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From Direct Membrane Filtration to Adsorption of Organic Micropollutants

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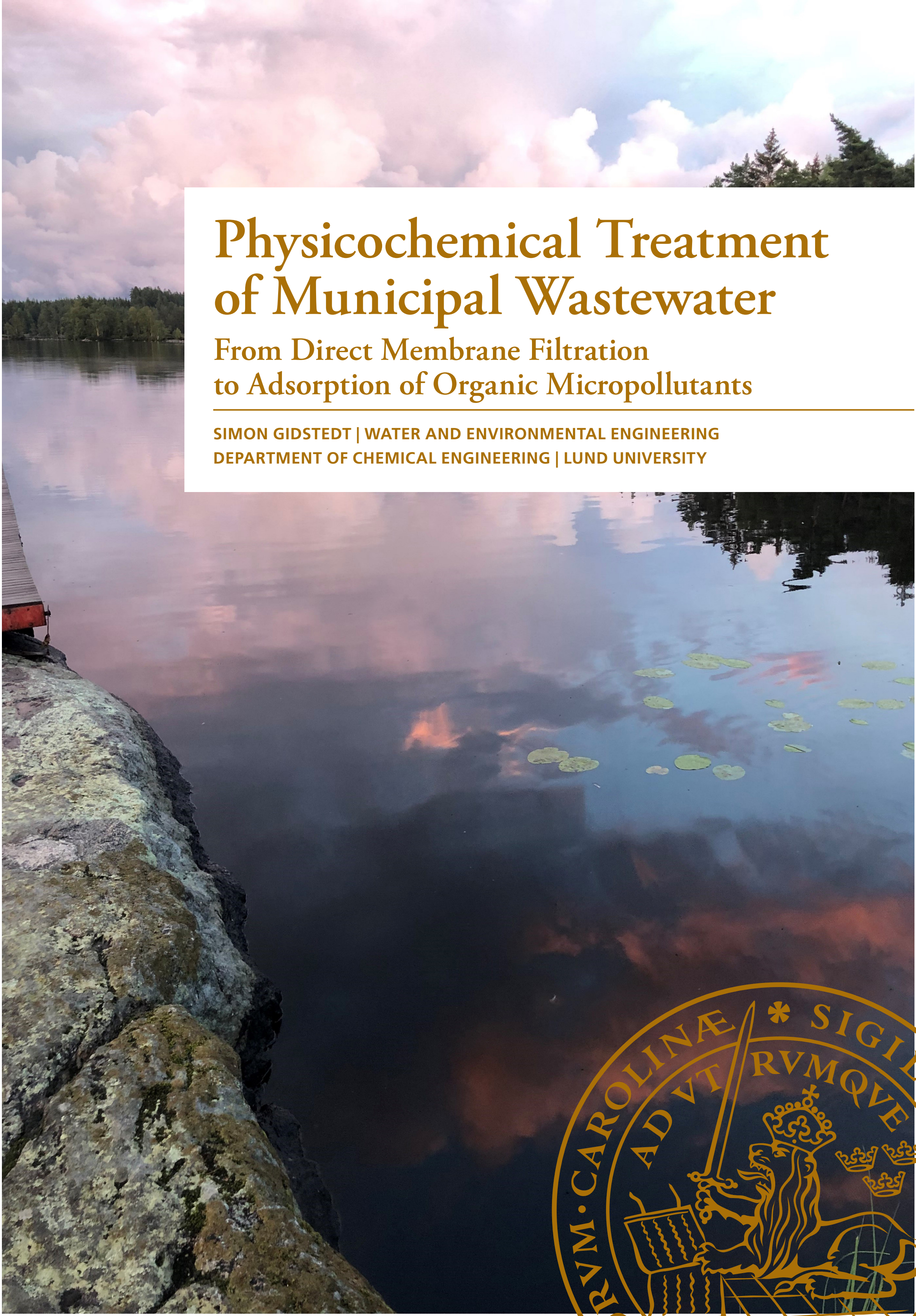

Physicochemical Treatment of Municipal Wastewater

From Direct Membrane Filtration to Adsorption of Organic Micropollutants

SIMON GIDSTEDT | WATER AND ENVIRONMENTAL ENGINEERING
DEPARTMENT OF CHEMICAL ENGINEERING | LUND UNIVERSITY

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WITHOUT PROPER WASTEWATER TREATMENT, our lakes and rivers would be polluted, leading to harmful effects on the aquatic environment. Pharmaceutical residues are one group of pollutants that needs to be removed. It is also important to minimize the amount of energy used in wastewater treatment in order to reduce the consumption of fossil fuels and reduce global warming. This dissertation concerns the treatment of municipal wastewater by using chemical precipitation, multiple stages of filtration and activated carbon for adsorption. The findings will hopefully contribute to ensuring that we protect the aquatic environment so that we can continue to enjoy it in the future.

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Physicochemical Treatment of Municipal Wastewater

From Direct Membrane Filtration to Adsorption of Organic Micropollutants

Simon Gidstedt

LICENTIATE DISSERTATION

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Abstract

Pharmaceutical residues and other organic micropollutants are transported via municipal wastewater to wastewater treatment plants (WWTPs). However, current treatment processes are not designed to remove organic micropollutants, and several such compounds have been found in recipients, where they can have negative effects on aquatic organisms. Additional processes are therefore required to reduce the release of organic micropollutants to the environment.

Additional treatment is likely to increase the energy demand at WWTPs. However, municipal wastewater contains organic matter that can be used for energy production in the form of biogas. Using physicochemical treatment methods would allow more of the organic content to be separated from the wastewater, increasing biogas production compared to conventional treatment with activated sludge.

This dissertation discusses the potential and limitations of physicochemical wastewater treatment on pilot scale by chemically enhanced primary treatment (CEPT) with microsieving, direct membrane filtration (DMF) and adsorption with activated carbon.

The results of pilot-scale tests showed that optimized CEPT with microsieving was very important for the high retention of particles, organic content and phosphorus, and for higher flux in subsequent DMF. However, the fouling of the membranes was severe, which prevented long-term operation. Biomethane produced from the sludge after microsieving could potentially produce enough energy to cover ~60% of the electricity required for such a treatment train (CEPT with microsieving and DMF). Supplementary biological treatment downstream of the treatment train could be used to reduce the remaining oxygen demand, and thus also reduce the dissolved organic content. The removal of organic micropollutants by CEPT with microsieving and DMF was minor. However, by subsequent filtration through granular activated carbon, a high removal could be achieved. The carbon filter could only be operated for 4 days due to the fouling of the membranes, and longer filtration times are suggested in future research.

Further laboratory-scale tests using powdered activated carbon showed that more organic micropollutants could be adsorbed when finer filtration was used as pre-treatment. Ultrafiltration was better than microfiltration, which was better than microsieving. This was related to lower concentrations of dissolved organic matter in the effluents the finer the filter used. The adsorption was highest in the effluent from a full-scale WWTP using biological treatment. This effluent had the lowest concentration of dissolved organic matter of all effluents. Finally, it was observed that granular activated carbon could be used to remove organic micropollutants after the WWTP and that the removals were similar to that by using powdered activated carbon.

Keywords: Direct membrane filtration; granular activated carbon; wastewater; chemically enhanced primary treatment; microsieving

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Plans are worthless, but planning is everything.

−Dwight D. Eisenhower

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My colleagues at Sweden Water Research for showing me the importance of our work within the water sector, and for letting me present my work at the Sweden Water Research Days. I hope I will meet many of you in the future.

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Thanks to my family for their support and for being there through happiness and sorrow. Thank you, Cilla, for believing in me and for you ever-lasting support.

Finally, thanks to everyone at the Department of Chemical Engineering for a pleasant and friendly working environment. I wish you all the best, and lots of granted applications!

Preface

This licentiate dissertation is the result of nearly four years of work as an industrial PhD student at Sweden Water Research AB and in the Water and Environmental Group at the Department of Chemical Engineering at Lund University. The research was conducted within the LESS is MORE project, funded by the INTERREG South Baltic Programme 2014-2020, the Swedish Agency for Marine and Water Management (Grant no. 2243-17) and Sweden Water Research AB. Thanks to this financing, I was able to participate in a project to construct a pilot-scale treatment plant, work with inspiring researchers, and be part of a very welcoming research environment, both at the Department of Chemical Engineering and at Sweden Water Research.

This dissertation presents a number of research studies on physicochemical wastewater treatment, including the removal of organic micropollutants by activated carbon adsorption. The research is mainly based on pilot-scale studies, but also on results from laboratory experiments. Part of the work resulted in a Master's Dissertation entitled *Direct Membrane Filtration – Investigating Fouling and Antifouling in Wastewater Treatment*, written by Omer Khalid (presented July 23rd, 2021). I was responsible for the original concept and co-supervised the student. A manuscript is being prepared which will be submitted for publication.

Abstract

Pharmaceutical residues and other organic micropollutants are transported via municipal wastewater to wastewater treatment plants (WWTPs). However, current treatment processes are not designed to remove organic micropollutants, and several such compounds have been found in recipients, where they can have negative effects on aquatic organisms. Additional processes are therefore required to reduce the release of organic micropollutants to the environment.

Additional treatment is likely to increase the energy demand at WWTPs. However, municipal wastewater contains organic matter that can be used for energy production in the form of biogas. Using physicochemical treatment methods would allow more of the organic content to be separated from the wastewater, increasing biogas production compared to conventional treatment with activated sludge.

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The results of pilot-scale tests showed that optimized CEPT with microsieving was very important for the high retention of particles, organic content and phosphorus, and for higher flux in subsequent DMF. However, the fouling of the membranes was severe, which prevented long-term operation. Biomethane produced from the sludge after microsieving could potentially produce enough energy to cover ~60% of the electricity required for such a treatment train (CEPT with microsieving and DMF). Supplementary biological treatment downstream of the treatment train could be used to reduce the remaining oxygen demand, and thus also reduce the dissolved organic content. The removal of organic micropollutants by CEPT with microsieving and DMF was small. However, by subsequent filtration through granular activated carbon, a high removal could be achieved. The carbon filter could only be operated for 4 days due to the fouling of the membranes, and longer filtration times are suggested in future research.

Laboratory-scale tests using powdered activated carbon showed that the tighter filtration used in the pre-treatment, the more organic micropollutants could be adsorbed. Ultrafiltration was better than microfiltration, which was better than microsieving. This was related to lower concentrations of dissolved organic matter in the effluents the finer the filter used. The adsorption was highest in the effluent from a full-scale WWTP using biological treatment. This effluent had the lowest concentration of dissolved organic matter of all effluents. Finally, it was observed that granular activated carbon could be used to remove organic micropollutants after the WWTP and that the removals were similar to that by using powdered activated carbon.

Populärvetenskaplig sammanfattning

Avloppsvatten med läkemedelsrester renas med filter och aktivt kol

VA-bolagen står inför flera utmaningar vad gäller rening av avloppsvatten. Till exempel att minska energianvändningen på reningsverken, samt att rena avloppsvattnet från läkemedelsrester. I den här studien har olika typer av filtrering i kombination med aktivt kol testats med målet att rena från läkemedelsrester samt förbättra energibalansen på reningsverk.

Läkedelsrester är en typ av mikroföroreningar som finns kvar i urin och avföring efter konsumtion och som transporteras med avloppsvattnet till reningsverken. Tyvärr så renas bara en bråkdel av läkemedelsresterna och man har uppmätt relativt höga koncentrationer av läkemedelsrester i vattendrag efter reningsverk där de utgör en fara för akvatiska miljön. Det finns undersökningar där man sett att hormonstörande ämnen från preventivmedel påverkat könsfördelningen hos vattenlevande djur. Man har också sett att ångestdämpande läkemedel gjort fiskar djärvare vilket påverkat deras chanser att överleva.

Många reningsverk i världen använder mikroorganismer för att behandla avloppsvatten. En behandling som fungerar bra för att bryta ned föroreningar (till exempel organiskt material) till koldioxid men som fungerar dåligt för att rena från läkemedelsrester. Rening med mikroorganismer kräver också att vattnet luftas vilket är energikrävande. I vår studie använde vi i stället filtrering för att separera och ta vara på organiskt material från avloppsvattnet. Efter att det organiska materialet filtrerats bort kan det omvandlas till energi i form av biogas genom att det rötas. Alltså ett plus i energibalansen på ett reningsverk.

Vanlig filtrering tar dock inte bort läkemedelsrester utan en extra behandling behövs efter en filtrering, eller för den delen, efter en behandling med mikroorganismer. För detta ändamål kan man använda aktivt kol i en process som kallas adsorption där läkemedelsresterna fastnar på kolet. I vår studie ville vi testa om filtrering av avloppsvatten fungerar lika bra som rening med mikroorganismer som en förbehandling till aktivt kol.

Genom experiment såg vi att filtrering fungerade bra som förbehandling till aktivt kol men att rening med mikroorganismer fungerade bättre. Anledningen var att efter filtrering fanns det mer lösta ämnen kvar i avloppsvattnet än efter rening med mikroorganismer och att dessa ämnen störde reningen från läkemedelsrester med aktivt kol. I experimenten såg vi också att desto finare filtrering som användes desto mer läkemedelsrester kunde tas bort i en efterföljande behandling med aktivt kol.

Med filtrering och aktivt kol kan vi alltså ta vara på det organiska materialet i avloppsvattnet för att producera biogas samt förhindra utsläppen av läkemedelsrester och därmed skydda hav, sjöar och vattendrag från att bli förorenade.

List of Papers

This dissertation in based on the following original articles, which will be referred to in the text by their Roman numerals:

Paper I **Gidstedt, S.**, Betsholtz, A., Falås, P., Cimbritz, M., Davidsson, Å., Micolucci, F., Svahn, O. (2022). A comparison of adsorption of organic micropollutants onto activated carbon following chemically enhanced primary treatment with microsieving, direct membrane filtration and tertiary treatment of municipal wastewater. *Science of the Total Environment,* 811, Article 152225.

Paper II **Gidstedt, S.**, Betsholtz, A., Cimbritz, M., Davidsson, Å., Hagman, M., Karlsson, S., Takman, M., Svahn, O., Micolucci, F. (2022). Chemically enhanced primary treatment, microsieving, direct membrane filtration and GAC filtration of municipal wastewater: a pilot-scale study. Manuscript submitted for publication.

My contribution to the papers

Paper I I participated in the planning, construction and operation of the pilot-scale plant. I also participated in sampling, and in the planning and execution of adsorption tests on laboratory scale. I analysed most of the results and wrote the paper with input from the other authors.

Paper II I participated in the planning, construction and operation of the pilot-scale plant. I also planned the experiments together with the other authors. I participated in sampling, performed most of the analysis, and planned and performed laboratory-scale experiments on bio-methane potential and oxygen uptake rate. I analysed the results and wrote the manuscript with contributions from the other authors.

Abbreviations and Symbols

BET	Brunauer-Emmett-Teller
BOD	Biological oxygen demand (mg L^{-1})
CEPT	Chemically enhanced primary treatment
COD	Chemical oxygen demand (mg L^{-1})
DMF	Direct membrane filtration
EDS	Energy-dispersive X-ray spectroscopy
DOC	Dissolved organic carbon (mg L^{-1})
GAC	Granular activated carbon
LOQ	Limit of quantification
Me^{3+}	Metal ion (III)
MF	Microfiltration
OUR	Oxygen uptake rate (mg O_2 L^{-1} h^{-1})
PAC	Powdered activated carbon
SEM	Scanning electron microscopy
SOUR	Specific oxygen uptake rate (mg O_2 g^{-1} VSS h^{-1})
SS	Suspended solids (mg L^{-1})
TMP	Transmembrane pressure (bar)
TOC	Total organic carbon (mg L^{-1})
TP	Total phosphorus (mg L^{-1})
UF	Ultrafiltration
VS	Volatile solids (mg L^{-1})
VSS	Volatile suspended solids (mg L^{-1})
WWTP	Wastewater treatment plant

Contents

1 Introduction

The management and treatment of municipal wastewater is necessary from both an environmental and health point of view. The original purpose was to prevent the spread of disease, but the aim of wastewater treatment today is also to prevent the eutrophication and pollution of receiving waters. The focus of modern wastewater treatment is to reduce the organic content and the amounts of nitrogen and phosphorus in wastewater, and to reduce eutrophication and oxygen depletion (Harremoës, 1998; la Cour Jansen et al., 2019), however, another group of pollutants has received increasing attention during recent years.

Organic micropollutants, such as pharmaceutical residues, are released into the aquatic environment via wastewater, where they can have negative effects, even at low concentrations (ng L^{-1} to µg L^{-1}) (Santos et al., 2010). Hence the name "micropollutants". Several studies have reported their limited removal in conventional wastewater treatment (Clara et al., 2005; Radjenović et al., 2009; Reemtsma et al., 2006; Reungoat et al., 2011; Ternes et al., 2004) and highlighted their presence in drinking water sources (Daneshvar et al., 2012; Kong et al., 2015; Tixier et al., 2003). The observed effects of pharmaceutical residues on aquatic organisms are intersex, feminization of males, behavioural changes and antibiotic resistance (Alan et al., 2008; Brodin et al., 2013; Brodin et al., 2017; Gagné et al., 2011; Qiao et al., 2018; Tetreault et al., 2011). Long-term effects and cocktail effects are also of considerable concern (Gosset et al., 2020; Kümmerer, 2009; Matić Bujagić et al., 2021; Pomati et al., 2007).

The removal of pharmaceutical residues by conventional wastewater treatment varies. Some pharmaceuticals, e.g., paracetamol and naproxen, are partly or completely degraded by activated sludge treatment (Lahti & Oikari, 2011; Radjenović et al., 2009). Others, such as diclofenac, can be degraded in biofilm processes (Falås et al., 2013), while others, such as carbamazepine, are resistant to biological degradation (Clara et al., 2005; Falås et al., 2013; Radjenović et al., 2009). Conventional wastewater treatment plants (WWTPs) must therefore be upgraded to increase the removal of organic micropollutants, including pharmaceutical residues.

Activated carbon is a highly porous material with a high surface area, and can be used to adsorb organic micropollutants. Adsorption studies in natural water and wastewater effluents have shown its potential to remove a wide variety of organic micropollutants (Boehler et al., 2012; Corwin & Summers, 2011; Fundneider et al., 2021; Joss et al., 2005; Kennedy et al., 2015; Ternes et al., 2002). Granular activated carbon (GAC) and powdered activated carbon (PAC) can both be used for this purpose. PAC has been shown to have a significantly higher adsorption capacity (Kårelid et al., 2017a; Real et al., 2017). However, GAC is gaining more attention as it can be reactivated and reused. The ability to reactivate GAC considerably reduces the carbon dioxide footprint of the process, making GAC a potentially better alternative than PAC (Böhler et al., 2022).

Another goal is to reduce the energy consumption of wastewater treatment. Many WWTPs use aerated activated sludge treatment to degrade and remove organic carbon; a process that generates carbon dioxide and consumes much energy (Lazarova et al., 2012). The need for aeration could be reduced, or even removed, by using advanced separation processes, allowing more of the organic carbon to be used for biogas production. Physicochemical treatment, such as chemically enhanced primary treatment (CEPT) and direct membrane filtration (DMF) are processes suitable for this purpose.

DMF involves the direct treatment of wastewater with a porous membrane (Ravazzini et al., 2005). Studies on DMF have shown its potential to treat wastewater and concentrate organic matter for enhanced biogas production, however, fouling is a major problem in such processes (Hube et al., 2020). Various combinations of anti-fouling techniques have been investigated with promising results (Hube et al., 2021; Kimura et al., 2021). However, most studies on DMF have been performed on laboratory scale, and studies are thus required on a larger scale for validation.

Another approach to reduce fouling during DMF is the use of pre-treatment, such as CEPT combined with microsieving. CEPT with microsieving has previously been suggested to be a net energy-producing process due to the high potential for biogas production (Remy et al., 2014). High retention of suspended solids (SS) and phosphorus is possible through optimized dosing of precipitation chemicals (Väänänen et al., 2016).

Pilot-scale tests using CEPT with microsieving and subsequent DMF (microfiltration) have shown stable DMF operation with high removal of chemical oxygen demand (COD) and phosphorus, low energy demand, and high biogas production potential (Hey et al., 2017, 2018). Organic micropollutants could also be removed by adding a further treatment step, such as adsorption by activated carbon.

1.1 Research objectives

The main research objective of the work presented in this dissertation was to investigate and evaluate the wastewater treatment concept of CEPT with microsieving and DMF followed by activated carbon adsorption for the removal of organic micropollutants. This treatment concept was investigated in terms of the removal of wastewater constituents, including organic micropollutants, biomethane production and membrane operation. The potential of supplementary biological treatment after microsieving and DMF was also investigated. The potential of organic micropollutant removal by GAC in tertiary treated wastewater was investigated and compared to removal by PAC, also in tertiary treated wastewater. Most of the investigations were performed on pilot scale, with complementary tests being performed on laboratory scale.

The specific objectives of this work were as follows.

- To find suitable precipitation chemicals to optimize CEPT.
- To investigate the influence of applying CEPT upstream of microsieving and DMF on treatment performance and membrane fouling.
- To determine the biomethane potential of the sludge from microsieving, and to evaluate the energy coverage for the treatment train (CEPT with microsieving and DMF).
- To determine the aeration required for supplementary biological treatment downstream of microsieving and DMF.
- To determine the potential of PAC for the adsorption of organic micropollutants downstream of CEPT with microsieving and DMF.
- To investigate the potential of using GAC filtration for the removal of organic micropollutants, compared to PAC, in tertiary treated wastewater.

1.2 Outline

This dissertation is based on two studies on CEPT with microsieving and DMF, together with additional results from coagulation and flocculation experiments. **Paper I** presents a study on the potential of adsorbing organic micropollutants by PAC after the physicochemical processes in the treatment train. **Paper II** presents the removal of wastewater constituents, biomethane production, membrane operation, possible supplementary biological treatment, and organic micropollutant removal by subsequent GAC filtration.

The basic principles of physicochemical wastewater treatment processes are described, as well as detailed investigations of the adsorption of specific organic micropollutants onto activated carbon. In addition, the potential and limitations of physicochemical wastewater treatment are discussed.

An introduction to the topic, including the potentially hazardous effects of organic micropollutants in the aquatic environment, is given in Chapter 1. The basic principles of physicochemical processes and their applicability in wastewater treatment are presented in Chapter 2. Chapter 3 describes the materials and methods used in this work. Chapter 4 presents the main results given in **Papers I** & **II**, together with the results of laboratory-scale experiments with precipitation chemicals. Concluding remarks and a synthesis of the work are given in Chapter 5. Finally, suggestions for future research can be found in Chapter 6.

2 Physicochemical Treatment

Physicochemical processes, such as chemical coagulation and flocculation, separation by sedimentation, size exclusion (sieving or membrane filtration) and adsorption, can be used for the treatment of municipal wastewater as a complement, or alternative, to biological treatment. However, this depends on the nature of the incoming wastewater and the demands on the outlet water. In contrast to biological treatment, physicochemical treatment can be carried out at low temperatures and is not affected by toxins in the wastewater, but is less suitable for the removal of nitrogen. The work presented in this dissertation is based on several physicochemical processes, the basic principles of which are described below.

2.1 Chemically enhanced primary treatment

CEPT involves the use of a coagulant and/or a flocculant aid before the primary treatment (traditionally sedimentation in clarifiers) to enhance the removal of SS and organic matter, which can be measured as total organic carbon (TOC), biological oxygen demand (BOD), or COD. Coagulation has been, and is, widely used in Sweden and Norway for pre- and post-precipitation to remove phosphorus (Henze & Ødegaard, 1994). Using a cationic polymer as the coagulant will have the greatest influence on SS and BOD removal, while metal salts can be used to remove more phosphorus (Väänänen, 2017).

The principle of coagulation in wastewater is based on the neutralisation of surface charges on particles and colloids in the water by adding a coagulant, such as a metal salt ($AlCl_3$, $AlSO_4$, $FeCl_3$ or $FeSO_4$), or a cationic polymer (Ødegaard, 1998). Neutral colloids aggregate into larger particles called flocs, increasing their settling velocity and enhancing separation in a clarifier.

If a metal salt is used as coagulant, the flocculation process can be aided by adding a polymer to the wastewater, after the addition of the coagulant. This leads to larger and more strongly bound flocs, which further increases the settling velocity and enables the use of other separation techniques. One example of a separation technique is microsieving, which requires strong flocs that will not break up during filtration (Ljunggren, 2006). Typical doses of coagulants and flocculants using CEPT and subsequent microsieving in disc and drum filters are about 1-7 mg polymer L^{-1} with 10-30 mg Fe^{3+} L^{-1} or 5-20 mg Al^{3+} L^{-1} (Ljunggren, 2006; Remy et al., 2014; Väänänen et al., 2016).

2.2 Microsieving in primary treatment

Microsieving involves filtration with fine mesh sieves with pore sizes down to ~10 µm, and can be applied in rotating drum sieves, disc sieves or belt sieves. Larger filter openings have also been used for primary treatment of wastewater to achieve high hydraulic capacity and a low area footprint (Ljunggren, 2006). A drum filter microsieve can replace primary clarifiers for the separation of SS and BOD on a lower surface area. A higher degree of removal can be achieved when CEPT is combined with microsieving (Ljunggren et al., 2007; Rusten & Ødegaard, 2006). The sludge is removed from the sieve by backwashing. A drum filter is illustrated in Figure 1.

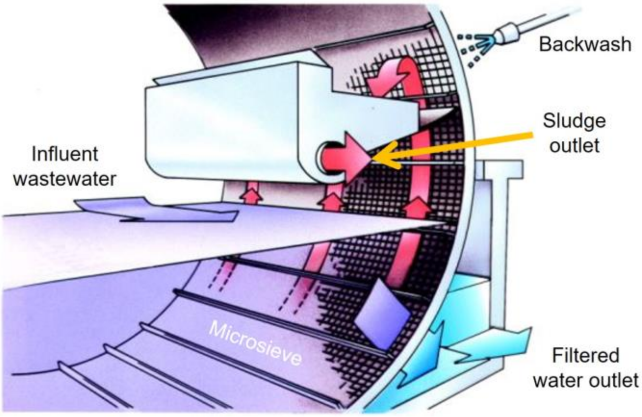


Figure 1. Illustration of drum filter microsieve configuration. Used with permission from Hydrotech.

2.3 Membrane filtration

Like microsieving, membrane filtration is also primarily based on separation by size exclusion. However, membrane filtration is performed on a much smaller scale, with smaller pores. The smallest pores are found in reverse osmosis membranes (<1 nm) followed by nanofiltration (< 2 nm), ultrafiltration (UF; 2-100 nm) and micro-filtration (MF; 0.1-2 µm) (Khulbe et al., 2008). The correct choice of membrane is very important and depends on the composition of the feed and the objective of the treatment.

The liquid that passes through the membrane is called the permeate, and the remaining liquid is called the concentrate or retentate (Figure 2). The transport of liquid through the membrane is referred to as the flux, which is the rate of flow divided by the area of the membrane (L m^{-2} h^{-1}). A high flux is desirable to achieve a high treatment capacity, but too high a flux can cause severe fouling of the membrane, leading to a decline in flux. Membrane filtration is commonly driven by the pressure difference between the feed and permeate side of the membrane, and is called the transmembrane pressure (TMP). The filtration capacity of a membrane can also be referred to as the permeability, which is the flux divided by the TMP.

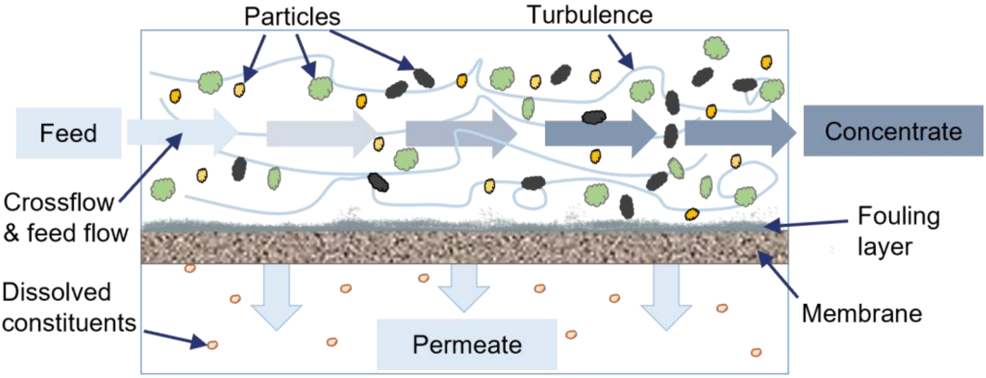


Figure 2. A simplified illustration of membrane filtration using crossflow.

As mentioned, the membrane can become fouled during filtration. There are different types of fouling, such as pore blocking, adsorption, and the formation of a cake layer on the membrane surface. Adsorption and pore blocking can cause irreversible fouling which is more difficult to remove than reversible fouling in the form of a cake layer. Fouling is commonly reduced having a crossflow along the membrane surface (Choi et al., 2005), but other techniques, such as periodic membrane cleaning, backwashing, operation optimization and pre-treatment of the feed, can also be used (Tijing et al., 2015). If the membrane is severely fouled, chemical cleaning is required.

2.3.1 Direct membrane filtration

DMF is described as the treatment of wastewater by a porous membrane without preceding biological treatment (Nieuwenhuijzen et al., 2000), and therefore differs from that in a membrane bioreactor. The organic matter in wastewater can be concentrated using DMF, and then anaerobically digested to produce methane. DMF has been described as treatment with a potentially low energy demand (Kimura et al., 2021; Ravazzini et al., 2005; Zhao et al., 2019), and it has been suggested that this form of treatment has the potential to be net energy positive (Hey et al., 2018; Kimura et al., 2021; Nascimento et al., 2017).

Previous studies on DMF have shown that fouling is the main problem (Hube et al., 2020). It is therefore important to reduce fouling in order to maintain an adequate flux and thus reap the economic benefits of the process. Chemical pre-treatment, gas sparging, chemically enhanced backwashing and vibrating modules have been studied as means of mitigating fouling in DMF. Typical fluxes and TMPs have been reported to be 4 to 20 L m^{-2} h^{-1} and 30 to 800 mbar (Hey et al., 2018; Kimura et al., 2021; Lateef et al., 2013; Nascimento et al., 2017).

Membrane filtration has also been suggested for the removal of organic micropollutants (Abtahi et al., 2018; Chon et al., 2012; Khanzada et al., 2020; Urtiaga et al., 2013), however, nanofiltration and reverse osmosis are better suited for this. In most applications of DMF for wastewater treatment, either MF or UF has been used (Hube et al., 2020), to maintain an adequate flux. Some degree of retention has been reported using UF, which was related to the adsorption of micropollutants in membrane pores (Khanzada et al., 2020).

2.4 Activated carbon adsorption

Activated carbon is a porous material with a high surface area per unit mass, and has long been used in various applications, for example, to purify water or air, and for medical purposes (Çeçen & Aktaş, 2011). In the present work, only adsorption from liquids was considered. The principle of adsorption involves the interaction of a compound with a boundary layer (e.g., the graphene layer of an activated carbon), which results in a change in phase of the compound (Dąbrowski, 2001). In this way, compounds dissolved in a liquid can be removed from the liquid as they are adsorbed onto the surface of the activated carbon. One advantage of activated carbon is its ability to adsorb a wide range of compounds, such as pharmaceutical residues and other organic micropollutants. The porosity of activated carbon is classified into: micropores (< 2 nm), mesopores (2-50 nm) and macropores (> 50 nm) (Sing et al., 1985). Micropores are the most dominant pores in terms of surface area, and are the most important for the adsorption of organic micropollutants (Radovic, 2001). A simplified illustration of the surface of a grain of activated carbon, and an adsorbed micropollutant, is shown in Figure 3.

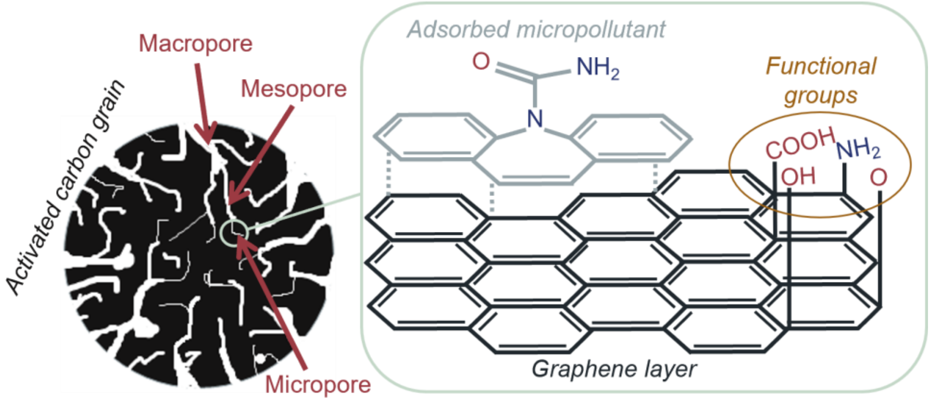


Figure 3. A simplified illustration of a grain of activated carbon, its surface and an adsorbed organic micropollutant.

The adsorption process is complex and is governed by several factors. The interactions between the adsorbent (activated carbon) and the adsorbate (compound) can be electrostatic, π-π (a type of dispersion force) or hydrophobic (Moreno-Castilla, 2004). Electrostatic interactions occur due to the charges on the adsorbent and adsorbate. This is dependent on the molecular structure of the adsorbent and the adsorbate, and the pH of the solution in which adsorption takes place. The surface charge of an activated carbon is highly influenced by its functional groups, such as $-COOH$, $-NH_2$, $-O$ and $-OH$, and their presence and distribution are dependent on the way in which the carbon is activated. If the adsorbate and adsorbent have opposite charges, electrostatic attraction will enhance adsorption. Functional groups also influence π-π interactions between the adsorbent and the adsorbate. For example, phenolic groups release electrons to the graphene layers, while carboxylic groups withdraw electrons. Hydrophobic interactions are governed by the repulsive forces between hydrophobic compounds and water molecules. Therefore, the more hydrophobic a compound is, the more easily it is adsorbed and removed from a water solution. (Moreno-Castilla, 2004).

Commercially available activated carbon can be manufactured from a variety of materials, including charcoal, wood, peat and coconut shells (Çeçen & Aktaş, 2011). It is important to consider the source of carbon bearing in mind climate impact, as well as the possibility of reactivating the carbon after it has been used. This is only possible with GAC, and not PAC. The carbon is activated by an activation process (e.g., thermal or chemical) which is important to get a large specific surface area, commonly measure as the Brunauer-Emmett-Teller surface area (Aktaş & Çeçen, 2006). A large specific surface area is further closely related to the performance of activated carbon in micropollutant removal (Mailler et al., 2016).

2.4.1 Adsorption of organic micropollutants from wastewater

The adsorption of organic micropollutants differs depending on their chemical properties. It has been shown that hydrophobic micropollutants (such as carbamazepine and diuron) and positively charged micropollutants (such as atenolol and metoprolol) are more easily adsorbed than hydrophilic and negatively charged ones (de Ridder et al., 2011; Guillossou et al., 2020; Kovalova et al., 2013). To remove a compound that is hard to adsorb, such as sulfamethoxazole (negatively charged) will thus require more activated carbon than to remove on that is easy to adsorb, such as metoprolol.

The negative influence of dissolved organic matter on the adsorption of organic micropollutants in wastewater effluents has been recognized (Aschermann et al., 2018; Boehler et al., 2012; Guillossou et al., 2020; Zietzschmann et al., 2014). Dissolved organic matter, commonly measured as dissolved organic carbon (DOC), adsorbs on the activated carbon, or blocks the carbon pores, making the adsorption sites unavailable for micropollutants. To reduce the influence of DOC, it is therefore better to employ adsorption as the final step in the treatment train, where most of the DOC will have been removed in the preceding steps (Boehler et al., 2012). Both powdered and granular activated carbon can be used for wastewater treatment.

2.4.2 Powdered activated carbon

PAC has particle sizes on the µm-scale and can be added as a slurry at a WWTP. One or several mixed contact reactors can be used to ensure sufficient time for the compounds to be adsorbed (Figure 4) (Margot et al., 2013). The PAC can be recirculated back to the biological treatment step for longer contact times, and thus greater removal. Doses of about 10-30 mg L^{-1} in secondary and final WWTP effluents have been shown to be sufficient to achieve high degrees of removal of organic micropollutants, depending on whether the PAC is recirculated or not (Kårelid et al., 2017b; Meinel et al., 2016). If PAC is added in the final step at a WWTP, subsequent separation is required to retain the PAC.

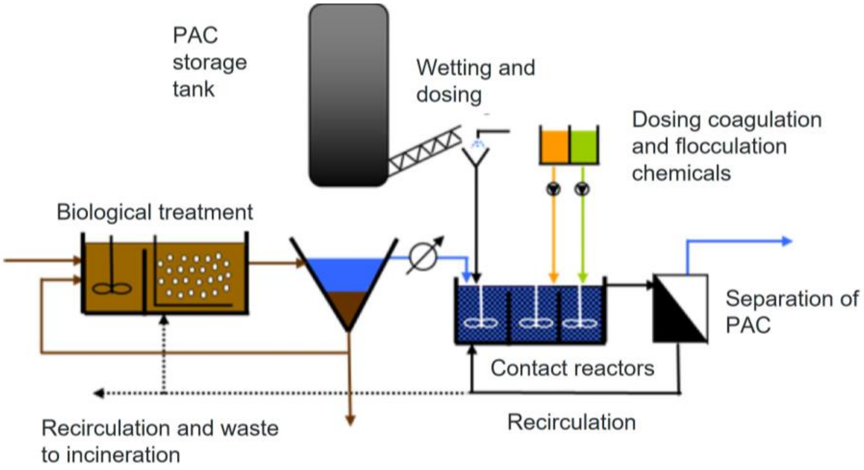


Figure 4. Example of a configuration for dosing PAC at a WWTP. (Adapted from Abegglen & Siegrist, 2012). Used with permission from Prof. Siegrist.

It is also possible to add PAC directly to the biological reactor at a WWTP and still achieve adequate micropollutant removal (Cimbritz et al., 2019; Streicher et al., 2016). This configuration required very little additional space at the WWTP. However, the PAC will be removed together with the sludge, which restricts the use of the sludge for agricultural purposes.

2.4.3 Granular activated carbon

Activated carbon is also available as granules (GAC); the grains usually being 0.2-5 mm in size (Çeçen & Aktaş, 2011). GAC can be used as a filter bed, or in a packed column, through which the wastewater can be filtered, as illustrated in Figure 5. During operation, pressure will build up in the filter and regular backwashing is needed to maintain an adequate filtering capacity. As wastewater is filtered through the filter bed, micropollutants and other DOC are adsorbed, and the GAC eventually becomes exhausted. When this happens, micropollutants will start to break through the filter and it must be regenerated or replaced with new GAC.

Comparisons of activated carbon have shown that PAC has a higher adsorption capacity and faster adsorption kinetics than GAC (Kårelid et al., 2017a; Meinel et al., 2015; Real et al., 2017); the latter can be related to the higher fraction of mesopores in PAC (Real et al., 2017; Suzuki, 1991). Because of this, GAC has economically been less attractive than PAC for removal of micropollutants. However, the possibility of reactivating GAC for reuse makes removal by GAC comparable to that of PAC (Böhler et al., 2022). Moreover, there are indications that biofilm growth in a GAC filter can degrade some of the adsorbed micropollutants, which may extend the time before GAC becomes saturated (Betsholtz et al., 2021; Sbardella et al., 2018).

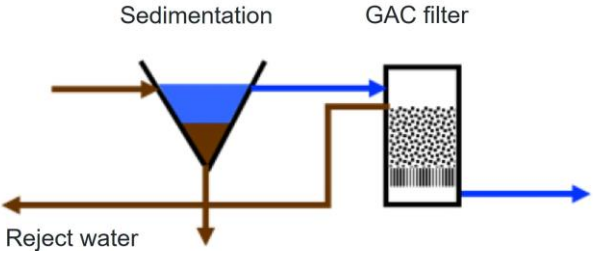


Figure 5. Example of a configuration for GAC filtration at a WWTP. (Adapted from Abegglen & Siegrist, 2012). Used with permission from Prof. Siegrist.

Several factors influence the degree of removal of organic micropollutants by GAC filtration. The number of bed volumes treated in the filter is one of the most important as it is a measure of the load of micropollutants and DOC on the filter. Poorly adsorbing compounds, such as gabapentin, will start to break through the filter earlier than those that adsorb well, such as carbamazepine. The number of bed volumes that can be treated before breakthrough occurs depends on the process parameters, the type of GAC used, the characteristics of the wastewater, and the specific compound considered (Benstoem et al., 2017). In a meta-analysis of 44 studies, Benstoem et al. found that a 20% breakthrough of diclofenac was reached after 800-20,000 bed volumes, which could be explained by a broad range of process parameters, types of GAC used and the different wastewater matrices used in the studies.

One important operational parameter is the empty bed contact time, which is the time taken for the wastewater to pass through an empty filter bed or column. A filter time of 20-30 min has been reported to be appropriate for the adsorption of micropollutants (Böhler et al., 2022; Fundneider et al., 2021).

It is also important to consider pre-treatment to reduce the load of DOC and SS on the GAC filter. As mentioned above (Section 2.4.1), DOC has a negative impact on the adsorption of organic micropollutants on activated carbon, and as the GAC filter retains particles, a high SS load would result in the need for excessive backwashing of the filter (Bornemann et al., 2012).

3 Methods

This chapter briefly describes the methods used in the studies presented in **Papers I** & **II** and in additional precipitation experiments. The first introductory part concerns some background to why some experiments were conducted in pilot scale, while others were performed in laboratory scale.

The treatment train of CEPT with microsieving, DMF and GAC is a new concept that could have the potential of being implanted on full scale. CEPT with microsieving, and GAC filtration have separately been tested in large scale. However, most studies on DMF have been conducted on laboratory scale for relatively short periods, or with synthetic wastewater (Hube et al., 2020). DMF has the potential to treat wastewater on a large scale, but more studies are required on pilot scale with real wastewater to verify the stability of operation and performance. Therefore, the work described in this dissertation was primarily based on experiments conducted on pilot scale at a municipal full-scale WWTP (Svedala, Sweden) where real fresh incoming wastewater was continuously supplied.

The performance of CEPT with microsieving and DMF was evaluated by measuring the removal of SS, TOC and total phosphorus (TP). Flux and TMP data from DMF were used, together with the recovery of permeability after membrane cleaning, to evaluate the membrane operation. The biomethane potential of the sludge from the microsieve was used to calculate the energy balance.

As a proof of concept, GAC filtration of the membrane permeate was also performed on pilot scale. Twenty-one organic micropollutants were chosen for analysis based on the EU watch list and compounds detected in recipients in southern Sweden (Gomez Cortes et al., 2020; Svahn & Björklund, 2017). The organic micropollutants were measured in samples collected upstream of, in between, and downstream of, the pilot scale plant. Unlike many previous studies on the removal of organic micropollutants, the wastewater in this work was not spiked; measurements were made of the actual levels of organic micropollutants.

Prior to the pilot-scale experiments, different precipitation chemicals and doses were tested on laboratory scale for CEPT with microsieving. Adsorption experiments were also performed on laboratory scale with PAC and water samples from the pilot-scale plant, enabling measurements with many PAC doses at fixed concentrations of organic micropollutants in the water samples.

3.1 Laboratory-scale experiments

3.1.1 Coagulation and flocculation

Prior to the experiments on pilot scale, different coagulants and flocculants were tested in the laboratory to determine the best combination of chemicals for good floc formation. The objective was to create large flocs that were strong enough to not break up in the microsieving step. Fresh wastewater was collected as a grab sample after the sand trap at Svedala WWTP. Two Al-based coagulants (PAX-XL100 and PAX-XL60) and one Fe-based coagulant (PIX-111) (Kemira, Sweden) were investigated using jar tests, with chemical doses from 3.3 to 13.2 mg Fe^{3+} or Al^{3+} L^{-1} together with 2 mg L^{-1} of an anionic polyacrylamide polymer (A150; Kemira, Sweden). The tests were performed in a program-controlled flocculator (Flocculator 2000, Kemira). The flocs were separated on a woven microfilter cloth with openings of 100 µm. Photographs of the jar tests and subsequent filtration are shown in Figure 6.

Figure 6. Jar tests with Al- and Fe-based coagulants and anionic polymer (left) and the separation of flocs on a microfilter cloth (right).

The flocculation procedure was as follows.

1. Primary wastewater was poured into 1 L beakers.
2. Rapid mixing (200 rpm) for 10 s.
3. Coagulant was added during the first 3 s of rapid mixing in Step 2.
4. Slow mixing (40 rpm) for 2 min.

5. Rapid mixing (200 rpm) for 10 s.
6. Polymer was added during the first 3 s of rapid mixing in Step 5.
7. Slow mixing (40 rpm) for 6 min.
8. The flocculated wastewater was poured over a cloth with 100 µm pores to separate the flocs from the water.

The procedure was repeated for each of the coagulants and the result of flocculation was evaluated by measuring the turbidity (NTU; Nephelometric Turbidity Units) of the filtered water using a turbidimeter (HACH 2100P, Germany). The coagulant giving the lowest turbidity was used in further experiments for choosing a flocculant.

The best coagulant was tested with 6 polyacrylamide polymers (N100, A100, A110, A120, A150, Hydrex 6161) (Kemira, Sweden and Veolia, France) where one was non-ionic (N100) and the rest were anionic with low to high charge densities. The polymer doses were increased to 2-4 mg L^{-1} based on higher dosing used in previous studies (Hey et al., 2017; Väänänen et al., 2016). The same flocculation procedure was used as above, and turbidity was again used for evaluating the results. The three best-performing polymers were then tested on pilot scale (see Section 3.2.1).

3.1.2 Adsorption using PAC

The adsorption of organic micropollutants in different process streams from the pilot-scale plant and the WWTP by PAC was investigated (**Paper I**). Wastewater was collected as grab samples after microsieving, MF, UF and after tertiary treatment at the WWTP. PAC (SAE Super, Norit, Netherlands) was prepared as a suspended stock solution in deionized water.

Wastewater was transferred into Falcon tubes (50 mL) and PAC was added at 12 concentrations (0, 5, 10, 15, 20, 25, 30, 40, 50, 60, 80, 100 mg L^{-1}). After 24 h of agitation on a horizontal shaker at 20°C, the PAC was separated from the solution by centrifugation and subsequent filtration (0.45 µm). The doses and agitation time were based on the results of previous studies with PAC (Meinel et al., 2016). DOC and ultraviolet absorbance at 254 nm (UVA_{254}) were determined within 24 h. Filtered samples for micropollutant analysis were stored at −20°C.

3.2 The pilot-scale plant

A pilot-scale treatment plant was built and operated periodically at Svedala WWTP (Figure 7), Sweden, for approximately 10 months. The plant consisted of two parts. The first, larger part, received wastewater from the sand trap at the WWTP. This was treated with CEPT, microsieving, MF and UF (parallel filtration), and GAC filtration after UF (lower part of Figure 7). A photograph of the first part of the pilot plant is shown in Figure 8. The second part received tertiary (biologically and chemically) treated wastewater from the WWTP, and consisted of a column for sand filtration followed by a similar column for GAC filtration (upper, right part of Figure 7).

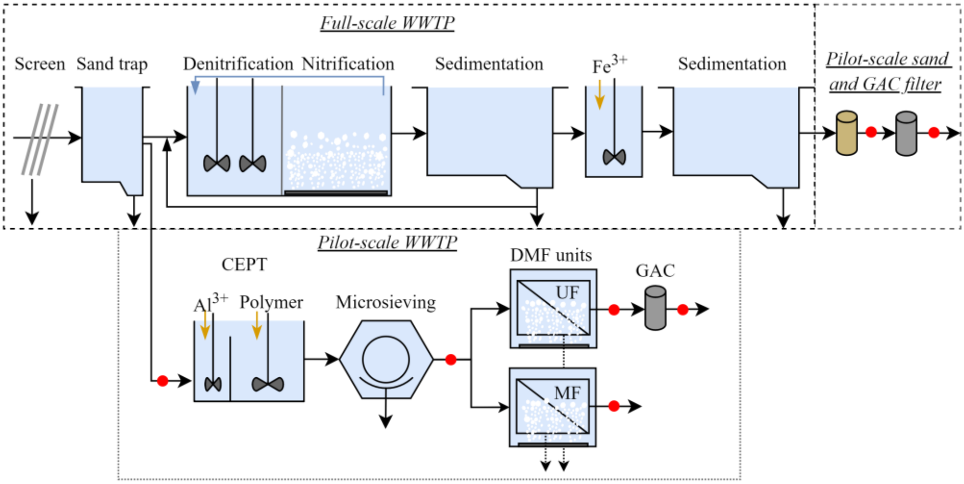


Figure 7. A simplified process scheme of the Svedala WWTP and pilot-scale treatment plant. Red dots indicate sampling locations.

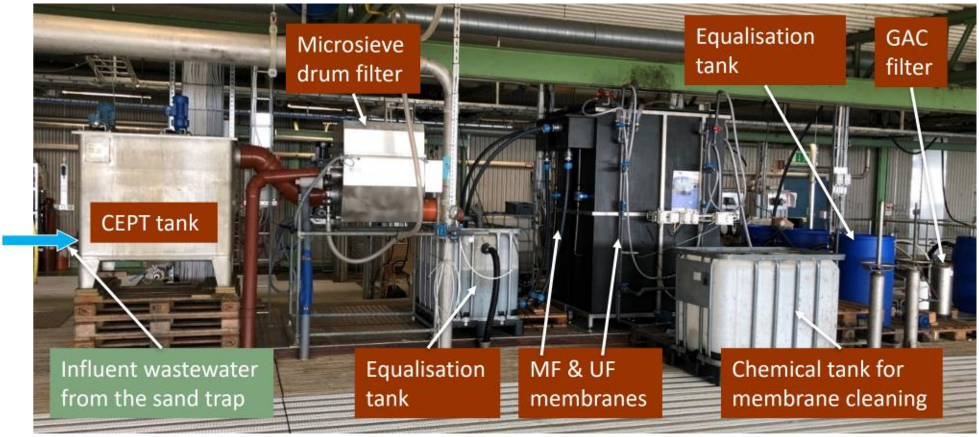


Figure 8. The pilot-scale treatment plant employing physicochemical processes at Svedala WWTP.

3.2.1 Initial tests and operation of the pilot-scale plant

After the tests using different coagulants and flocculants on laboratory scale (see Section 3.1.1) further tests were performed on pilot scale using PAX-XL60 and the three best polymers (A110, A120, Hydrex 6161) found in the laboratory tests. These pilot-scale tests were conducted as the flocs were likely exposed to greater shear forces in the subsequent microsieving on pilot scale than in laboratory scale. Wastewater (2.7-3.0 m^3 h^{-1}) was pumped from the sand trap to the CEPT tank into which the precipitation chemicals were dosed into two stirred compartments (200 L for coagulation and 800 L for flocculation) (Figure 9, left side) using peristaltic pumps (Prominent, Germany). Ten mg Al^{3+} L^{-1} coagulant and 3 mg L^{-1} polymer were used and subsequent microsieving was performed in a drum filter (Hydrotech, Sweden) with a woven fabric with openings of 100 µm (Figure 9, right side). Each polymer was tested for 2-4 h. Wastewater was collected as grab samples after microsieving, and the turbidity and SS were analysed. The best polymer was chosen for further experiments.

Figure 9. Coagulation and flocculation in the CEPT tank (left) and the drum filter used for microsieving (right).

After further testing and optimizing the doses of precipitation chemicals, the dose of coagulant was slightly increased and CEPT was performed with 12 mg Al^{3+} L^{-1} (PAX-XL60) and 3 mg L^{-1} polymer (Hydrex 6161, Veolia, France). After the chemicals and doses for the CEPT had been decided, longer experiments, up to 5 days, were conducted and subsequent DMF was applied. The influence of applying CEPT upstream of microsieving was investigated in terms of treatment, and filtration capacity in DMF. The maximum capacity of the microsieve was not determined. A capacity of 32 m^3 m^{-2} h^{-1} with CEPT and 40 µm openings has been reported previously (Ljunggren et al., 2007).

Parallel membrane filtration using UF and MF membranes (UFX-10pHt and MFP2, Alfa Laval, Denmark, 40 m^2 each) that were submerged in tanks (Figure 10). The tanks were aerated from the bottom to induce a crossflow across the membrane surfaces and reduce fouling. Wastewater was drawn through the membranes using pumps on the permeate side. Fouling was reduced by periodically turning the pumps on and off. Membrane filtration was monitored, and data were collected for the flux, TMP, wastewater temperature and pH. Due to operational failure, MF was only included in the experiments presented in **Paper I**. The UF permeate was further filtered in a 19 L GAC column (Figure 11). The empty bed contact time in the GAC filter was kept at 10 min by maintaining the flow at 114 L h^{-1}.

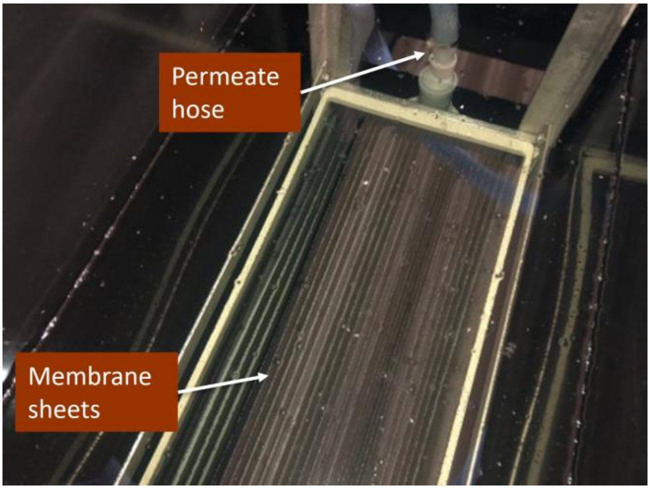


Figure 10. The flat sheet UF membrane unit submerged in tap water.

3.2.2 Operation of pilot-scale sand and GAC filters

Pilot-scale sand and GAC filters (19 L each) (Figure 11) were connected in series with tertiary effluent as feed, as shown in Figures 7. The operational parameters for the filters were similar to the ones for the GAC filter downstream of UF; the empty bed contact time was kept at 10 min for both filters by maintaining a flow of 114 L h^{-1}.

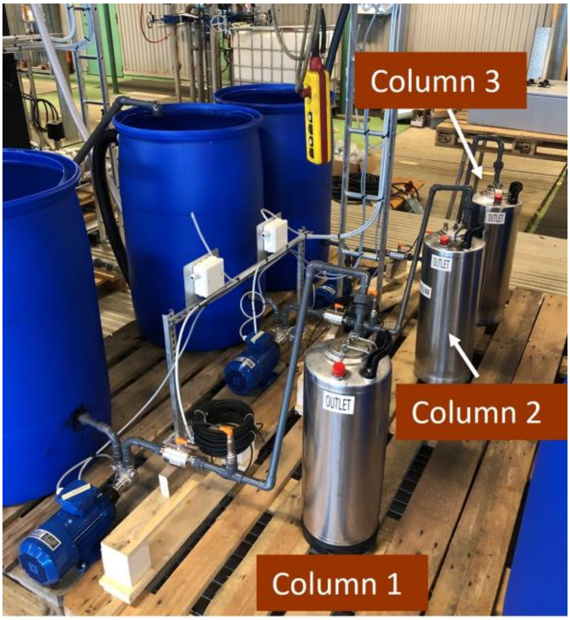


Figure 11. Columns containing sand and GAC. Column 1 contained GAC for treatment downstream of CEPT, microsieving and DMF (UF). Column 2 contained sand and column 3 contained GAC. Column 2 and 3 were connected in series (sand first) for treatment downstream of the WWTP.

3.2.3 Sampling

Grab and 24 h composite samples were collected for the analysis of organic micropollutants and other wastewater constituents. Samples to be analysed for organic micropollutants were stored at −20°C in high-density polyethylene bottles until analysed at MoLab at Kristianstad University, Sweden. Samples for other analyses were either taken directly to a laboratory at Lund University for analysis, or stored at 4°C for analysis within 24 h.

3.3 Analytical methods

3.3.1 Organic micropollutants

Samples were prepared using solid phase extraction followed by elution and reconstitution. Aliquots of 1-10 µL per sample were analysed using ultra-performance liquid chromatography (UPLC) followed by tandem mass spectrometry (MS/MS) (Waters Acquity UPLC H-Class, Xevo TQS Waters Micromass, Manchester, UK) using three different methods at different pH values (Svahn & Björklund, 2016, 2019). A list of the organic micropollutants together with their limit of quantification and relative standard deviations is given in the Appendix (Table A1).

The concentrations of the organic micropollutants were used to calculate the removal by PAC and GAC, and the adsorption onto PAC. The number of bed volumes put through the GAC filter, and the average incoming DOC concentration were used to calculate the average load of DOC on the GAC. This was used to compare the removal by PAC and GAC.

Freundlich isotherms (Eq. 1) were used to evaluate the results from the PAC experiment (see Section 3.1.2) and to compare the adsorption onto PAC from the wastewater after the different processes:

$$q_e = K_F * C_e^{1/n} \qquad \text{(Eq. 1)}$$

where:

q_e is the adsorbed concentration of organic micropollutants on the activated carbon (ng mg^{-1}),

K_F is the Freundlich constant (ng mg^{-1})(ng L^{-1})$^{-1/n}$,

C_e is the organic micropollutant concentration at equilibrium (ng L^{-1}) and

$1/n$ is the Freundlich intensity parameter (dimensionless).

3.3.2 Nutrients, organic carbon and particles

Total nitrogen, TP, COD and TOC were analysed using Hach cuvettes and a spectrometer (Hach DR2800, Germany). A UV-vis spectrometer (Hach DR6000, Germany) was used to measure UVA_{254}. The concentrations of SS and volatile SS (VSS) were determined according to the European standard (EN 872). The total solids concentration was determined by drying 40-50 mL of mixed sample at 105°C for 12-24 h and weighing. The sample was then ignited at 550°C for 1 h and then weighed again to determine the concentration of VSS.

3.3.3 Oxygen uptake rate

Supplementary treatment of the physicochemically treated wastewater was investigated with the aim of reducing the remaining organic content after CEPT, microsieving and DMF. The aeration required for a biological process was determined by measuring the oxygen uptake rate (OUR; a measure of the biodegradability of the wastewater) in the wastewater leaving the pilot plant (**Paper II**).

Grab samples collected at the pilot plant were mixed with activated sludge from Svedala WWTP, allylthiourea (a nitrification inhibitor) and put in separate (1.2 L) reactors that were connected to the OUR equipment (BM-Advance Respirometer, Surcis, S.L., Spain). The reactors were stirred and aerated periodically, and the dissolved oxygen was monitored. The oxygen uptake was measured between the periods of aeration, and the OUR was calculated by the software from the equipment provider. The experiments were run for ~3 h for each sample. To allow comparison of the results, the OUR results were normalised to the concentration of VSS in the reactors, and denoted the specific OUR (SOUR). The aeration required for each wastewater sample was calculated as the accumulative oxygen uptake from the start of the experiment until the OUR reached the target reference value.

3.3.4 Biomethane potential

The carbon-rich sludge from the microsieve was used as substrate in tests to determine the biomethane potential by anaerobic digestion (**Paper II**). Grab samples of sludge from the microsieve and waste activate sludge from the WWTP were mixed separately with sludge from a mesophilic anaerobic digester (Klagshamn WWTP, Sweden) in tightly sealed 500 mL glass jars. The jars were placed in a water bath (37°C) and were periodically stirred. The biogas produced was led via a carbon dioxide trap (sodium hydroxide solution) to an AMPTS II system (Bioprocess Control, Sweden). The flow of methane was measured continuously over 30 days.

3.3.5 Membrane operation

Operational data from DMF was used to evaluate the influence of applying CEPT upstream of microsieving on the membrane permeability. Tree cycles of operation were performed: (1) DMF of raw wastewater (no CEPT before microsieving), (2) and (3) DMF with CEPT before microsieving. The volumetric flux was calculated as the permeate flow per unit membrane area (Eq. 2), and was further used to calculate the flux normalised to 15°C of the membrane (Eq. 3) as described by Hey et al. (2017):

$$J_V = \frac{Q_P}{A_M} \qquad \text{(Eq. 2)}$$

$$J_{\text{standard}} = J_{\text{measured}} * \frac{(42.5+T_{\text{standard}})^{1.5}}{(42.5+T_{\text{measured}})^{1.5}} \qquad \text{(Eq. 3)}$$

where:

J_{measured} is the volumetric flux (L m^{-2} h^{-1}),

Q_P is the measured permeate flow (L h^{-1}),

A_M is the membrane area (m^2),

J_{standard} is the standardised volumetric flux (L m^{-2} h^{-1}),

T_{standard} is the standard temperature (15°C) and

T_{measured} is the measured temperature (°C).

The TMP was calculated as the average pressure difference between the feed and permeate side (Eq. 4). The permeability normalised to 15°C was calculated according to (Eq. 5):

$$TMP = P_F - P_P \qquad \text{(Eq. 4)}$$

$$\text{Permeability}_{\text{standard}} = \frac{J_{\text{standard}}}{TMP} \qquad \text{(Eq. 5)}$$

where:

TMP is the transmembrane pressure (bar),

P_F is the measured pressure on the feed side (bar),

P_P is the measured pressure on the permeate side (bar) and

$\text{Permeability}_{\text{standard}}$ is the standardised permeability (L m^{-2} h^{-1} bar^{-1}).

3.3.6 Analysis of membranes after filtration and cleaning

After filtration and cleaning on pilot scale, the UF membrane was analysed in the laboratory to investigate fouling. Pieces of the membrane were stored for 6 months in tap water containing 45 mg sodium bisulphite L^{-1} to prevent microbial growth (4°C), until analysis. A piece of a pristine UF membrane (washed with sodium hydroxide) was used as a reference sample. Before analysis, the pieces of membrane were air dried, coated with a thin layer of gold and palladium, and then examined with scanning electron micro-scopy (SEM) and energy-dispersive X-ray spectroscopy (EDS).

4 Results and Discussion

This chapter describes the findings and results of the laboratory- and pilot-scale studies. The first sections deal with the optimization of, and treatment with, CEPT and microsieving with different chemicals and doses in the laboratory. Similar tests were then performed on pilot scale. The influence of applying CEPT upstream of microsieving on DMF is described based on operational performance up to 4 days at a time. The removal of SS, TOC and TP was investigated and results of a brief investigation of membrane fouling are presented. Thereafter, the potential of using supplementary biological treatment is described, followed by the presentation of an energy balance of the treatment concept. Finally, the results on the removal of organic micropollutants by PAC and GAC are presented.

4.1 CEPT and microsieving on laboratory scale

Jar tests with three coagulants (PAX-XL100, PAX-XL60, PIX-111) and one polymer (A150), followed by microsieving, were performed on laboratory scale. It was found that higher doses of coagulant resulted in lower turbidity of the microsieve filtrate (Figure 12). Furthermore, the two Al-based coagulants (PAX-XL100, PAX-XL60) reduced the turbidity more than the Fe-based one (PIX-111), as also reported by Remy et al. (2014). The best performing coagulant (PAX-XL60, Kemira, Sweden) was therefore used for the following experiments.

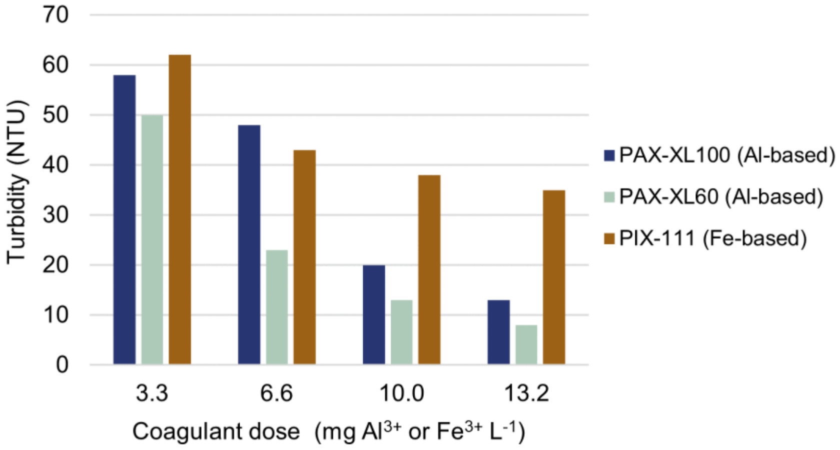


Figure 12. Turbidity after coagulation, flocculation and microsieving. Coagulation and flocculation were done using jar tests with four different doses of three coagulants and 2 mg polymer (A150) L^{-1}. The turbidity and SS concentration of the untreated wastewater were 274 NTU and 394 mg L^{-1}, respectively.

After identifying the best coagulant, 6 polymers (N100, A100, A110, A120, A150, Hydrex 6161) were investigated to determine which gave the best separation of flocs by microsieving. Two doses (10 and 15 mg Al^{3+} L^{-1}) of coagulant and 3 doses (2, 3 and 4 mg L^{-1}) of polymer were tested. The results are shown in Figure 13.

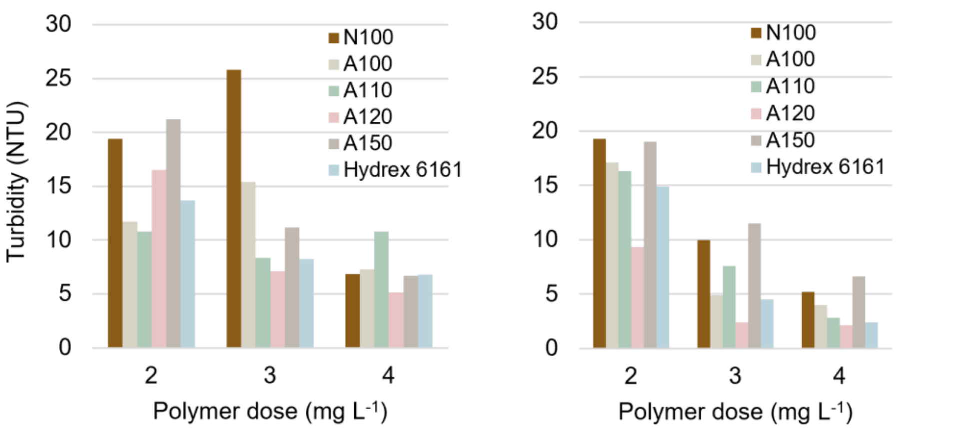


Figure 13. Turbidity after coagulation, flocculation and microsieving. Coagulation and flocculation were done using jar tests with 10 (left) and 15 (right) mg Al^{3+} L^{-1} (PAX-XL60) with three different doses of six different polymers. The turbidity and SS concentration of the raw wastewater samples were 110 NTU and 193 mg L^{-1}, respectively (left), and 139 NTU and 225 mg L^{-1}, respectively (right).

The three best-performing polymers were found to be A110, A120 and Hydrex 6161 (Figure 13). All three were anionic polymers with low- or medium charge density, and all had high molecular weights. The lowest observed turbidity after microsieving was 2.1 NTU, corresponding to a 98% reduction in turbidity, using 15 mg Al^{3+} L^{-1} and 4 mg A120 L^{-1}. The difference in turbidity in the filtrates decreased with increasing polymer dose. However, high dosing leads to higher costs, and the risk of residual polymer in the filtrate, which may foul the membranes in subsequent DMF. A dose of 3 mg L^{-1} polymer was thus used in the following experiments, so as to maintain adequate performance without overdosing. Cationic polymers can also be used to achieve high retention, however, they may also have a negative effect on subsequent membrane filtration (Hey, 2016).

4.2 Treatment performance on pilot scale

The main results of this dissertation come from experiments on pilot scale. After the investigation of CEPT on laboratory scale, experiments were started at the pilot plant. More experiments with different polymers were performed and the treatment performance by CEPT, microsieving and DMF was investigated.

4.2.1 Investigating precipitation chemicals

The final stage in identifying the best polymer was to investigate the ones that performed best in the laboratory-scale experiments, on pilot scale. The pilot-scale set-up differed from that in the laboratory, mainly in that microsieving was performed in a rotating drum filter on pilot scale, and the flocs are exposed to greater turbulence than in the laboratory. Chemical dosing of 10 mg Al^{3+} L^{-1} and 3 mg L^{-1} polymer was used. Both SS and turbidity was analysed to evaluate the performance. The results are shown in Figure 14.

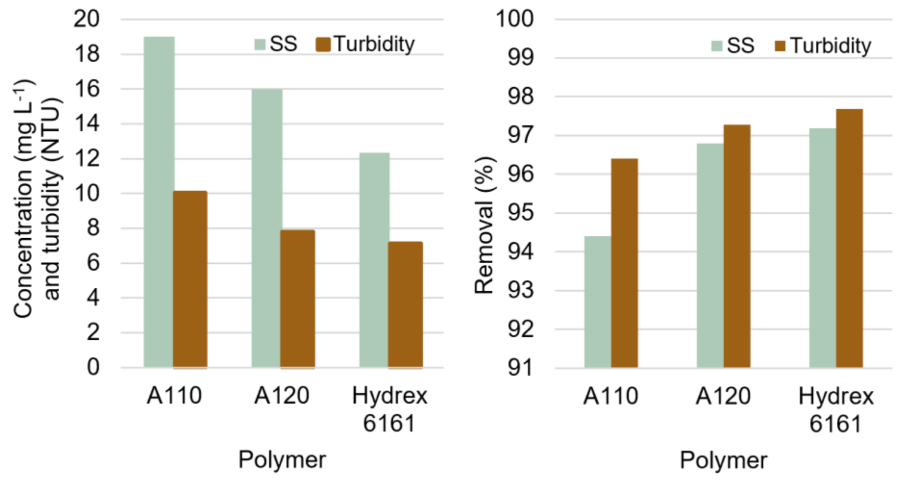


Figure 14. Concentration of SS and turbidity (left) and corresponding removal (right) after coagulation, flocculation and microsieving on pilot scale. Chemical dosing: 10 mg Al^{3+} L^{-1} and 3 mg polymer L^{-1}. The concentration of SS in the influent wastewater varied between 340 and 500 mg L^{-1} and the turbidity between 380 and 410 NTU.

The lowest turbidity (7 NTU) and concentration of SS (12 mg L^{-1}) in the microsieve filtrate was observed when using Hydrex 6161 (Figure 14). The second best-performing polymer was A120, which had given the best results in the laboratory experiments (7 NTU) using similar doses (10 mg Al^{3+} L^{-1} and 3 mg polymer L^{-1}). Because of the great reduction in both SS and turbidity (> 97%), Hydrex 6161 was chosen for further experiments in pilot scale together with PAX-XL60 as a coagulant.

4.2.2 Treatment by CEPT, microsieving and DMF

Further treatment with (and without) CEPT with microsieving and DMF was evaluated over periods up to 4 days, including the analysis of SS, TOC and TP (**Paper II**). DMF without CEPT upstream of microsieving was investigated to determine the removal that could be achieved with DMF only, and how much CEPT improved treatment with DMF (Figure 15). Treatment with DMF could only be evaluated with UF due to the operational failure of MF.

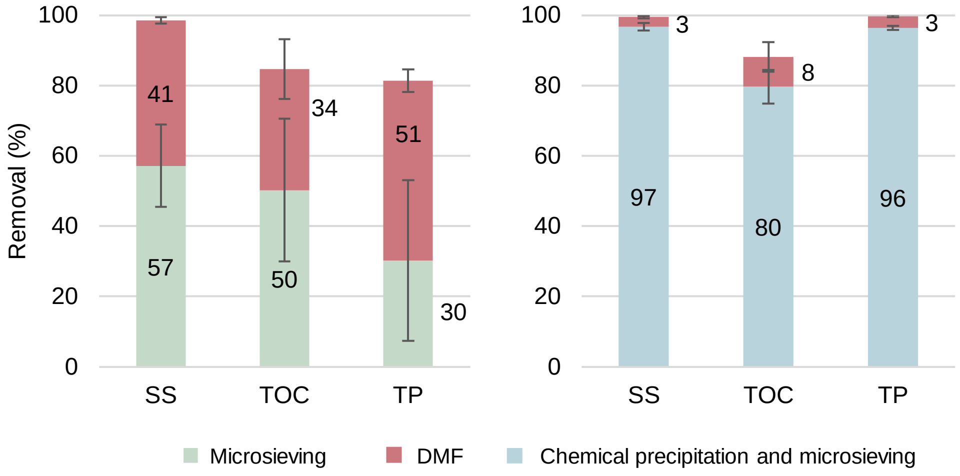


Figure 15. The performance of treatment without (left) and with (right) CEPT before microsieving and subsequent DMF. The error bars represent the standard deviation calculated from the data (n = 5-8).

Treatment with only microsieving and DMF (no CEPT) provided relatively high removal of SS (98%), TOC (84%) and TP (81%) (Figure 15, left); the average concentrations in the membrane permeate being 6, 25 and 1.9 mg L^{-1}, respectively. The presence of SS in the permeate could be a sign of leakage, but since the removal was relatively high, it was deemed that the source of the particles was release from the biofilm in pipes downstream of the membrane tank. The removal of DOC by DMF was on average 35%, which implies that most of the DOC was smaller than the pores of the membrane. It should be noted that the performance of this kind of process is dependent on the amount of constituents dissolved in the incoming raw wastewater, which may vary (Levine et al., 1985, 1991).

Treatment with CEPT before microsieving and DMF improved the performance, especially regarding TP (Figure 15, right). CEPT and microsieving reached almost the same degree of removal of SS and TOC as did microsieving and DMF (without CEPT). The concentration of TP was on average 0.4 mg L^{-1} after microsieving. The average concentrations of SS, TOC and TP after DMF were 3, 22 and 0.07 mg L^{-1}, respectively. The removal of DOC by CEPT and microsieving was on average 43%, and when including DMF, it increased to 50%. This implies that CEPT enabled greater removal of DOC than UF.

The removal of total nitrogen was low (35%) but was higher than expected. Mostly particle-bound nitrogen would be removed in this type of treatment.

4.3 Membrane operation

CEPT improved the separation of organic material by microsieving, and thus also reduced the load on the UF membrane (**Paper II**). The probability of heavy fouling will be reduced with a lower load of organics, and it will be easier to maintain an adequate flux. However, polymer residues (used in CEPT) in the microsieve filtrate could also potentially foul the membrane (Zhang et al., 2020). To evaluate the influence of CEPT upstream of microsieving on DMF performance, the flux and TMP were recorded (Figure 16) during operation using the control software, and used to calculate the permeability (Figure 17). Three cycles were performed; two with CEPT and one without CEPT.

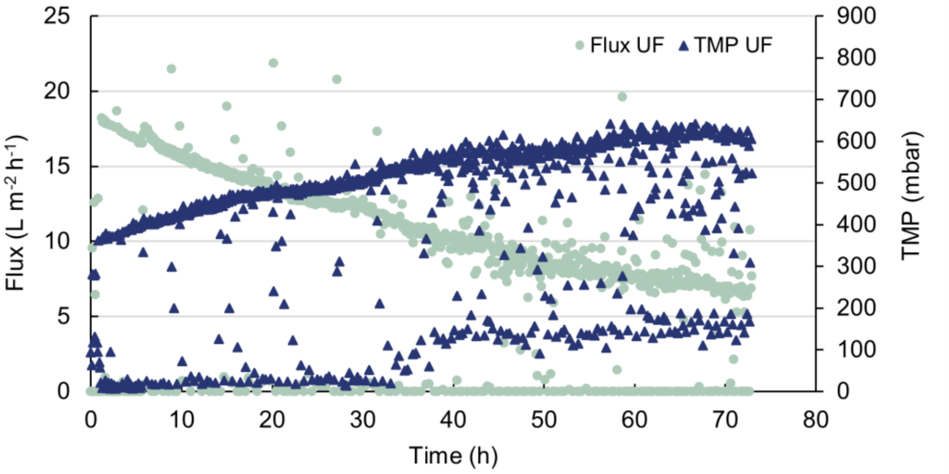


Figure 16. Flux and TMP from one cycle of DMF (UF) with CEPT applied upstream of microsieving.

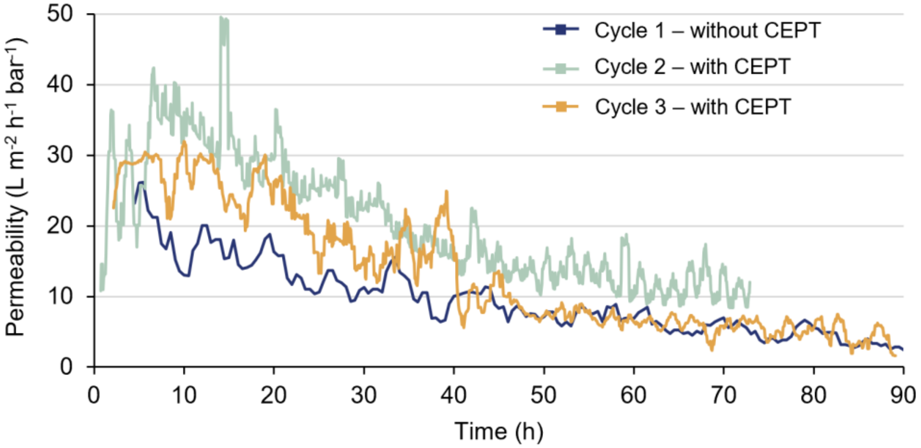


Figure 17. DMF membrane permeability from three cycles of membrane operation. One without CEPT and two with CEPT before microsieving.

The intention was to maintain a constant flux while varying the TMP, as the permeate pump was set at a fixed flow rate. However, the TMP increased and the flux decreased during operation (Figure 16). This was observed during all cycles of operation, and a maximum of 90 h of operation was achieved before the TMP threshold was reached (650-800 mbar). Comparing the permeability during the three cycles (Figure 17) showed that employing CEPT before microsieving (Cycles 2 & 3) generally resulted in a higher permeability than without CEPT (Cycle 1), at least during the first 40 h of filtration.

Both the flux and TMP were within the range of values reported in previous studies (Hey et al., 2018; Kimura et al., 2021; Lateef et al., 2013; Nascimento et al., 2017). Hey et al. (2017), who also used CEPT and microsieving as pre-treatment prior to DMF (MF), observed a stable standardised flux of 6.1 L m^{-2} h^{-1} at 30 mbar, corresponding to a permeability of 203 L m^{-2} h^{-1} bar^{-1}. This permeability is significantly higher that the values obtained in the present work, which is partly due to that they used MF while we used UF. Low fluxes are preferable to reduce fouling, but this also results in a lower filtration capacity. Hey et al. (2017) did not report any specific fouling problems, which could be due to the low flux and TMP, and cleaning with hydrogen peroxide was successful. Fouling was more serious in the present work, as described below.

4.3.1 Membrane fouling and cleaning

The DMF membrane was cleaned after use according to the manufacturer's protocol, which included hydrogen peroxide (12 g m^{-2}) at high pH (for organic fouling) and citric acid (100 g m^{-2}) at low pH (for inorganic fouling), however, the membrane's permeability could not be restored (< 70% recovery). Furthermore, the results (**Paper II**) showed that cleaning after Cycle 2, with only hydrogen peroxide restored the permeability to 89%, however, a high chemical dose of hydrogen peroxide (36 g L^{-1}), membrane soaking for approximately 48 h and backwashing with chemicals were necessary. Since no citric acid was necessary to clean the membrane, it was deemed that fouling was mostly organic. Others have reported almost complete restoration of membrane permeability after cleaning (Kimura et al., 2017; Lateef et al., 2013) using either physical or chemical cleaning methods.

Membrane cleaning was performed after Cycle 3 with hydrogen peroxide (24 g m^{-2}) and citric acid (200 g m^{-2}), which resulted in a much lower recovery of the permeability (54%). Only chemical backwashing was performed with no soaking. The reason for the heavier fouling after Cycle 3 could, however, not be identified. It is possible that the type of cleaning (chemical backwashing) or the chemicals used were not suitable. It is also possible that some flocs broke in the microsieve, and/or that polymer residues were adsorbed on the membrane surface, or that a change in the influent wastewater composition resulted in more severe fouling, such as pore blocking.

The UF membrane used for DMF was investigated in the laboratory using SEM and EDS. From the SEM images, it appeared that, after cleaning, the used UF membrane was cleaner than the pristine membrane (Figure 18). The surface of the pristine membrane appeared to be rough, which could be due to sodium crystals from washing with sodium hydroxide, or dust accumulated during storage. The used and cleaned UF membrane appeared smoother with a few irregularities, which could be fouling or dust. It is possible that there was a remaining fouling layer after cleaning that was so thin that it could not be detected, or that it was altered during sample preparation (air drying and coating with gold and palladium). It is also possible that the pores of the used and cleaned membrane were blocked, but this could not be detected in this analysis.

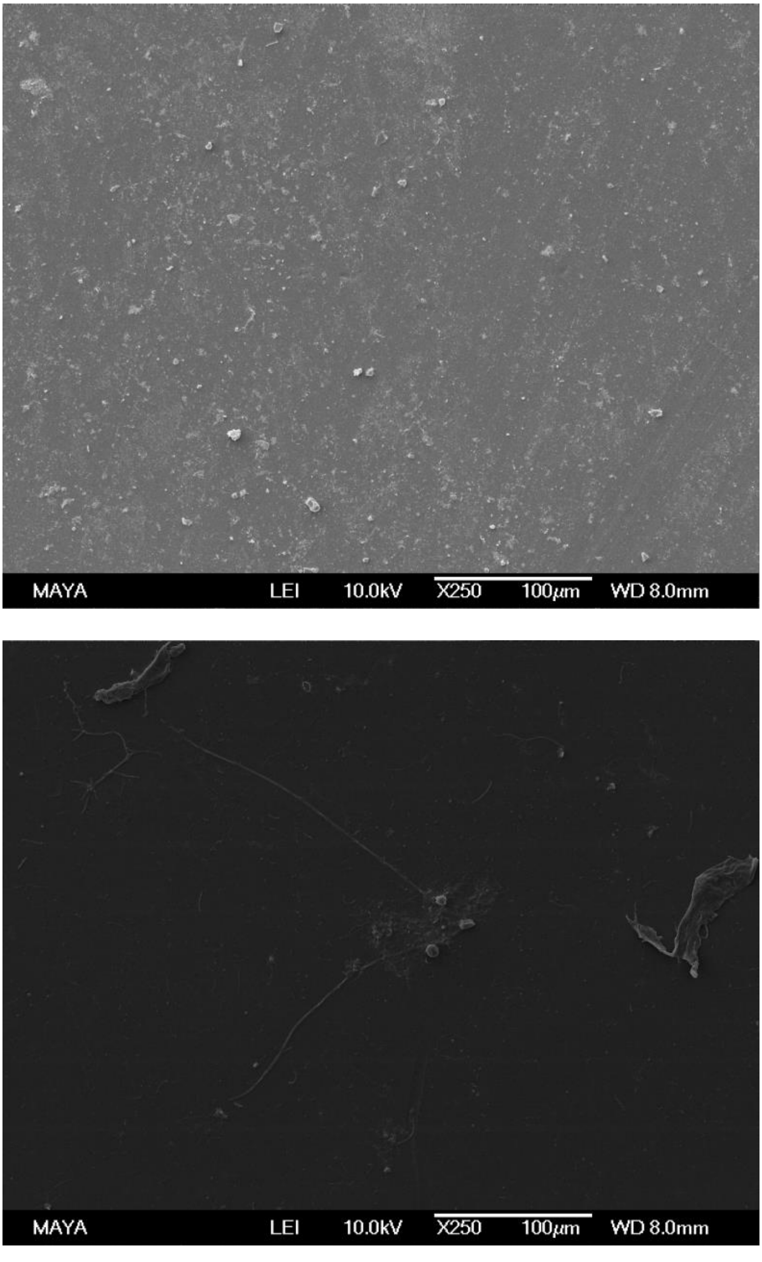


Figure 18. Two screen shots from SEM showing the pristine membrane (upper part of the figure) and the used UF membrane (lower part of the figure) after cleaning.

The EDS analysis showed that carbon was the most common element on the surface of both the pristine, and the used, cleaned UF membrane samples (Figure 19). The carbon could be from organic fouling and/or the membrane material, but because of the similar elemental composition of the pristine membrane, there was no significant evidence of fouling on the membrane surface. The main difference between the two membranes seemed to be the peak of sodium on the pristine membrane which could be remains from washing with sodium hydroxide. Nonetheless, the membrane could have been fouled internally. No indication of inorganic fouling was identified in the EDS analysis.

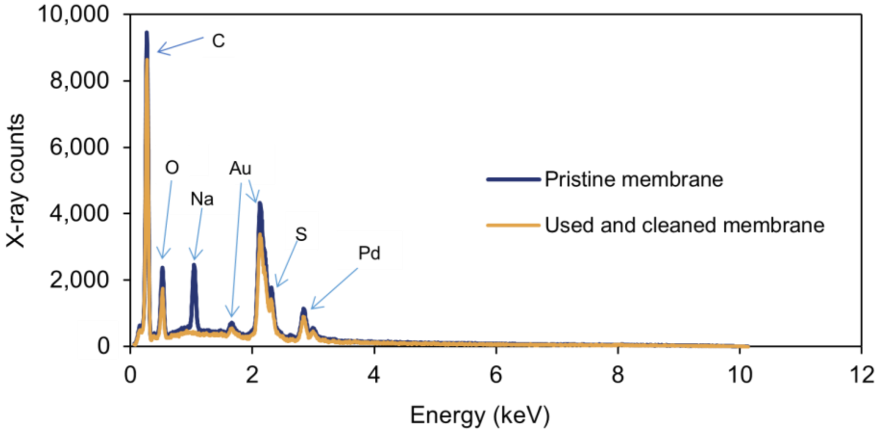
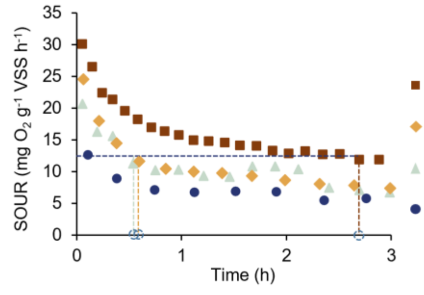


Figure 19. Elemental analysis of the surface of a piece of pristine UF membrane, and a used and cleaned UF membrane using EDS. Gold and palladium were used to prepare the membrane for SEM.

Since cleaning at the pilot plant seemed inadequate, a laboratory-scale study was conducted to further investigate fouling and cleaning in DMF. This study showed that hydrogen peroxide was a relatively poor cleaning agent compared to a commercial cleaning agent (Ultrasil-10, Ecolab, UK). The use of suitable cleaning agents is thus important to restore permeability and enable long-term operation of DMF. The laboratory study resulted in a Master's dissertation entitled *Direct Membrane Filtration – Investigating Fouling and Antifouling in Wastewater Treatment*.

4.4 Oxygen uptake rate

Further treatment could be used to reduce the organic content remaining after DMF, for example, an aerated biological process. Information on the DOC degradability and the aeration required was obtained by measuring SOUR in wastewater samples (**Paper II**). The results of these experiments can be found in Figure 20.



	$SOUR_{ini}$ (mg O_2 g VSS^{-1} h^{-1})	DOC_{ini} (mg L^{-1})	$SOUR_{ini}$/ DOC_{ini}	O_2 uptake (mg O_2 L^{-1})
■ Influent	30	31	1.0	34
◆ Microsieve filtrate	25	25	1.0	8
▲ DMF permeate	21	18	1.4	7
● WWTP effluent	13	14	0.9	-

Figure 20. SOUR in the wastewater samples from the pilot-scale plant (left). The blue dashed line indicates the initial value of SOUR in the WWTP effluent and was used as a reference. Vertical dashed lines indicate the times at which the initial SOUR of the WWTP effluent was reached. The table on the right gives the initial SOUR values ($SOUR_{ini}$), initial DOC concentrations (DOC_{ini}), a measure of DOC degradability ($SOUR_{ini}/DOC_{ini}$) and the oxygen uptake for each sample.

The highest initial value of SOUR was observed in the influent wastewater sample (collected after the sand trap at the WWTP and entering the pilot-scale plant; Figure 7), followed by the microsieve filtrate, DMF permeate and lastly the WWTP effluent. The initial DOC concentrations in the wastewater samples followed the same order. A measure of the biodegradability of the DOC was obtained by dividing the initial SOUR value by the initial DOC concentration. The highest value of this ratio, 1.4, was found in the DMF permeate, which indicates that a high fraction of the DOC was readily biodegradable. The corresponding value in the microsieve filtrate was 1.0.

The oxygen uptake in the DMF permeate (7 mg O_2 L^{-1}) was 21% of that in the influent (34 mg O_2 L^{-1}). The microsieve filtrate a had an oxygen uptake (8 mg O_2 L^{-1}), which was similar to that in the DMF permeate and corresponded to 24% of that in the influent. Conclusively, it was possible to reduce the need for aeration by more than 75% using CEPT with microsieving, while subsequent DMF led to only a small further reduction in the oxygen uptake.

4.5 Biomethane potential and energy balance

The measured biomethane potential of the sludge removed by the microsieve (with CEPT applied upstream) was, on average, 0.34 Nm^3 kg^{-1} volatile solids (VS) (**Paper II**), which is similar to the results presented by Hey et al. (2018). The corresponding value for the waste-activated sludge from the WWTP was almost as high (0.31 Nm^3 kg^{-1} VS) and was expected to be lower. The WWTP in Svedala does not have a primary clarifier (Figure 7), and thus a fraction of the activated sludge consists of primary sludge, which contributes to the higher biomethane potential.

The electricity consumption of a physicochemical treatment train using CEPT, microsieving and DMF has been estimated to be 0.55 kWh m^{-3} treated wastewater, including preliminary treatment, chemical dosing and sludge treatment (Hey et al., 2017). Based on this value, a biomethane production of 0.34 Nm^3 kg^{-1} VS and the mass balance calculated over the microsieve (**Paper II**), the potential electricity coverage was found to be 61%. Much of the energy in this kind of treatment train would probably be consumed by the membrane operation. In the present studies on pilot scale, where the energy consumption was high (in total 3.7 kWh m^{-3} permeate), the UF membrane consumed more than 50% of the electricity. Hey et al. (2018) reported that their MF unit consumed about 70% of the electricity used by the whole treatment train. The electricity consumption of the membrane unit could potentially be reduced by using intermittent aeration and vibrating membrane modules, instead of continuous aeration (Kimura et al., 2021). However, there are to date no scientific reports on large-scale tests with such applications.

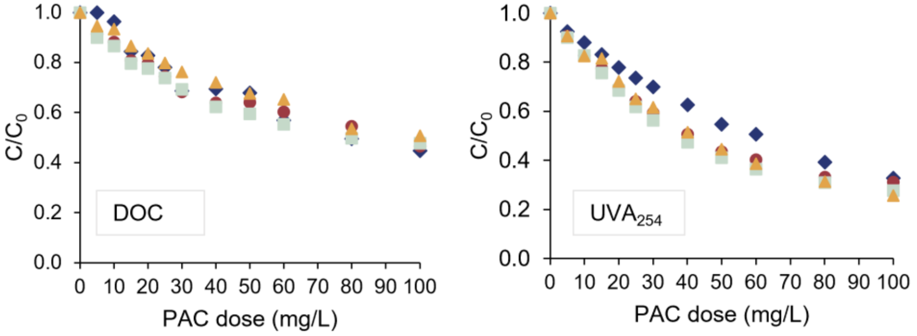
4.6 Adsorption onto PAC

Experiments were conducted with PAC to study the adsorption of compounds in the wastewater streams after physicochemical treatment, and to compare it to adsorption in tertiary (biologically and chemically) treated wastewater.

4.6.1 Removal of DOC and UVA_{254} following PAC adsorption

As expected, adding PAC to the four different wastewater samples from the pilot plant and downstream of tertiary treatment resulted in a reduction in DOC concentration and the UVA_{254} signal (Figure 21). The UVA_{254} signal was generally reduced to a higher extent in the microsieve filtrate and in the permeates from UF and MF, than in the tertiary effluent. The relative reductions in DOC concentrations were similar in the different samples, with somewhat higher degrees of removal in the UF and MF permeate. The lower degree of removal in the microsieve filtrate, despite its initially higher DOC concentration (C_0), could be due to adsorption and pore blocking by larger DOC compounds, or DOC that prevented smaller DOC compounds from adsorbing onto the PAC.

The absolute reduction in DOC concentration was in general lower in the sample from tertiary treatment than in the other samples, which indicates that the DOC in this wastewater sample had a lower affinity for PAC. Biological treatment can change the character of the DOC (Wang & Chen, 2018), which could partly explain why the DOC was removed to a lower extent by PAC in the tertiary effluent, than in the other samples.



Reference values (C_0)	▲ Microsieve filtrate	● MF permeate	■ UF permeate	◆ Tertiary effluent
DOC (mg L^{-1})	16.4	12.4	11.9	10.7
UVA_{254} (m^{-1})	29.0	21.8	20.7	18.9

Figure 21. Relative reduction in DOC and UVA_{254} signal in laboratory PAC experiments and initial values of DOC and UVA_{254}.

4.6.2 Adsorption isotherms for organic micropollutants

The removal of organic micropollutants by CEPT with microsieving and DMF was investigated on pilot scale, however, this treatment had only a limited effect on the micropollutants. Ciprofloxacin, ketoconazole and sertraline were the only micropollutants that were removed (> 80%) (**Paper II**), and their removal could be related to their ability to adsorb to sludge, as supported by sorption coefficients reported in previous studies (Hörsing et al., 2011; Ternes et al., 2004). Additional treatment, such as activated carbon adsorption, is thus required for the adequate removal of a larger number of organic micropollutants.

The adsorption of 12 organic micropollutants onto PAC was therefore investigated (**Paper I**). Adsorption isotherms for four organic micropollutants (metoprolol, carbamazepine, diclofenac and sulfamethoxazole) are shown in Figure 22.

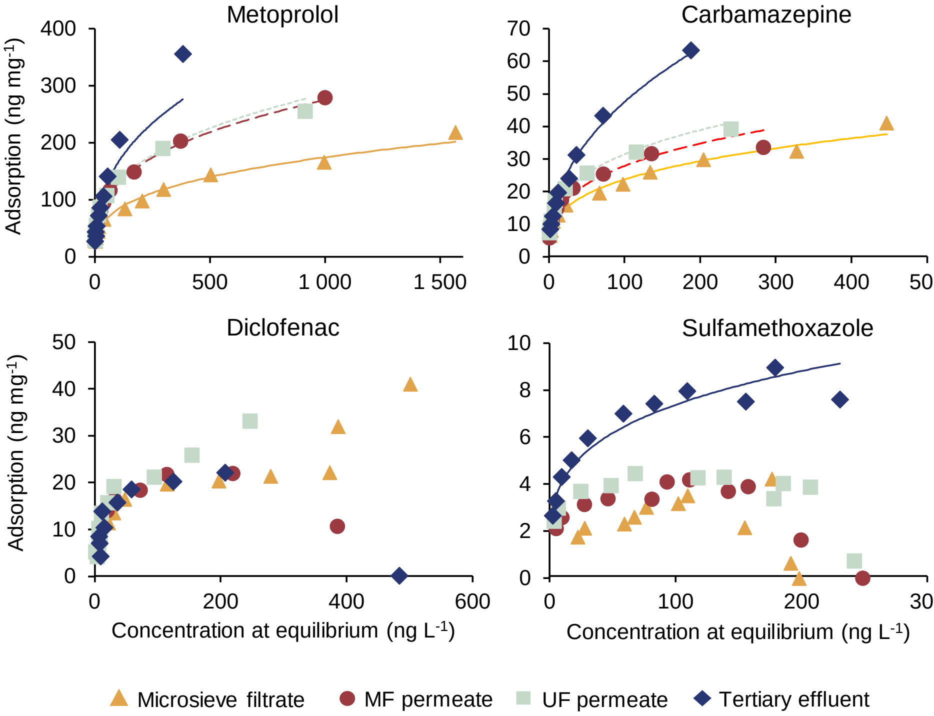


Figure 22. Adsorption of four organic micropollutants onto PAC in wastewater samples from the physicochemical pilot-scale plant, and downstream of tertiary treatment at the WWTP. Freundlich isotherms are shown for good fits ($R^2 \geq 0.90$). Note the different scales on the axes.

The adsorption of these four organic micropollutants was generally higher in the tertiary effluent than in the other samples (Figure 22). The higher adsorption of compounds in the tertiary effluent was related to both the lower concentration and the lower adsorption of DOC in this sample (Figure 21). The higher concentrations of DOC in the other three samples may have resulted in increased competition for adsorption sites, and thus lower adsorption of the organic micropollutants, as reported in previous studies (Altmann et al., 2014; Zietzschmann et al., 2014). The lowest adsorption was generally observed in the case of the microsieve filtrate, however, the adsorption of compounds in the membrane permeates was sometimes equally low, despite the fact that they had undergone more extensive treatment. One possible explanation is that low-molecular-weight DOC is more prone to adsorb onto PAC than high-molecular-weight DOC, as reported by Zietzschmann et al. (2014), and the membrane permeates contained low-molecular-weight DOC.

Isotherms could be fitted ($R^2 \geq 0.90$) to most of the data from the adsorption experiments with PAC (Figure 22). However, for the organic micropollutants with a lower affinity to activated carbon, such as diclofenac and sulfamethoxazole, isotherms could only be fitted to the data from some of the samples (e.g., sulfamethoxazole in the tertiary effluent). The lack of fitted isotherms for some samples was due to negative trends in adsorption at higher equilibrium concentrations. The difference in the affinity of organic micropollutants to PAC is related to their molecular properties, as discussed in **Paper I**. The most important properties were found to be hydrophobicity and charge, as reported in previous studies (de Ridder et al., 2011; Guillossou et al., 2020).

4.7 Organic micropollutant removal by PAC and GAC

The removal of organic micropollutants from the tertiary effluent sample by PAC was compared with the removal by GAC filtration on pilot scale downstream of tertiary treatment on full scale at the WWTP (Figure 23) (**Paper I**). The adsorption experiment by PAC was carried out by adding PAC to the sample, whereas GAC filtration was performed during approximately 6 months, corresponding to a hydraulic load of 23,000 bed volumes and an average DOC load of 0.83 g per g GAC. The load of DOC on the GAC was inversed to get a dose of GAC normalized per weight DOC, which could be compared with a specific dose of PAC. The removal of organic micropollutants by PAC and GAC could then be compared using a similar dose (1.2 g activated carbon g^{-1} DOC). The removal by GAC was calculated as the weighted average between a water load of 0 and 23,000 bed volumes.

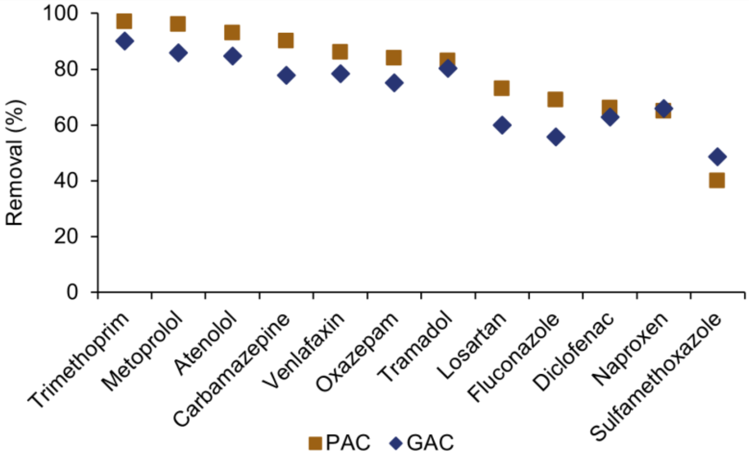


Figure 23. Removal of organic micropollutants by PAC and GAC in tertiary treated wastewater, with a dose of 1.2 g activated carbon per g DOC. Removal by GAC was based on the weighted mean micropollutant removal during the GAC process from 0 to 23,000 bed volumes.

The removal of micropollutants was high with both types of carbon. The slightly higher degree of removal by PAC was expected, as this has been reported in previous studies (Kårelid et al., 2017a; Meinel et al., 2015; Real et al., 2017). Furthermore, the order of removal of the organic micropollutants was similar for PAC and GAC. The removal of positively charged compounds, such as trimethoprim and atenolol, was usually higher than for negatively charged ones, such as naproxen and sulfamethoxazole.

The two types of carbon used in this study had similar Brunauer-Emmett-Teller surface areas, which should give rise to similar adsorption capacities. However, the distribution of pores also influences the adsorption mechanism. Real et al. (2017) found that the lower fractions of mesopore in GAC hindered the diffusion of the targeted substances into the micropores, reducing adsorption. Nevertheless, the degree of removal using PAC and GAC in the present work was more similar than expected based on previous studies, indicating high potential of using both GAC and PAC for the removal of organic micropollutants. The possibility of reactivating an exhausted GAC filter could give GAC an advantage over PAC, as suggested in Böhler et al. (2022).

4.8 GAC filtration after DMF

Removal of organic micropollutants by GAC filtration downstream of CEPT with microsieving and DMF (UF) was investigated on pilot scale (**Paper II**). Due to the relatively rapid fouling of the membranes (see Section 4.3), the GAC filter could only be operated for about 4 days (440 bed volumes). Organic micropollutant removal was compared with that in a GAC filter operated downstream of the tertiary treatment at Svedala WWTP. The results are given in Table 1.

Table 1. Concentrations and removal of organic micropollutants by GAC after CEPT with microsieving and DMF and after tertiary (biological and chemical) wastewater treatment. The compounds are sorted according to concentration after DMF.

	Concentration (ng L^{-1})					Removal (%)	
	After DMF	After DMF & GAC filtration	After tertiary treatment	After tertiary treatment & GAC filtration	LOQ*	With GAC filtration after DMF	With GAC filtration after tertiary treatment
Paracetamol	37,319	57	<LOQ	<LOQ	2.0	>99	-
Ibuprofen	15,987	225	1,127	<LOQ	100	99	>99
Naproxen	7,417	17	678	24	10	>99	96
Atenolol	2,535	<LOQ	1,445	13	2.0	>99	99
Metoprolol	1,577	<LOQ	2,017	26	2.0	>99	99
Diclofenac	1,402	10	1,296	25	2.0	99	98
Losartan	913	22	1,349	57	1.0	98	96
Venlafaxine	752	5.5	640	7.5	1.0	99	99
Carbamazepine	614	2.0	643	8.9	0.5	>99	99
Tramadol	529	3.0	909	8.1	1.0	99	99
Fluconazole	288	0.3	60	4.0	0.3	>99	93
Ciprofloxacin	246	1.9	<LOQ	<LOQ	10	99	-
Trimethoprim	242	<LOQ	132	1.3	1.0	>99	99
Citalopram	191	2.8	360	1.2	1.0	99	>99
Sulfamethoxazole	164	<LOQ	240	4.7	2.0	>99	98
Propranolol	142	<LOQ	76	<LOQ	2.0	>99	99
Oxazepam	109	1.0	493	8.7	1.0	99	98
Ketoconazole	73	<LOQ	14	<LOQ	10	>99	>99
Imidacloprid	36	<LOQ	12	<LOQ	2.0	>99	98
Estrone	34	<LOQ	29	<LOQ	0.2	>99	>99
Sertraline	10	<LOQ	34	<LOQ	0.5	>99	>99

*LOQ: limit of quantification.

The concentrations of organic micropollutants were generally higher after CEPT with microsieving and DMF than after full-scale treatment at the WWTP. Pharmaceuticals such as paracetamol, ibuprofen and naproxen are known to be biodegradable (Joss et al., 2006), and were removed in the activated sludge treatment at the WWTP. In contrast, the degree of removal was low following CEPT with microsieving and DMF (**Paper II**).

Removal by GAC filtration was high both downstream of DMF and after tertiary treatment (≥ 93%). Both GAC filters had been operated for approximately 4 days and the throughputs were relatively low (~500 bed volumes). High removal was therefore expected as almost all of the adsorption sites on the GAC were still available. During this period of operation, the average concentration of DOC entering the GAC filter after DMF was 28 mg L^{-1} and the removal of DOC by GAC was on average 65%. The amount of DOC adsorbed was calculated using the reduction in DOC and the volume put through the filter, giving a value of 25 mg DOC g^{-1} GAC. The concentration and the calculated amount of DOC adsorbed on the GAC filter following tertiary treatment were 11.9 mg L^{-1} and 15 mg DOC g^{-1} GAC, respectively. The removal of DOC was 66%. A continuous high load of DOC on the GAC downstream of DMF would probably result in more rapid saturation of the GAC.

The initial removal of organic micropollutants by GAC after CEPT with microsieving and DMF demonstrates the potential of this type of treatment for removing organic micropollutants. The main challenge in this study was to reduce fouling and maintain stable membrane filtration. Longer durations of operation are required for a thorough investigation of the potential of full-scale GAC filtration after DMF to remove organic micropollutants.

Conclusions

This dissertation describes the potential of chemically enhanced primary treatment with microsieving, direct membrane filtration and activated carbon adsorption, for the treatment of municipal wastewater, including the removal of organic micropollutants. The pilot-scale tests demonstrated the possibility of achieving high performance on a larger scale, while experiments in the laboratory demonstrated the potential for organic micropollutant adsorption, and provided information on the aeration required for supplementary biological treatment.

The appropriate choice of precipitation chemicals and their dosage was found to be highly important to achieve a high degree of removal of TOC and TP, and to ensure a high flux in DMF. It is strongly recommended that a variety of chemicals be tested before implementing CEPT. Very high removal of organic matter and phosphorus was achieved by CEPT with microsieving using 12 mg Al^{3+} L^{-1} and 3 mg L^{-1} polymer. Subsequent DMF led to even greater removal, with effluent TP concentrations < 0.1 mg L^{-1}.

Despite successful pre-treatment, the DMF membranes were fouled relatively quickly, which severely limits its potential for full-scale implementation. Inadequate cleaning of the membranes using hydrogen peroxide and citric acid further demonstrated the difficulties in achieving stable DMF operation, and the importance of proper membrane cleaning and managing fouling.

High removal of SS (> 90%) and organic matter was possible using CEPT with microsieving. Combined with a high biomethane potential of the sludge from microsieving, this could contribute to significant energy production. A potential electricity coverage of 61% was calculated. DMF was the most energy-intensive process in this treatment train. However, other studies have indicated the potential of DMF techniques that require substantially less energy.

A biological process following CEPT, microsieving and DMF could be used to further treat the wastewater, and would only need a fraction (< 25%) of the aeration required for aerobic degradation of the influent wastewater at Svedala WWTP. Implementing CEPT and microsieving would remove most of the need for aeration. Subsequent DMF would only lead to a further small reduction in the need for aeration.

The adsorption of organic micropollutants onto PAC was lower in the wastewater after physicochemical treatment than in tertiary treated wastewater. The lower adsorption of organic micropollutants was attributed to higher concentrations of DOC after physicochemical treatment. CEPT with microsieving followed by DMF produced a wastewater more suitable for micropollutant adsorption than CEPT and microsieving alone.

The removal of organic micropollutants using PAC in tertiary treated wastewater was similar to that of using GAC on tertiary treated wastewater; the degree of removal by PAC was in general just a little higher than that by GAC. The order of the degree of removal of different organic micropollutants was also similar for the two types of carbon.

The removal of organic micropollutants by CEPT with microsieving and DMF was small. However, all of the organic micropollutants studied were removed (≥ 98%) using a GAC filter after DMF, during approximately 4 days (440 bed volumes) of operation.

Future research

The results presented in this dissertation demonstrate the potential of CEPT with microsieving, DMF and activated carbon adsorption for the removal of organic micropollutants. The potential for high energy coverage by utilizing the separated sludge for biomethane production, and the potential of supplementary biological treatment were also demonstrated.

The most serious problem encountered during this work was fouling of the membrane during DMF. The problem of membrane fouling is well-known, and although there are promising techniques to reduce fouling, most have only been tested on laboratory scale. Studies on such techniques, and their influence on membrane fouling, on pilot scale are therefore suggested.

The potential of GAC filtration downstream of DMF to remove organic micropollutants was demonstrated for a short period. However, continuous, long-term experiments will be needed to further investigate the removal of organic micropollutants using a GAC filter. This is interesting due to the relatively high concentrations of DOC and the low concentrations of particles in the input to the GAC filter. High concentrations of DOC may result in quicker exhaustion of the GAC, but also the rapid establishment of a biofilm in the GAC filter, which may increase the removal of organic micropollutants.

DMF had a limited effect on the removal of DOC and was difficult to operate due to fouling. Therefore, another kind of filtration which is less sensitive to fouling should be tested after CEPT with microsieving, and as a pre-treatment to GAC filtration. Microsieving with small openings (10-20 µm) could be used to remove residual flocs after a first microsieving step with larger opening (100 µm). Microsieving would also have a higher filtration capacity than DMF. Another interesting option is a sand filter, in which a biofilm can grow and remove DOC.

Appendix

Table A1. The limits of quantification (LOQ) and relative standard deviation (RSD) in the analysis of the 21 organic micropollutants investigated

Compound	LOQ (ng L^{-1})	RSD (%)
Atenolol	0.1	0.5
Carbamazepine	0.5	0.7
Ciprofloxacin	5	3.3
Citalopram	1	1.8
Diclofenac	1	3.0
Estrone	0.05	1.2
Fluconazole	0.6	1.6
Ibuprofen	100	4.3
Imidacloprid	0.1	4.3
Ketoconazole	5	10.2
Losartan	0.1	3.2
Metoprolol	0.1	1.6
Naproxen	15	3.9
Oxazepam	0.6	1.6
Paracetamol	1	6.0
Propranolol	0.1	3.5
Sertraline	0.5	3.6
Sulfamethoxazole	0.1	2.4
Tramadol	2	2.5
Trimethoprim	1	1.4
Venlafaxine	1	6.3

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