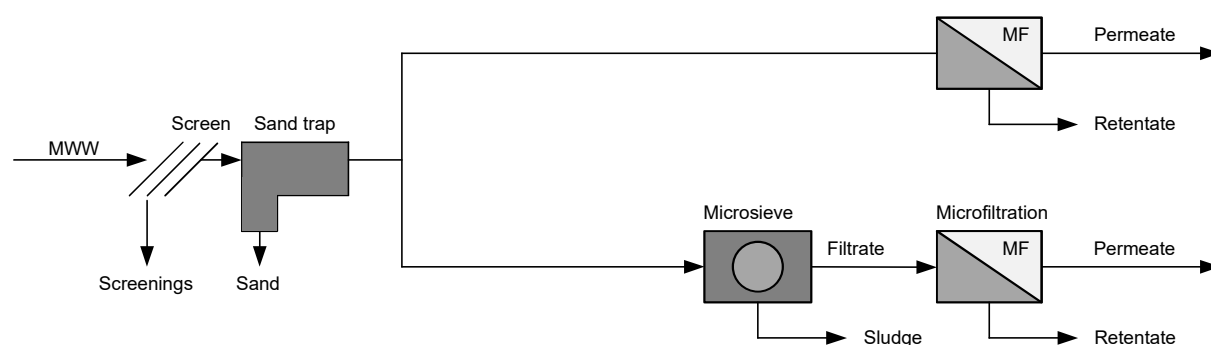


Figure 10. Schematic layout of the conducted direct membrane filtration experiments (**Paper I**) using raw or mechanically pretreated municipal wastewater.

4.2.2 Physicochemical pretreatment

The application of coagulants (PACl, FeCl_3) before microfiltration was investigated by Abdessemed and Nezzal (2002), Delgado Diaz *et al.* (2012) and Jin *et al.* (2015). Furthermore, coagulation (Remy *et al.*, 2014; Väänänen *et al.*, 2016) and coagulation/flocculation (Väänänen *et al.*, 2016) were combined with microsieving for primary treatment. However, no studies were found that investigated the effects of microfiltration combining coagulation, flocculation and microsieving for the pretreatment of raw municipal wastewater.

In this study, PACl and a cationic polymer were used for coagulation and cationic coagulation, respectively. Coagulation/flocculation was conducted using PACl and an anionic or cationic polymer. Figure 11 shows the conducted experiments regarding physicochemical pretreatment before microfiltration (**Paper I**).

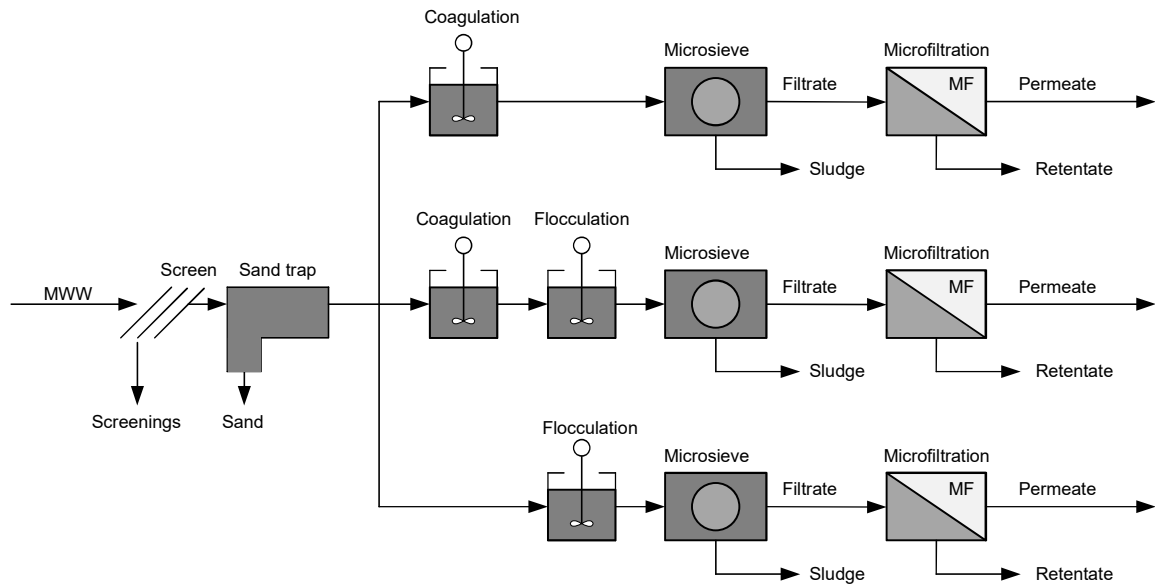


Figure 11. Schematic layout of the conducted direct membrane filtration experiments (**Paper I**) applying the physicochemically pre-treated microsieve filtrate. PACI was used for coagulation, PACI was used in combination with the anionic or cationic polymer for coagulation and flocculation, and the cationic flocculant was used for cationic polymer coagulation.

4.3 Direct forward osmosis experiments

4.3.1 Mechanical pretreatment before forward osmosis

As an overview of the mechanical pretreatment experiments before forward osmosis, Figure 12 depicts the conducted forward-osmosis experiments using 2 M NaCl and Öresund seawater as the draw solution. Microsieving and microfiltration were conducted in the same manner as described in the direct membrane filtration section and in **Paper I**.

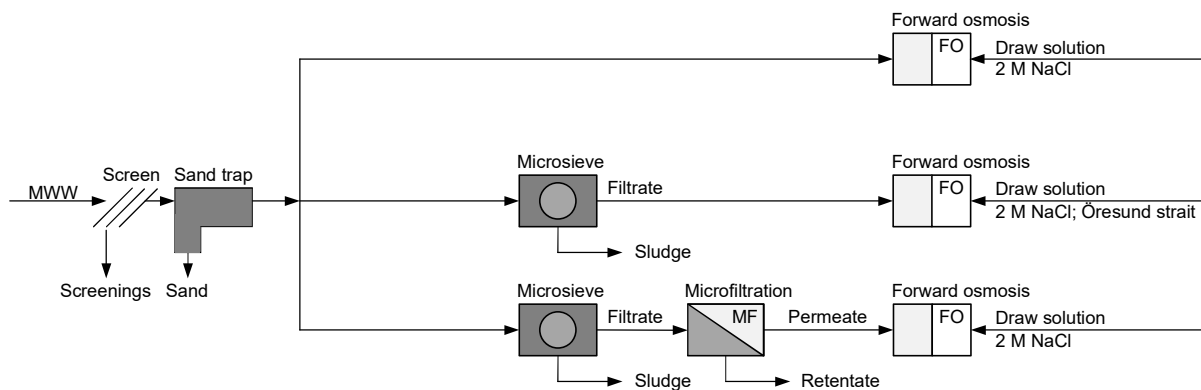


Figure 12.

Schematic layout of the conducted forward osmosis experiments (**Paper II**) applying raw municipal wastewater, microsieve filtrate and microfiltration permeate. In all experiments, 2 M NaCl was used as the draw solution. Öresund seawater was only used with the microsieve filtrate.

4.3.2 Physicochemical pretreatment before forward osmosis

To investigate the effects of physicochemical pretreatment before forward osmosis, different process configurations were applied (see Figure 13). The feed solutions for forward osmosis consisted of either microsieve filtrate or microfiltration permeate. Coagulation, flocculation, microsieving and microfiltration were conducted as described in **Paper I**. Furthermore, the model draw solution (2 M NaCl) and Öresund seawater were used for the microsieve filtrate (**Paper III**, **Paper IV** and this thesis) and 2 M NaCl was used for the produced microfiltration permeate (**Paper III**).

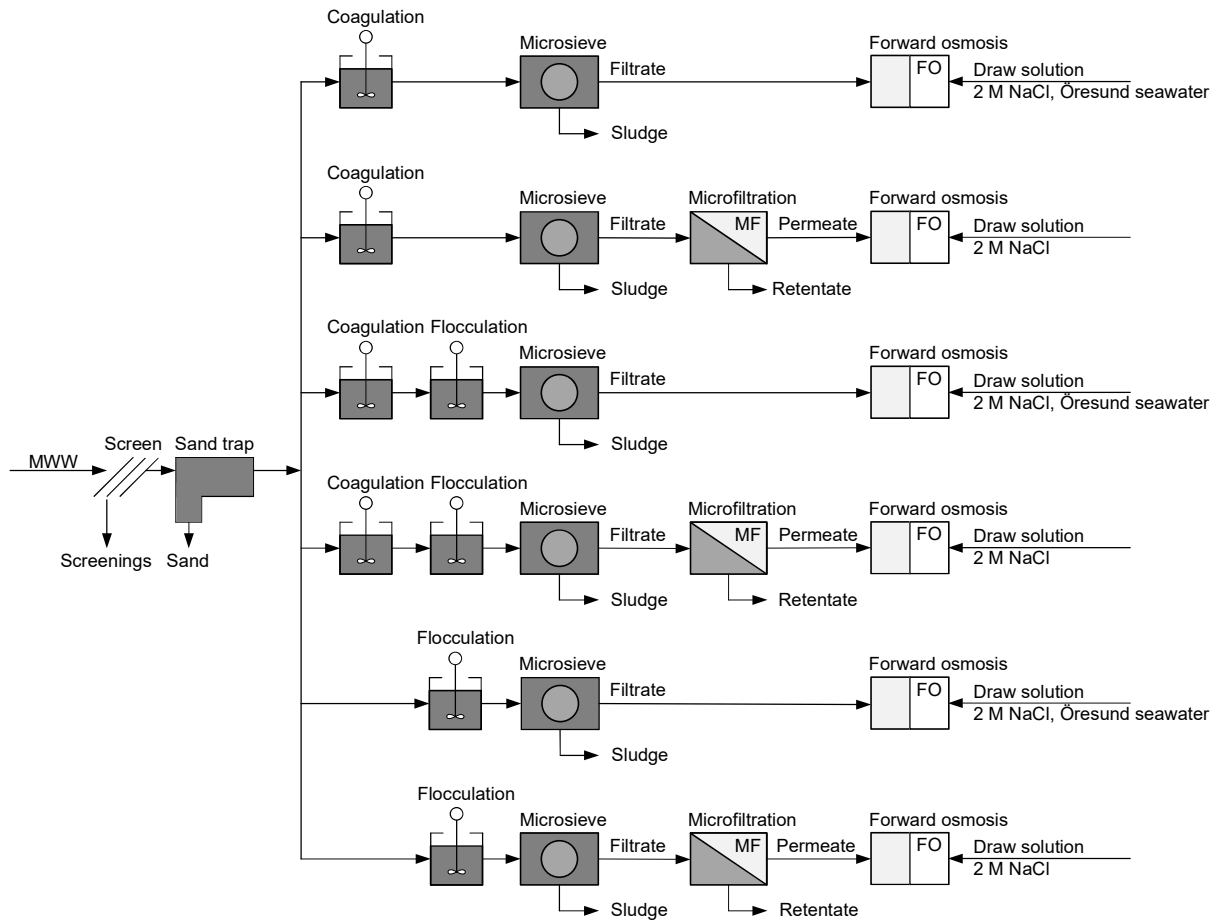


Figure 13. Schematic layout of the conducted forward osmosis experiments (**Paper III and IV**) using the physicochemical pre-treated microsieved filtrate. PACI was used for coagulation, the anionic or cationic polymers were used for coagulation and flocculation, and the cationic flocculant was used for cationic polymer coagulation. In all experiments, 2 M NaCl was used as the draw solution. Öresund seawater was only used with the microsieved filtrate as the feed solution.

5 Results and Discussion

5.1 Direct membrane filtration

Microfiltration was directly applied for treating (1) raw municipal wastewater and (2) microsieve filtrate with a 24 hours delay. The water flux when using the raw municipal wastewater varied strongly (see left figure in Figure 14) during operation in comparison to the very stable water flux that was observed when using the microsieve filtrate (see right figure in Figure 14). After 166 hours and 190 hours, respectively, both membranes underwent membrane cleaning (as shown by the dotted vertical line in both figures in Figure 14). Thereafter, the feed to the membrane tanks was switched to rule out system bias. However, during the second period, operational problems in the form of pipe and instrument clogging occurred due to the presence of debris in the membrane tank when treating the raw municipal wastewater. This resulted in constantly increasing trans-membrane pressure until the water level over the membrane finally overflowed (max. 10 kPa) from the membrane tank. Consequently, all of the later experiments were conducted by employing microsieving before microfiltration to ensure operational stability (**Paper I**).

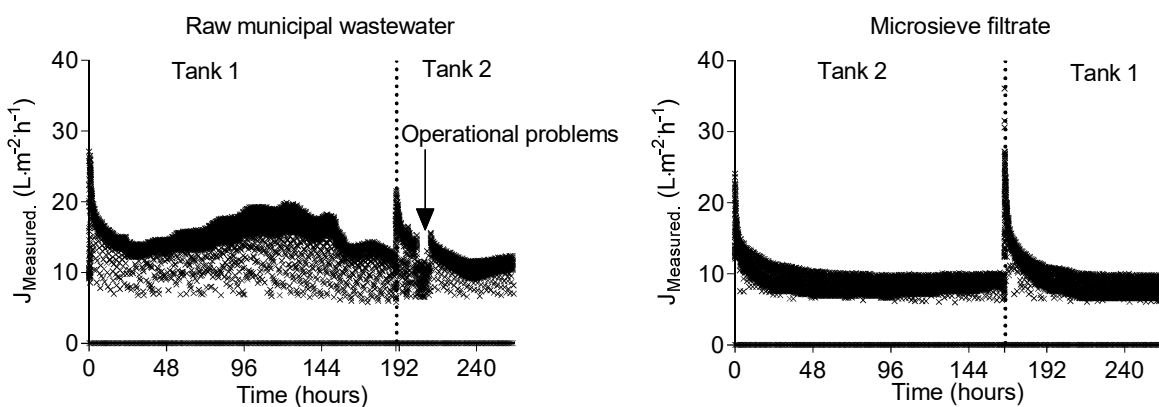


Figure 14.

Microfiltration permeate flux course using the raw municipal wastewater (left, Tank 1) and the microsieve filtrate (right, Tank 2) as feed solutions for microfiltration. The vertical dotted line represents membrane cleaning. Thereafter, the feed to the membrane tanks was switched; Tank 2 received raw municipal wastewater (left) and Tank 1 received microsieve filtrate (right) to rule out system bias (**Paper I**).

Furthermore, direct microfiltration of raw wastewater could result in problems because the combination of air scouring and small particles, e.g., sand, were assumed to have an abrasive effect on the microfiltration membrane, thus shortening the technical lifespan of the membrane.

5.1.1 Mechanical pretreatment

The microsieve filtrate was subjected to microfiltration, which resulted in an average permeate flux of $2.6 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ (see left figure in Figure 15). This permeate flux is substantially lower than the achieved permeate flux of $20 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ in the study of Lateef *et al.* (2013). However, the applied trans-membrane pressure in **Paper I** was significantly lower (0.03 bar) than the applied trans-membrane pressure(s) found in the studies shown in Table 1. The purpose in this study was to maintain a stable trans-membrane pressure to observe permeate flux instead of maintaining a stable permeate flux and compensating for the permeate flux loss by increasing the trans-membrane pressure.

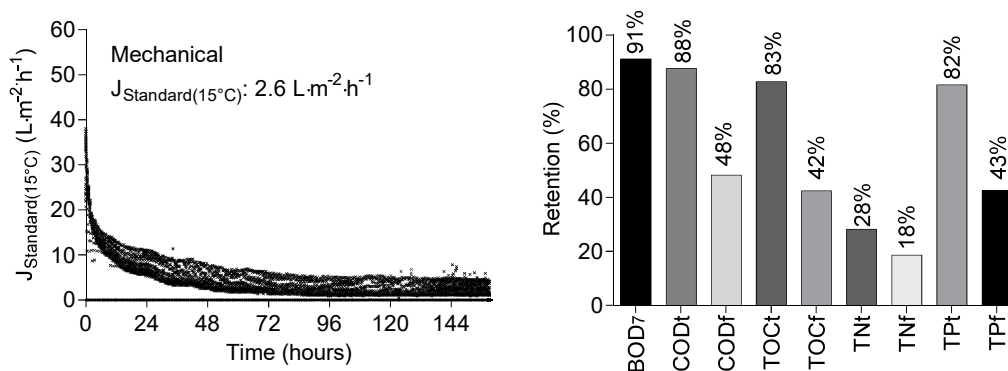


Figure 15.

Microfiltration permeate flux ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) over time (h) (left figure). Wastewater component retention (%) using microsieve filtrate as feed for microfiltration (right figure). The retention of the components was calculated according to Equation 1 by using the concentration ($\text{mg}\cdot\text{L}^{-1}$) in the raw municipal wastewater (c_{in}) and the concentration ($\text{mg}\cdot\text{L}^{-1}$) in the microfiltration permeate (c_{out}) (**Paper I**).

Microfiltration achieved high BOD₇, COD_t, TOC_t and TP_t retentions (%), as shown on the right side of Figure 15. Retentions of 91% ($17 \text{ mg}\cdot\text{L}^{-1}$) for BOD₇, 88% ($73 \text{ mg}\cdot\text{L}^{-1}$) for COD_t and 83% ($23 \text{ mg}\cdot\text{L}^{-1}$) for TOC_t were found because part of the COD fraction in municipal wastewater is particulate (Henze *et al.*, 2002). The presented retention values (%) correspond to those presented by Ahn *et al.* (2001) and Lateef *et al.* (2013).

Furthermore, the total nitrogen (TNt) and soluble total phosphorus (TPf) retentions were 28% ($37 \text{ mg}\cdot\text{L}^{-1}$) and 43% ($2.3 \text{ mg}\cdot\text{L}^{-1}$), respectively, which correspond to the values found by Ahn *et al.* (2000) for TN 36% and by Kolega *et al.* (1991) for TP 42-75%. At the full-scale Källby wastewater treatment plant, post-precipitated sludge is recirculated back before the sand trap, resulting in a high amount of particulate phosphorus in the raw municipal wastewater. Thus, the TPf values was used to calculate phosphorus retention.

5.1.2 Physicochemical pretreatment

5.1.2.1 Coagulation

The combination of PACl and microsieving achieved retentions of 22% ($586 \text{ mg}\cdot\text{L}^{-1}$) for SS, 40% ($188 \text{ mg}\cdot\text{L}^{-1}$) for BOD₇, 24% ($699 \text{ mg}\cdot\text{L}^{-1}$) for COD_t, 31% ($178 \text{ mg}\cdot\text{L}^{-1}$) for TOC_t, and 95% ($0.2 \text{ mg}\cdot\text{L}^{-1}$) for TPf on the microsieve.

In comparison to the aforementioned values using PACl as a coagulant, the use of only the cationic polymer in combination with microsieving achieved higher retentions of 97% ($12 \text{ mg}\cdot\text{L}^{-1}$) for SS, 89% ($16 \text{ mg}\cdot\text{L}^{-1}$) for BOD₇, 87% ($48 \text{ mg}\cdot\text{L}^{-1}$) for COD_t, and 82% ($19 \text{ mg}\cdot\text{L}^{-1}$) for TOC_t and a lower retention of 12% ($2.0 \text{ mg}\cdot\text{L}^{-1}$) for TPf on the microsieve. The obtained retention values considering the carbon fractions, i.e., SS, COD_t, TOC_t, correspond to the values found in Väänänen *et al.* (2016).

Despite the high carbon and phosphorus retentions that were observed when using PACl in combination with microsieving, microfiltration retained 96% ($11 \text{ mg}\cdot\text{L}^{-1}$) of BOD₇, 94% ($61 \text{ mg}\cdot\text{L}^{-1}$) of COD_t, 94% ($16 \text{ mg}\cdot\text{L}^{-1}$) of TOC_t and 98% ($0.1 \text{ mg}\cdot\text{L}^{-1}$) of TPf when PACl was used (see in the right side of Figure 16). The high COD retention of 94% corresponds to the range of 81%-95% found by Abdessemed and Nezzal (2002), Delgado Diaz *et al.* (2012) and Jin *et al.* (2015).

Furthermore, a higher permeate flux was obtained with PACl ($6.2 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) (see the left side of Figure 16) in comparison with the permeate flux with mechanical pretreatment ($2.6 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$). This finding agrees with the observations in Abdessemed and Nezzal (2002), who state that the permeate flux can be improved significantly when PACl is used.

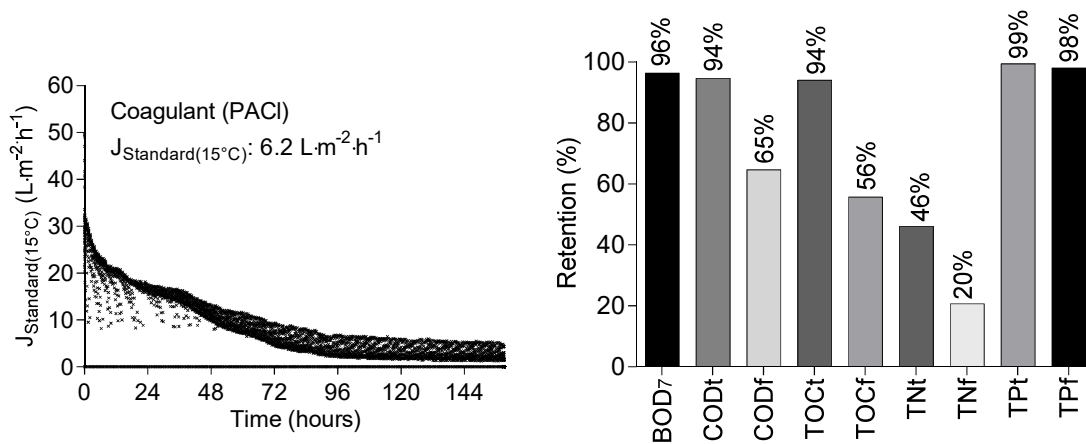


Figure 16. The microfiltration permeate flux ($L \cdot m^{-2} \cdot h^{-1}$) over time (h) (left figure). Wastewater component retentions (%) using PACI in combination with microsieving as physicochemical pretreatment before microfiltration (right figure). The retention is calculated according to Equation 1 and by using the concentration ($mg \cdot L^{-1}$) in the raw municipal wastewater (c_{in}) and concentration ($mg \cdot L^{-1}$) in the microfiltration permeate (c_{out}) (Paper I).

Cationic coagulation in combination with microsieving achieved higher carbon retentions than PACI (see the right portion of Figure 17). However, compared with PACI, only 15% of TPf was retained ($2.4 mg \cdot L^{-1}$). This low TPf removal resulted from the absence of metals with strong binding affinities e.g., Al^{3+} or Fe^{3+} . Furthermore, the obtained permeate flux of $2.6 L \cdot m^{-2} \cdot h^{-1}$ (see the left side of Figure 17), was less than half of the permeate flux that was achieved when using PACI ($6.2 L \cdot m^{-2} \cdot h^{-1}$). This low permeate flux shows that the cationic coagulant that was used negatively affected the membrane's performance.

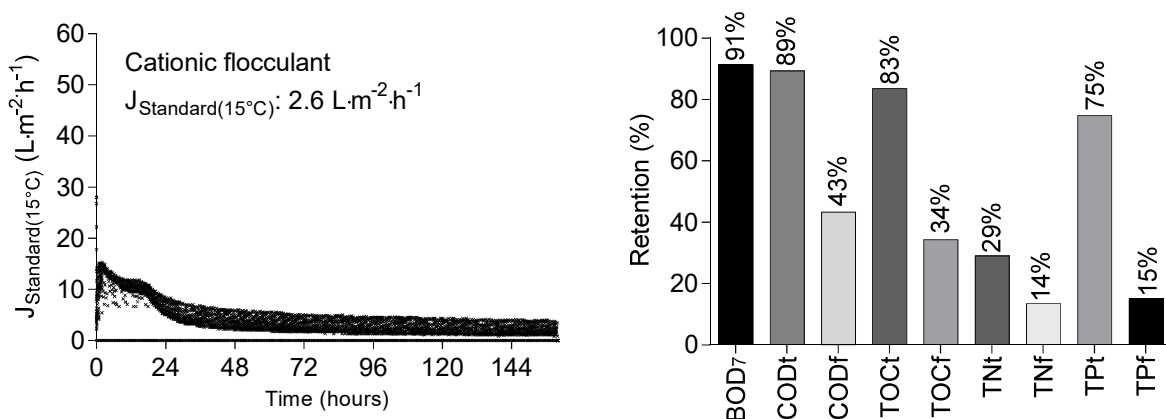


Figure 17. Microfiltration permeate flux ($L \cdot m^{-2} \cdot h^{-1}$) over time (h) (left figure). Wastewater component retentions (%) using a cationic flocculant as the coagulant in combination with microsieving for physicochemical pretreatment before microfiltration (right figure). The retention is calculated according to Equation 1 using the concentrations ($mg \cdot L^{-1}$) in the raw municipal wastewater (c_{in}) and microfiltration permeate (c_{out}) (Paper I).

5.1.2.2 Coagulation and flocculation

The combination of coagulation/flocculation with microsieving can achieve SS, COD and TP removals of >95%, 70-95% and >95, respectively, according to Remy *et al.* (2014) and Väänänen *et al.* (2016). In this study, the retentions of SS, COD and TPf by the microsieve were 98% ($13 \text{ mg}\cdot\text{L}^{-1}$), 90% ($77 \text{ mg}\cdot\text{L}^{-1}$), and 96% ($0.1 \text{ mg}\cdot\text{L}^{-1}$) applying anionic flocculant, and 99% ($6 \text{ mg}\cdot\text{L}^{-1}$), 89% ($60 \text{ mg}\cdot\text{L}^{-1}$) and 98% ($0.05 \text{ mg}\cdot\text{L}^{-1}$) for the cationic flocculant, respectively. Furthermore, 91% ($31 \text{ mg}\cdot\text{L}^{-1}$) or 89% ($20 \text{ mg}\cdot\text{L}^{-1}$) of the BOD_7 was retained when the anionic and cationic flocculants were used, respectively.

Microfiltration of the pre-treated microsieve filtrate following the use of the coagulant (PACl) and anionic flocculant resulted in final retentions of 96% ($16 \text{ mg}\cdot\text{L}^{-1}$) for BOD_7 , 94% ($41 \text{ mg}\cdot\text{L}^{-1}$) for COD_t , and >99% ($0.05 \text{ mg}\cdot\text{L}^{-1}$) for TPf (see the right side of Figure 18).

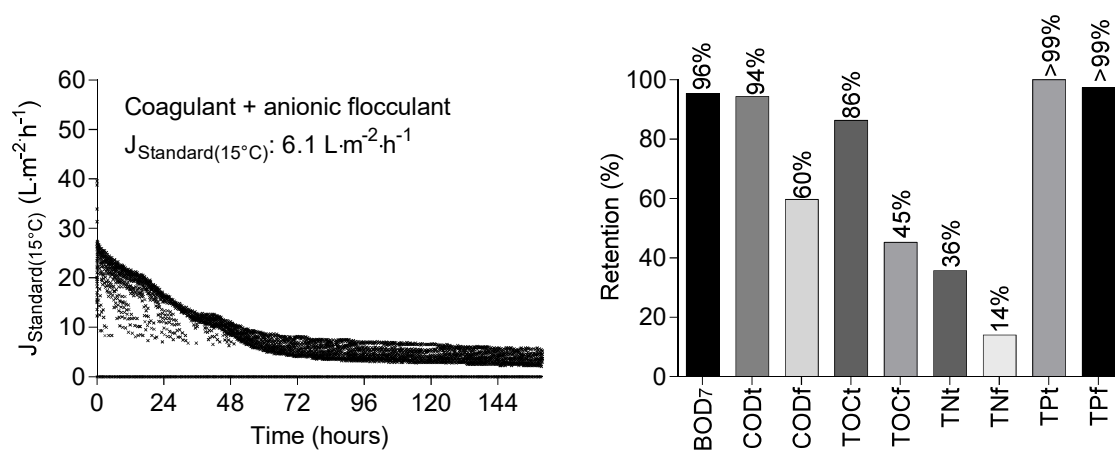


Figure 18.

Microfiltration permeate flux ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) over time (h) (left figure). Wastewater component retentions (%) using the coagulant and anionic flocculant in combination with microsieving for physicochemical pretreatment before microfiltration (right). The retention is calculated according to Equation 1 using the concentrations ($\text{mg}\cdot\text{L}^{-1}$) in the raw municipal wastewater (C_{in}) and microfiltration permeate (C_{out}) (**Paper I**).

Coagulation/cationic flocculation achieved final retentions of 94% ($13 \text{ mg}\cdot\text{L}^{-1}$) for BOD_7 , 91% ($44 \text{ mg}\cdot\text{L}^{-1}$) for COD_t , and >99% ($0.01 \text{ mg}\cdot\text{L}^{-1}$) for TPf which is shown in the right figure in Figure 19.

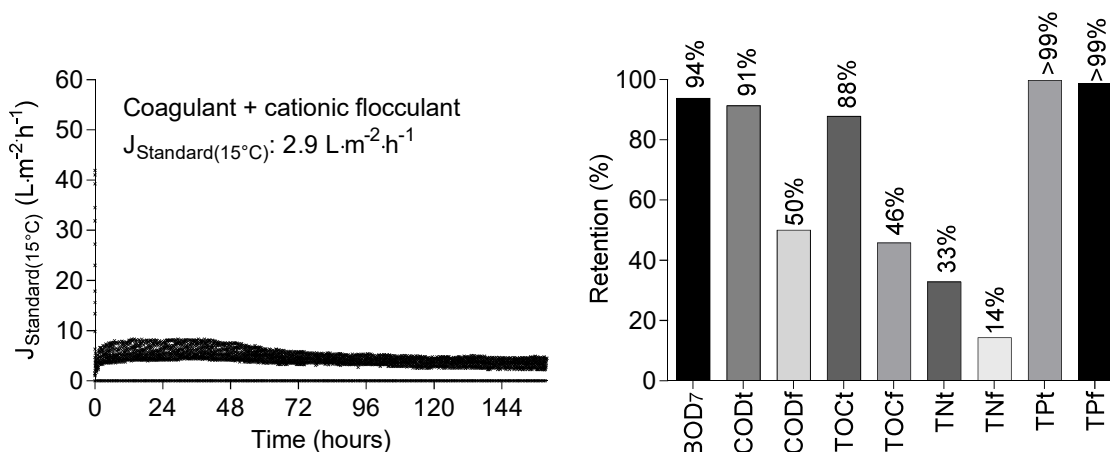


Figure 19.

Microfiltration permeate flux ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) over time (h) (left). Wastewater component retentions (%) using the coagulant and cationic flocculant in combination with microsieving as physicochemical pretreatment before microfiltration (right). The retention is calculated according to Equation 1 using the concentrations ($\text{mg}\cdot\text{L}^{-1}$) in the raw municipal wastewater (C_{in}) and in the microfiltration permeate (C_{out}) (**Paper I**).

The retention performances of both coagulant/flocculant applications were similar, but the achieved permeate flux differed significantly. The permeate flux using coagulation with the anionic flocculant ($6.1 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, left figure in Figure 18) was twice as large as the permeate flux achieved using the coagulant and cationic flocculant ($2.9 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, left figure in Figure 19).

Nevertheless, the involvement of the cationic flocculant in combination with microfiltration shows high carbon (BOD_7 , COD , TOC) retentions. In both cases, i.e., cationic coagulant, and coagulant/cationic flocculant, the obtained permeate flux was within the same magnitude as the permeate flux obtained for the mechanically pre-treated microsieve filtrate ($2.6 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, left figure in Figure 15).

5.1.2.3 Overview of the direct membrane filtration experiments

Microfiltration was conducted at a low trans-membrane pressure (3 kPa) and applied used for treating mechanically and physicochemically pre-treated raw municipal wastewater. The achieved results in terms of water flux, retention and the final concentrations in the permeate are shown in Figures 15 to 19 and are summarised in Table 5.

Table 5.

Overview of the achieved average standard permeate flux ($J_{\text{Standard}(15^{\circ}\text{C})}$, $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) and corresponding normalised permeate flux ($J_{\text{Norm.}}$, $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$), the final permeate concentration ($\text{mg}\cdot\text{L}^{-1}$) and corresponding retention (%) of the microfiltration used for the mechanical and different physicochemically pre-treated feed types. The retention (%) is calculated based on the initial concentration ($\text{mg}\cdot\text{L}^{-1}$) in the raw municipal wastewater and the final concentration ($\text{mg}\cdot\text{L}^{-1}$) in the microfiltration permeate (**Paper I**).

| Parameters | Mechanical | PACI | Cationic polymer | PACI + anionic polymer | PACI + cationic polymer |
|---|------------|---------|------------------|------------------------|-------------------------|
| $J_{\text{Standard}(15^{\circ}\text{C})}$ ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) | 2.6 | 6.2 | 2.6 | 6.1 | 2.9 |
| $J_{\text{Norm.}}$ ($\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$) | 87 | 207 | 87 | 203 | 97 |
| BOD ₇ ($\text{mg}\cdot\text{L}^{-1}$) | 17±5 | 11±2 | 13±2 | 16±13* | 13±2 |
| Retention (%) | (91±3) | (96±1) | (91±3) | (96±3) | (94±4) |
| COD _t ($\text{mg}\cdot\text{L}^{-1}$) | 73±27 | 61±35 | 48±6 | 41±26 | 44±10 |
| Retention (%) | (88±5) | (94±1) | (89±4) | (94±3) | (91±3) |
| COD _f ($\text{mg}\cdot\text{L}^{-1}$) | 64±25 | 41±11 | 47±4 | 41±26 | 42±8 |
| Retention (%) | (48±15) | (65±5) | (43±11) | (60±11) | (50±16) |
| TOC _t ($\text{mg}\cdot\text{L}^{-1}$) | 23±7 | 16±3 | 17±1 | 25±18 | 15±3 |
| Retention (%) | (83±7) | (94±2) | (83±6) | (86±9) | (88±6) |
| TOC _f ($\text{mg}\cdot\text{L}^{-1}$) | 21±5 | 16±3 | 17±1 | 17±8 | 15±3 |
| Retention (%) | (42±16) | (56±9) | (34±9) | (45±11) | (46±19) |
| TN _t ($\text{mg}\cdot\text{L}^{-1}$) | 37±10 | 42±4 | 36±6 | 34±10 | 30±6 |
| Retention (%) | (28±17) | (46±11) | (29±9) | (36±8) | (33±7) |
| TN _f ($\text{mg}\cdot\text{L}^{-1}$) | 32±5 | 41±4 | 36±6 | 34±11 | 30±6 |
| Retention (%) | (18±7) | (20±5) | (14±5) | (14±9) | (14±12) |
| TP _t ($\text{mg}\cdot\text{L}^{-1}$) | 2.4±0.6 | 0.2±0.3 | 2.5±0.8 | 0.02±0.01 | 0.02±0.01 |
| Retention (%) | (82±3) | (99±1) | (75±9) | (>99±0) | (>99±0) |
| TP _f ($\text{mg}\cdot\text{L}^{-1}$) | 2.3±0.7 | 0.1±0.1 | 2.4±1.0 | 0.05±0.03 | 0.03±0.01 |
| Retention (%) | (43±17) | (98±1) | (15±9) | (97±3) | (99±1) |

*measured only two times.

Furthermore, in Table 6, the retentions of BOD, COD, TOC, TN and TP in this study (**Paper I**) are put combined with the retentions (%) reported in the studies listed in Table 2.

Table 6.

Overview of the retentions (%) listed in Table 2 and **Paper I** (Table 5) for BOD, COD, TOC, TN and TP using microfiltration and ultrafiltration of raw municipal wastewater, primary effluent, and septic effluent.

| Feed | BOD | CODt | TOC | TN | TP | Reference |
|----------------------|--------|--------|-----|--------|--------|--|
| Raw MWW | 95% | 98% | 71% | 36% | | Ahn <i>et al.</i> , 2001 |
| Raw MWW | | 70-80% | | | | Lateef <i>et al.</i> , 2013 |
| Raw MWW* | | 95% | | | | Jin <i>et al.</i> , 2015 |
| Raw MWW | 91% | 88% | 83% | 28% | 82% | Paper I (Mechanical pretreatment) |
| Raw MWW** | 96% | 94% | 94% | 46% | 99% | Paper I (PACl) |
| Raw MWW** | 91% | 89% | 83% | 29% | 75% | Paper I (Cationic polymer) |
| Raw MWW** | 96% | 94% | 86% | 36% | >99% | Paper I (PACl + anionic polymer) |
| Raw MWW** | 94% | 91% | 88% | 33% | >99% | Paper I (PACl + cationic polymer) |
| Primary Eff. | 61-89% | | | | 42-75% | Kolega <i>et al.</i> , 1991 |
| Primary Eff. | 44% | 49% | | | | Juby <i>et al.</i> , 2000 |
| Primary Eff. | 48% | 50% | | | | Juby <i>et al.</i> , 2001 |
| Primary Eff. | 47% | 49% | | | | Sethi and Juby, 2002 |
| Primary Eff. | 57% | 65% | 8% | 0% | | Juby <i>et al.</i> , 2013 |
| Septic tank effluent | | 93% | 93% | 66% | | Ahn and Song, 1999 |
| Raw MWW | 94-99% | 97% | | | | Abdessemed <i>et al.</i> , 1999 |
| Raw MWW | | 69% | | 17% | 36% | van Nieuwenhuijzen <i>et al.</i> , 2000 |
| Raw MWW | 47-52% | 51-65% | | 13-22% | 18-53% | van Nieuwenhuijzen, 2002 |
| Raw MWW | | 37% | | 9% | 20% | Ravazzini <i>et al.</i> , 2005 |
| Primary Eff.* | | 88% | | | | Abdessemed and Nezzal, 2002 |
| Primary Eff. | | 42% | | 0% | 20% | Ravazzini <i>et al.</i> , 2005 |
| Primary Eff.* | | 81-95% | | | | Delgado Diaz <i>et al.</i> , 2012 |

*Chemicals were used in the form of coagulant, e.g., FeCl₃, PACl. **The used chemicals are described in the reference column in parentheses.

5.2 Direct membrane filtration and wastewater discharge limits

Different pretreatment methods, i.e., mechanical and physicochemical, before microfiltration, were tested for municipal wastewater treatment (**Paper I**). The discharge limits for municipal wastewater treatment plants in Sweden are regulated by European Directive (91/271/EEC), and total nitrogen removal is required for municipal wastewater treatment plants larger than 10 000 PE. For wastewater treatment plants with sizes of 2 000–10 000 PE, removal of BOD₇ and total phosphorus (TPt) is required to achieve discharge values of less than 15 mg·L⁻¹ and 0.5 mg·L⁻¹, respectively. As shown in Table 5, three treatment concepts fulfil the wastewater discharge demands: PACl only, PACl + anionic flocculation, and PACl + cationic flocculation (**Paper I**). However, as shown in Table 5, only 30-42% of TNt was retained, which agrees with the value of 36% observed by Ahn *et al.* (2001). Thus, the low retention for total nitrogen does not comply with the present wastewater discharge demands for large wastewater treatment plants (>10 000 PE).

However, the permeate flux using PACl + cationic flocculant was significant lower (2.9 L·m⁻²·h⁻¹) than the permeate fluxes when using PACl (6.2 L·m⁻²·h⁻¹) and PACl + anionic flocculant (6.1 L·m⁻²·h⁻¹); thus, increased membrane area demands and increased material costs are expected to compensate for the lower permeate flux.

Furthermore, considering that the discharge demands regarding, e.g., TPt, are expected to become more stringent in the future, the use of PACl + anionic flocculant is preferred because it results in TPt and TPs discharge of ≤0.05 mg·L⁻¹. Moreover, the combination of PACl + anionic polymer resulted in higher carbon removals (SS, CODt, TOCt) in combination with microsieving than PACl alone, potentially leading to lower membrane fouling, and the retained carbon can be used for biogas utilisation, as shown in Remy *et al.* (2014), Falk (2015) and **Paper IV**.

To compare each pretreatment method before microfiltration, Table 5 shows the obtained permeate fluxes, final BOD₇, CODt, CODf, TOCt, TOCf, TNt, TNf, TPt and TPf concentrations in the microfiltration permeate (mg·L⁻¹) and their respective degree of retention (%). In addition to the aforementioned measurements, the volatile fatty acid (VFA) content, alkalinity, total dissolved solids content, and electroconductivity decreased (as average values) by 81%, 37%, 11%, and 9%, respectively. The pH increased by 0.3 units due to the stripping of dissolved CO₂.

5.3 Direct forward osmosis

5.3.1 Mechanical pretreatment

The forward-osmosis membranes, i.e., AIM and HTI, were tested using three different feed solutions, i.e., raw municipal wastewater (Raw MWW), microsieve filtrate (MSF) and microfiltration permeate (MFP), for the feasibility of their use for municipal wastewater treatment. The feed solutions were produced in the pilot-scale plant and forward-osmosis experiments were conducted in a bench-scale study and using 2 M NaCl as the model draw solution (**Paper II**). The obtained results show that microsieving before forward osmosis can improve both the water flux and BOD₇, TPt and TPf solute rejections. However, with increasing pretreatment steps, i.e., microfiltration, the water flux increased for the AIM membrane, but the BOD₇, TPt and TPf solute rejection did not improve (as shown in Table 7).

Table 7.

Overview of the achieved water flux (J_w , L·m⁻²·h⁻¹), normalised water flux (J/J_0), final concentration in the forward osmosis permeate and corresponding solute rejection value (%) of the AIM and HTI membranes (**Paper II**). Forward osmosis was applied for different feed solutions, i.e., raw municipal wastewater (Raw MWW), microsieve filtrate (MSF) and microfiltration permeate (MFP). In this study, 2 M NaCl was used as the draw solution.

| Parameter | Raw MWW | | MSF | | MFP | |
|---|---------|------|------|------|------|------|
| | AIM | HTI | AIM | HTI | AIM | HTI |
| J_w (L·m ⁻² ·h ⁻¹) | 9.3 | 9.6 | 9.3 | 12.8 | 10.0 | 11.0 |
| J/J_0 | 0.84 | 0.74 | 0.94 | 0.83 | 0.90 | 0.89 |
| BOD ₇ (mg·L ⁻¹) | 1.1 | 1.5 | 0.3 | 1.5 | 0.4 | 1.5 |
| Rejection (%) | 100% | 99% | 100% | 99% | 98% | 96% |
| TNt (mg·L ⁻¹) | 38 | 40 | 27 | 15 | 41 | 25 |
| Rejection (%) | 32% | 37% | 14% | 61% | 0% | 28% |
| TNf (mg·L ⁻¹) | 38 | 38 | 26 | 13 | 41 | 25 |
| Rejection (%) | 16% | 28% | 0% | 53% | 0% | 20% |
| TPt (mg·L ⁻¹) | 0.15 | 0.21 | 0.03 | 0.10 | 0.04 | 0.04 |
| Rejection (%) | 99% | 98% | 100% | 99% | 99% | 98% |
| TPf (mg·L ⁻¹) | 0.03 | 0.06 | 0.03 | 0.04 | 0.04 | 0.04 |
| Rejection (%) | 98% | 97% | 98% | 98% | 98% | 98% |

Using forward osmosis to treat wastewater results in high carbon and phosphorus rejection without the involvement of chemicals (Xie *et al.*, 2013a; Zhang *et al.*, 2014; Wang *et al.*, 2016). Despite the availability of forward osmosis studies considering synthetic wastewater, raw wastewater and primary effluent, no studies have reported the rejection of BOD₇ from these types of wastewater and effluent. Nevertheless, because BOD is a fraction of COD and TOC (Henze *et al.*, 2002), high COD solute rejections of 72%-99% (Zhang *et al.* 2014a; Ansari *et al.*, 2016a; Wang *et al.*, 2016) and high TOC solute rejections of >95% (Xie *et al.*, 2013) have

been reported, implying that high BOD solute rejection occurs. Furthermore, the high TPt and TPf solute rejections in this work agree with the solute rejection values found in the range of 90-99% for synthetic wastewater (Valladares *et al.*, 2013; Xue *et al.*, 2015) and raw wastewater (Wang *et al.*, 2016). Moreover, high TP solute rejections (95-99%) were reported for anaerobic digester centrate (Holloway *et al.*, 2007; Xie *et al.*, 2014; Ansari *et al.*, 2016b) and urine (97-99%, Zhang *et al.*, 2014b).

5.3.2 Physicochemical pretreatment

The effects of physicochemical pretreatment, i.e., coagulation only, coagulation/flocculation, and flocculation only, in combination with microsieving and microfiltration on forward-osmosis performance in terms of water flux and solute rejection have not been widely reported.

5.3.2.1 Coagulation

Since AlCl_3 dissociates to form Al^{3+} and Cl^- in wastewater and the forward-osmosis membrane has a negatively charged surface (Elimelech *et al.*, 1994; Szymczyk *et al.*, 2010) that attracts Al^{3+} and other naturally occurring cations, e.g., Ca^{2+} and Fe^{3+} , charge neutralisation occurs and the flux decreases (Chester *et al.*, 2009).

Because a cationic polymer was used for coagulation, the same effect in terms of water flux decrease was observed, indicating that the membrane can attract cationic polymers. However, in both cases, the water flux was higher for the microsieve filtrate than for the microfiltration permeate. The higher water flux observed for the microsieve filtrate potentially occurred because the microsieve filtrate still contains suspended solids, which form a protective layer on the membrane surface (**Paper II**). Thus, the membrane is less exposed to the cationic species, e.g., Al^{3+} , Ca^{2+} , and Fe^{3+} , and the cationic polymer.

Furthermore, in comparison to the values of mechanical pretreatment, i.e., microsieve filtrate, the water flux and solute rejection (BOD_7 , TPt) were not improved by using PACl and the cationic polymer. However, a higher water flux was achieved when using the microfiltration permeate.

5.3.2.2 Coagulation and flocculation

During coagulation and anionic flocculation, the created flocs increased their binding strengths, linking the negatively charged polymer groups to the positively charged sites in the flocs and consequently neutralising the overall charge of the flocs. Thus, the remaining SS in the microsieve filtrate is assumed to be less prone to interactions with the negatively charged membrane surface, which could explain the higher water flux in comparison with only PACl. However, the final BOD_7 , TPt

and TPs concentrations in the permeate were similar to those obtained when using PACl.

Furthermore, since high carbon solute rejections can be achieved by using PACl + anionic polymer in combination with microsieving and microfiltration, simultaneous retention of dissolved organic carbon (DOC) is assumed, which is an important fouling factor for thin-film-composite reverse-osmosis membranes (Chester *et al.*, 2009). Thus, a high water flux ($12.0 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$) was achieved in comparison to the achieved water fluxes with mechanical pretreatment (**Paper II**).

5.3.2.3 Total nitrogen removal

The achieved TNt and TNf solute rejection with mechanical pretreatment in this study were in the range of 26-41%, which lies in the solute rejection range of synthetic wastewater of 0-59% (Valladares *et al.*, 2013; Xue *et al.*, 2015). However, the TNt and TNf solute rejections in this study were lower than the values of 68-95% found for raw wastewater by Xie *et al.* (2015) and Wang *et al.* (2016). However, higher TNt and TNf solute rejections were achieved with physicochemical pretreatment (33-65%) in comparison to mechanical pretreatment (26-41%). In comparison to the high carbon rejection achieved when using forward osmosis, the low rejection of ammonium nitrogen potentially occurred for several reasons: (1) membrane defects on the active layer (Zhang *et al.*, 2014), (2) diffusion across the membrane since ammonium is a monovalent ion (Wang *et al.*, 2016) and (3) the pH dependence of the ammonium/ammonia balance that is moved towards the uncharged ammonia above pH 9.3 (Xue *et al.*, 2015). With increasing pH, the rejection of ammonium and ammonia decreases (Zhang *et al.*, 2014; Xue *et al.*, 2015).

The obtained water fluxes (J_w , $\text{L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$), the final concentrations ($\text{mg}\cdot\text{L}^{-1}$) in the permeate, and the solute rejections (%) of the different physicochemical pre-treated feed solutions are summarised in Table 8. This table provides an overview that can be used to evaluate and compare the use of chemical and mechanical pretreatment methods before forward osmosis.

Table 8.

Overview of the achieved water flux (J_w , $L \cdot m^{-2} \cdot h^{-1}$), normalised water flux (J/J_0), final concentrations ($mg \cdot L^{-1}$) in the forward osmosis permeate and corresponding solute rejection values (%) of the AIM (**Paper III**). Forward osmosis was applied to different physiochemical pre-treated feed solutions from both microsieve filtrate (MSF) and microfiltration permeate (MFP). In this study, 2 M NaCl was used as the draw solution.

| Parameter | PACl | | Cationic polymer | | PACl + anionic polymer | | PACl + cationic polymer | |
|--|------|------|------------------|------|------------------------|------|-------------------------|-------|
| | MSF | MFP | MSF | MFP | MSF | MFP | MSF | MFP |
| J_w ($L \cdot m^{-2} \cdot h^{-1}$) | 8.9 | 11.0 | 9.0 | 10.9 | 9.8 | 12.0 | 8.7 | 9.2 |
| J/J_0 | 0.92 | 0.83 | 0.89 | 0.78 | 0.81 | 0.90 | 0.81 | 0.91 |
| BOD_7 ($mg \cdot L^{-1}$) | 7 | 3 | 21 | 14 | 5 | 5 | 18 | 16 |
| Rejection | 98% | 87% | 44% | 44% | 71% | 86% | 1% | 44% |
| TNt ($mg \cdot L^{-1}$) | 19 | 32 | 11 | 26 | 13 | 13 | 14 | 18 |
| Rejection | 44% | 39% | 65% | 26% | 42% | 41% | 39% | 38% |
| TNf ($mg \cdot L^{-1}$) | 19 | 30 | 12 | 26 | 35 | 35 | 13 | 19 |
| Rejection | 65% | 40% | 63% | 26% | 35% | 35% | 42% | 33% |
| TPt ($mg \cdot L^{-1}$) | 0.06 | 0.05 | 0.1 | 0.05 | 0.05 | 0.04 | 0.05 | <0.03 |
| Rejection | >99% | 88% | 78% | >99% | 98% | 37% | 95% | >99% |
| TPf ($mg \cdot L^{-1}$) | 0.06 | 0.05 | 0.1 | 0.05 | 0.05 | 0.04 | 0.05 | <0.03 |
| Rejection | >99% | 61% | 8% | >99% | 82% | 1% | 98% | >99% |

5.3.3 Öresund seawater as the draw solution

Instead of using a model draw solution (2 M NaCl), seawater from Öresund was used to investigate water flux and solute rejection for full-scale evaluation using only the microsieve filtrate. Because forward osmosis requires an osmotic pressure gradient across the membrane, the osmotic pressure of the feed solutions and Öresund seawater were measured. The osmotic pressure measurements revealed that the Öresund seawater and raw municipal wastewater had osmotic pressures of 250 kPa (2.5 bar) and 2-5 kPa (0.02-0.05 bar), respectively. The model draw solution had an osmotic pressure of 10.68 MPa (106.8 bar).

Mechanical pretreatment achieved the highest BOD_7 solute rejection, whereas PACl and the anionic flocculant achieved the highest water flux. However, in comparison to the observations obtained for the model draw solution, the use of the cationic flocculant resulted in higher BOD_7 solute rejections, but the water flux remained within the same magnitude as the water flux observed for mechanical pretreatment. Furthermore, a non-linear relationship between osmotic pressure and water flux was ascertained because the osmotic pressure ratio was 43 and the water flux ratio was 8.3, which could be attributed to fouling issues due to the high osmotic gradient.

Thus, a higher water flux using 2 M NaCl can be achieved, and more foulants come into contact with the membrane, which lowers the water flux. Because the Öresund

seawater has a low osmotic pressure, a lower water flux is obtained and the membranes' surface is not as affected by the foulants.

The obtained water fluxes (J_w , $L \cdot m^{-2} \cdot h^{-1}$), final permeate concentration ($mg \cdot L^{-1}$) and corresponding solute rejections (%) using Öresund seawater as the draw solution are summarised in Table 9. Furthermore, the solute rejection values (%) found in the studies listed in Table 4 are summarised in Table 10 along with the achieved solute rejection values listed in Table 7 (**Paper II**), Table 8 (**Paper III**) and Table 9 (**Paper IV**).

Table 9.

Overview of the achieved water fluxes (J_w , $L \cdot m^{-2} \cdot h^{-1}$), final concentrations ($mg \cdot L^{-1}$) in the forward osmosis permeate and corresponding solute rejection values (%) of the AIM (**Paper IV** and this thesis). Forward osmosis was used with different feed solutions, i.e., mechanical and physicochemical pretreatments using only the microsieve filtrate (MSF). Öresund seawater was used as a natural draw solution.

| Parameters | BOD ₇ | TPt | TPf | J _w ($L \cdot m^{-2} \cdot h^{-1}$) |
|---|------------------|------|------|--|
| Mechanical ($mg \cdot L^{-1}$) | 1.6 | 0.02 | 0.02 | 1.1 |
| Rejection (%) | 97% | 99% | 99% | |
| PACl only ($mg \cdot L^{-1}$) | 3.7 | 0.02 | 0.02 | 1.5 |
| Rejection (%) | 98% | 96% | 88% | |
| PACl + anionic flocculant ($mg \cdot L^{-1}$) | 6.7 | 0.02 | 0.02 | 1.4 |
| Rejection (%) | 86% | 81% | 70% | |
| PACl + anionic flocculant ($mg \cdot L^{-1}$) | 6.1 | 0.02 | 0.02 | 1.1 |
| Rejection (%) | 80% | 99% | 99% | |
| Cationic flocculant ($mg \cdot L^{-1}$) | 6.6 | 0.02 | 0.02 | 1.2 |
| Rejection (%) | 83% | 84% | 79% | |

Table 10.

Overview of the solute rejections (%) from the studies listed in Table 4, including the achieved solute rejections (%) in Paper II (Table 7) and Papers III and IV (Table 8 and 9), for BOD₇, COD, TOC, TNT and TPt when using forward osmosis for treating synthetic wastewater, raw municipal wastewater, anaerobic digester centrate and urine.

| Feed | BOD ₇ | COD | TOC | TNt | TPt | Reference |
|-----------------------------|------------------|------|------|--------|--------|---------------------------------|
| Synthetic wastewater | | 99% | | 56-59% | 99% | Valladares <i>et al.</i> , 2013 |
| | | | | 0% | >90% | Xue <i>et al.</i> , 2015 |
| Raw municipal wastewater | | | >95% | >95% | | Xie <i>et al.</i> , 2013 |
| | | 72% | | | | Zhang <i>et al.</i> , 2014a |
| | | >99% | | | | Ansari <i>et al.</i> , 2016a |
| | | >99% | | 68% | >99% | Wang <i>et al.</i> , 2016 |
| Raw municipal wastewater | 100% | | | 32% | 99% | Paper II (2 M NaCl) |
| | MSF | | | 14% | 100% | Paper II (2 M NaCl) |
| | MSF | | | | 97% | Paper IV (Öresund) |
| | MFP | | | 0% | 99% | Paper II (2 M NaCl) |
| PACI | MSF | | | 44% | >99% | Paper III (2 M NaCl) |
| | MSF | | | | 96% | This thesis (Öresund) |
| | MFP | | | 39% | 88% | Paper III (2 M NaCl) |
| Cationic polymer | MSF | | | 65% | 78% | Paper III (2 M NaCl) |
| | MSF | | | | 84% | This thesis (Öresund) |
| | MFP | | | 26% | >99% | Paper III (2 M NaCl) |
| PACI + anionic polymer | MSF | | | 42% | 98% | Paper III (2 M NaCl) |
| | MSF | | | | 81% | This thesis (Öresund) |
| | MFP | | | 41% | 37% | Paper III (2 M NaCl) |
| PACI + cationic polymer | MSF | | | 39% | 95% | Paper III (2 M NaCl) |
| | MSF | | | | 99% | This thesis (Öresund) |
| | MFP | | | 38% | >99% | Paper III (2 M NaCl) |
| Anaerobic digester centrate | | | | 85-97% | >99% | Holloway <i>et al.</i> , 2007 |
| | | | | >90% | >97% | Xie <i>et al.</i> , 2014 |
| | | | | | >95% | Ansari <i>et al.</i> , 2016b |
| Urine | | | | 31-91% | 97-99% | Zhang <i>et al.</i> , 2014b |

MSF: Microsieve filtrate; MFP: Microsieve permeate

5.4 Direct forward osmosis and wastewater discharge limits

The use of forward osmosis for treating the microsieve filtrate (mechanical) resulted in BOD₇ rejection of ~100%, which corresponded to a final concentration of 0.3 mg·L⁻¹ in the permeate (**Paper II**). This concentration was lower than the concentrations observed when using the remaining pretreatment methods in this study (**Paper II** and **Paper III**). Furthermore, TPt and TPf were rejected to a high degree (≥98%), corresponding to 0.03 mg·L⁻¹ in the forward osmosis permeate. However, these rejection values were achieved for the model draw solution (2 M NaCl). Nevertheless, using Öresund seawater as draw solution, the rejections of 97% (1.6 mg·L⁻¹) for the BOD₇ and 99% (0.02 mg·L⁻¹) for both TPt and TPf remained high, and the final concentration in the forward osmosis permeate remained low. Considering the achieved solute rejection values and the corresponding low BOD₇ and TPt concentrations, microsieving in combination with forward osmosis can comply with the wastewater discharge demands for small- and medium-sized wastewater treatment plants in Sweden. Furthermore, DFO does not require coagulation and flocculation as it is needed with DMF.

5.5 Evaluation of direct membrane filtration and direct forward osmosis for full-scale applications

Two treatment concepts were selected to evaluate the specific electricity (kWh_{el}·PE⁻¹·year⁻¹, kWh_{el}·m⁻³), specific energy (kWh·PE⁻¹·year⁻¹, kWh·m⁻³) and specific area demand (m²·PE⁻¹) for full-scale applications based on the findings presented in **Paper I-IV**. The selection criterion for this evaluation was the fulfilment of the Swedish wastewater discharge demands for small- and medium-sized wastewater treatment plants (2 000-10 000 PE) because total nitrogen cannot be retained sufficiently to fulfil the outlet demands for larger wastewater treatment plants (>10 000 PE).

One configuration was chosen from each concept, i.e., DMF with coagulation/anionic flocculation (**Paper I**) and DFO including only microsieving (**Paper II**). However, the water flux value obtained from using Öresund seawater (1.1 L·m⁻²·h⁻¹) as the natural draw solution was used instead (**Paper IV**) of the value that was obtained by using 2 M NaCl (9.3 L·m⁻²·h⁻¹) as the model draw solution (**Paper II**). Both selected concepts were feasible for full-scale wastewater treatment

applications and achieved high permeate flux (**Paper I**) and/or high BOD₇, TPt and TPs retentions (**Paper I, Paper II**). Furthermore, because more carbon was retained when using both selected concepts, methane gas potential tests were performed and evaluated further for full-scale production (**Paper IV**).

The full-scale evaluation using both concepts included screening, an aerated sand trap, an equalisation tank (retrofitted primary settler), microsieving and anaerobic digestion that included sludge handling, e.g., gravitational thickening, dewatering, centrifugation and gas storage. Data from the Källby wastewater treatment plant was used for screening, the aerated sand trap and anaerobic digestion. Furthermore, coagulation/flocculation, microsieving and microfiltration, including clean-in-place (CIP), were evaluated based on full-scale data as these methods are stand-alone applications and are commercially available.

Forward osmosis using the AIM was integrated in existing forward-osmosis systems. However, the AIM membrane performances were used to obtain specific membrane area demands and specific area demands, including clean-in-place. Furthermore, Öresund seawater was used as a draw solution to evaluate DFO for wastewater treatment in southern Sweden (**Paper IV**).

Because chemicals were used in both concepts, e.g., coagulation, flocculation, clean-in-place and sludge handling (flocculation), the specific energy demand for the production of each chemical was calculated.

These evaluations showed that the specific electricity demand for both concepts, i.e., DMF and DFO, including anaerobic digestion, was 0.55 kWh_{el}·m⁻³ (see the grey shaded area in Figure 20), which is 0.2 kWh_{el}·m⁻³ lower than the median value of 105 Swedish wastewater treatment plants with sizes of 1 500-10 000 PE (**Paper I**) (see the left side of Figure 20). Furthermore, the specific electricity demands of both the DMF and DFO concepts correspond to the specific electricity demands (as median value) of 192 Swedish wastewater treatment plants with sizes of 1 500-100 000 PE (see the right side of Figure 20).

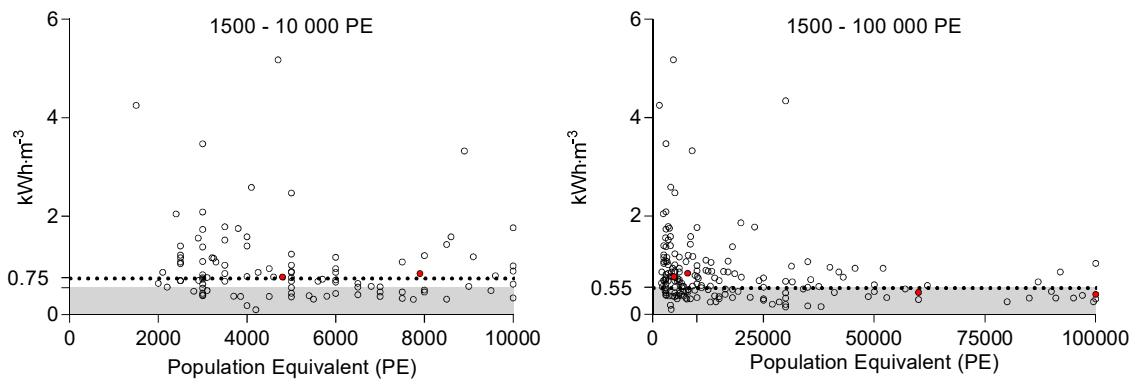


Figure 20.

Specific electricity consumption per cubic metre of treated wastewater at Swedish wastewater treatment plants. The left figure shows the specific electricity consumption for wastewater treatment plants with sizes of 1 500-10 000 PE ($n = 105$) (**Paper I**), and the right figure shows the specific electricity consumption for wastewater treatment plants with sizes of 1 500–100 000 PE ($n = 192$). The horizontal lines represent the median values of the specific electricity demands of the wastewater treatment plants and the grey shaded areas represent the specific electricity demands of the DMF and DFO concepts. The red dots represent the values of the selected wastewater treatment plants, as shown in Table 11, except for the Sjölanda wastewater treatment plant, which is 370 000 PE. Basic data were received with permission from Lustig and Dahlberg, 2012.

Lateef *et al.* (2013) concluded that direct membrane filtration is an attractive option because it sufficiently treats wastewater in a compact way. However, no studies have evaluated the specific area demands for direct membrane filtration (DMF) or direct forward osmosis (DFO). The evaluations in **Paper IV** show that the specific area demand including the main-stream treatment steps (i.e., screening, sand trap, equalisation tank, microsieving, microfiltration or forward-osmosis membrane including clean in place) and sludge-stream treatment steps (i.e., anaerobic digestion, including sludge handling, e.g., thickening, dewatering, and gas storage) was $0.046 \text{ m}^2 \cdot \text{PE}^{-1}$ for direct membrane filtration (DMF) and $0.051 \text{ m}^2 \cdot \text{PE}^{-1}$ for direct forward osmosis (DFO), including the Öresund seawater treatment by microsieving.

Although sufficient data are available regarding the specific electricity demands for Swedish wastewater treatment plants (see Figure 20) no data are available for the specific (active) area demands of wastewater treatment processes, including sludge treatment. Therefore, the specific area demand and specific electricity consumption were calculated for five municipal wastewater treatment plants with sizes of 4 800 to 370 000 PE, operating anaerobic digestion on site, and located near the Källby wastewater treatment plant (Table 11). The specific electricity demand ranged from $0.41\text{-}0.84 \text{ kWh}_{\text{el}} \cdot \text{m}^{-3}$ and is depicted by red dots in Figure 20. Furthermore, the specific area demand ($0.107\text{-}0.228 \text{ m}^2 \cdot \text{PE}^{-1}$) and the specific electricity demand showed that the specific area and energy demand decreases as the population equivalent increases.

Nevertheless, the specific electricity demand was $0.55 \text{ kWh}_{\text{el}} \cdot \text{m}^{-3}$ for both concepts (DMF and DFO) and was equal to the median specific electricity demand for wastewater treatment plants $>10\,000$ PE. However, both concepts clearly show that municipal wastewater can be treated using at least half of the surface area relative to wastewater treatment plants with sizes of 4 800 to 370 000 PE.

Table 11.

Specific electricity consumption and the (active) area demands of five selected conventional wastewater treatment plants, DMF with coagulation and flocculation (**Paper I**), and DFO with microsieving and Öresund seawater as draw solutions (**Paper IV**). All wastewater treatment plants include anaerobic digestion and sludge handling.

| Wastewater treatment plant | Size | Specific electricity consumption | | Specific area |
|----------------------------------|---------|--|--|-----------------------------------|
| | PE | $\text{kWh}_{\text{el}} \cdot \text{m}^{-3}$ | $\text{kWh}_{\text{el}} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$ | $\text{m}^2 \cdot \text{PE}^{-1}$ |
| Sjölunda* | 370 000 | 0.42 | 47.9 | 0.107 |
| Källby*.*** | 100 000 | 0.41 | 46.5 | 0.155 |
| Klagshamn* | 60 000 | 0.45 | 62.0 | 0.132 |
| DMF (Paper I) | 10 000 | 0.55 | 40.0 | 0.046 |
| FO (Öresund) (Paper IV) | 10 000 | 0.55 | 39.7 | 0.051 |
| Södra Sandby**.*** | 7 900 | 0.84 | 88.6 | 0.228 |
| Veberöd** | 4 800 | 0.77 | 54.8 | 0.190 |

*BOD₇, COD_t, TN_t and TP_t discharge demands. **BOD₇ and TP_t discharge demands. ***excluding ponds as the polishing step. (All values presented in Table 11, except the DMF and DFO data, were adapted with permission from Ulf Nyberg and Emma Sjöberg, VA SYD, Malmö, Sweden, 2016).

Furthermore, because more carbon can be retained in both DMF and DFO concepts in comparison to conventional wastewater treatment, this leads to a higher specific energy production of $102 \text{ kWh} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$ and $100 \text{ kWh} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$, respectively compared to $58 \text{ kWh} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$ (**Paper IV**). Converting methane gas to 40% electricity, 50% heat and 10% loss would result in an electricity- and energy positive wastewater treatment plant, see Table 12.

Table 12.

Summary of the specific electricity and energy surplus of the DMF and DFO seawater concepts, including sludge treatment and methane gas production (**Paper IV**).

| | Specific electricity surplus | | Specific heat surplus | |
|----------------------|--|---|--|----------------------------------|
| | $\text{kWh}_{\text{el}} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$ | $\text{Wh}_{\text{el}} \cdot \text{m}^{-3}$ | $\text{kWh} \cdot \text{PE}^{-1} \cdot \text{year}^{-1}$ | $\text{kWh} \cdot \text{m}^{-3}$ |
| DMF | 1 | 14 | 43 | 0.6 |
| DFO (Öresund) | 4 | 55 | 58 | 0.8 |

6 Conclusions

The main conclusion obtained from this work is that the Swedish discharge demands for small- and medium-sized wastewater treatment plants can be met without using biological treatment. The demands can be met by using either direct membrane filtration consisting of coagulation, anionic flocculation, microsieving and microfiltration or by direct forward osmosis consisting of microsieving and using Öresund seawater as a natural draw solution. Both concepts have a smaller area demand than conventional biological wastewater treatment but only direct membrane filtration is ready for full-scale testing. The work indicates that the two concepts are electricity and energy positive because more biogas can be produced than in conventional biological wastewater treatment.

6.1 Direct membrane filtration concept

The direct membrane filtration concept can be integrated at full-scale wastewater treatment plants because all applications, i.e., coagulation/flocculation, microsieving and microfiltration, are commercially available.

To ensure stable operation of the direct membrane filtration concept, microsieving before microfiltration is a necessity. Microfiltration can be performed at a low trans-membrane pressure (0.03 bar), but coagulation is necessary for increasing the permeate flux and for complying with the present wastewater discharge demands. To meet future discharge demands, e.g., increased carbon and phosphorus reduction, coagulation and flocculation will become a necessity. Considering the specific electricity consumption, air scouring is the most consuming operation.

6.2 Direct forward osmosis concept

The direct forward osmosis concept using microsieve filtrate as a feed solution and seawater as a natural draw solution can be considered as a future wastewater treatment concept.

The direct forward osmosis concept applied to microsieve filtrate can achieve higher BOD₇ and TPt solute rejections than the direct membrane filtration concept. Physicochemical pretreatment in combination with forward osmosis can improve the water flux but can result in lower solute rejections than mechanical pretreatment alone. The evaluation of the direct forward concepts resulted in positive gross electricity and energy production potentials.

7 Future work

Papers I-III show that municipal wastewater can be treated mechanically and physicochemically to fulfil the Swedish discharge demands for small- and medium-sized wastewater treatment plants. Furthermore, the generated retentate streams from the selected DMF and DFO concepts resulted in greater biogas production (**Paper IV**) than conventional wastewater treatment. Nevertheless, the selected DMF concept with physicochemical pretreatment (**Paper I**) and the DFO concept with mechanical pretreatment (**Paper II**) must be investigated further.

7.1 Direct membrane filtration

To verify the feasibility of the evaluated DMF-concept as presented and evaluated in **Paper IV**, up-scaling to a full-scale test would be the next step because all equipment is stand-alone and commercially available. The full-scale test could be performed at a small-sized wastewater treatment plant by initially treating a part of the total diurnal wastewater flow. The operation of the DMF-concept should be performed for at least one year to include seasonal variations such as diurnal flow, wastewater quality and temperature. Furthermore, performance tuning, e.g., alternating air-scouring, is necessary to decrease the specific electricity demand.

7.2 Direct forward osmosis

Forward osmosis using the AIM membrane can retain more BOD₇ and TPt than the DMF concept. However, pilot-plant testing can enable continuous and long-term studies. Öresund seawater could be used as a cheap draw solution because draw regeneration would not be necessary in this case. Furthermore, to meet the total nitrogen demands for large wastewater treatment plants, the solute rejection of ammonium should be improved significantly, which requires further membrane development. The addition of a physicochemical pretreatment stage before forward osmosis requires higher capital and operational costs. Nonetheless, 20% less

forward osmosis membrane area is required compared with the application of a mechanical pretreatment. The operational and economic advantages and disadvantages of this method should be investigated.

7.3. Sidestream treatment

In this study, the retained carbon in both concepts was tested and evaluated for biogas production and energy production. However, the presented concepts still contain process streams that must be handled. For example, the microfiltration permeate is particle free and may be suitable for nitrogen and phosphorus (coagulant could be left out) recovery in the form of struvite generation. Forward-osmosis retentate contains nitrogen, phosphorus and soluble carbon, which could be applied for struvite generation, as well as carbon for biogas production in a process similar to the IMANSTM process. Hence, further thought and investigations should be conducted to achieve a complete concept to treatment, respectively, include all generated side streams.

7.4 Micropollutants and microplastics

The DMF and DFO concepts both enable high retentions of carbon and phosphorus. However, the degree of retention of micropollutants, heavy metals and microplastics should be investigated.

7.5 Water disinfection

Since the direct membrane filtration and direct forward osmosis concepts include membrane processes, the membrane can act as a barrier for bacteria, parasites and virus. However, the rejection of these pathogens should be investigated considering water disinfection.

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