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Treatment methods for the removal of per- and polyfluoroalkyl substances (PFASs) from drinking water

Optimizing existing and exploring novel treatment techniques

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Abstract

Worldwide, the demand for safe drinking water is rising. As drinking water sources are increasingly impacted by anthropogenic and environmental changes it is becoming a challenge to produce safe and clean water. The contamination of water with the man-made group of chemical compounds by the name of per- and polyfluoroalkyl substances (PFASs) adds to this challenge. Conventional drinking water treatment is not designed to remove these compounds due to their unique properties and guideline values for allowed concentrations in the final drinking water are low. Thus, there is a need for advanced techniques for the removal of PFASs in drinking water production.

During the course of this thesis, existing treatment methods were optimized and novel methods were developed. Filtration through granular activated carbon (GAC) filters, originally implemented in drinking water treatment for the removal of compounds causing odor and taste, was evaluated in a full-scale drinking water treatment plant and conclusions on the cost-effectiveness were gained by adapting flow-rates and describing several operation scenarios (Paper I). The technique of membrane filtration was studied in pilot-scale and different types of adsorption materials were evaluated for the removal of PFASs from raw water directly versus removal from the membrane concentrate. The adsorption materials GAC and anion exchange both performed better with larger ingoing PFAS concentrations. Filtration through specifically designed anion exchange resins can be an option for membrane concentrate management (Paper II and III).

Electrochemical oxidation and was shown to degrade PFASs in various water types, including membrane concentrate (Paper IV). Further, the commercially available technique heterogeneously catalysed ozonation was applied in pilot-scale to treat contaminated tap water and was observed to remove certain PFASs efficiently (Paper V). This thesis contributes to an increased knowledge on water treatment techniques for the removal of PFASs, providing new insights into water treatment options to protect human health.

Keywords: drinking water, per- and polyfluoroalkyl substances, PFAS, granular activated carbon, advanced oxidation processes

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Behandlingsmetoder för avlägsnande av per- och polyfluoralkylämnen (PFAS) från dricksvatten
- Optimering av befintliga och utforskning av nya behandlingstekniker

Sammanfattning

Efterfrågan på säkert dricksvatten ökar globalt och eftersom dricksvattentäckter alltmer påverkas av antropogena störningar och miljöförändringar har det blivit en utmaning att producera säkert och rent vatten. Kontaminering av vatten med konstgjorda kemiska föreningar som kallas för högfluorerade alkylsubstanser (PFAS) bidrar till denna utmaning. De tillåtna koncentrationerna i det slutliga dricksvattnet är låga och i och med att konventionell dricksvattenberedning inte är utformad för att avlägsna dessa ämnen, finns det således ett behov av avancerade tekniker för avlägsnande av PFAS.

I denna avhandling gjordes försök med att både optimera befintliga behandlingsmetoder som man vet är effektiva för PFAS-borttagning, samt att utveckla nya metoder. Filtrering genom granulerat aktivt kol (GAC), som ursprungligen implementerades i dricksvattenrening för avlägsnande av föreningar som orsakar lukt och smak, utvärderades i ett fullskaligt dricksvattenverk. Slutsatser om kostnadseffektivitet uppnåddes genom att anpassa vattenflöden och genom att beräkna olika driftscenarier (Artikel I). Membranfiltrering studerades i pilotskala och olika typer av adsorptionsmaterial utvärderades för avlägsnande av PFAS från råvattnet direkt kontra avlägsnande från membrankoncentratet. Båda adsorptionsmaterial (GAC och jonbytesmaterial) fungerade bättre med högre ingående PFAS-koncentrationer och filtrering genom specifikt designade jonbytarmaterial kan vara ett lämpligt alternativ för membrankoncentrathantering (Artikel II och III). Experiment med elektrokemisk nedbrytning visade sig bryta ned PFAS i olika vattentyper, inklusive membrankoncentrat (Artikel IV). Den kommersiellt tillgängliga nedbrytningstekniken ”heterogen katalyserad ozonisering” applicerades i pilotskala för att behandla förorenat kranvatten och observerades avlägsna vissa PFAS effektivt (Artikel V). Denna avhandling bidrar till en ökad kunskap om vattenbehandlingstekniker för avlägsnande av PFAS och alternativ för vattenbehandling för att skydda människors hälsa.

Nyckelord: dricksvatten, högfluorerade ämnen, PFAS, granulerat aktivt kol, avancerade oxidationsprocesser

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Behandlungsmethoden zur Entfernung von per- und polyfluorierte Alkylverbindungen (PFAS) aus Trinkwasser - Optimierung bestehender und Untersuchung von neuartigen Behandlungstechniken

Zusammenfassung

Der Bedarf an sauberem, gesundheitlich unbedenklichem Wasser steigt weltweit. Vielfältige Eingriffe und Belastungen werden dabei zunehmend zu einer großen Herausforderung für eine sichere und saubere Produktion von Trinkwasser. Der Kontamination mit Chemikalien wie per- und polyfluorierte Alkylverbindungen (PFAS) kommt dabei eine große Bedeutung zu, da sie mit konventionellen Verfahren der Trinkwasseraufbereitung nicht aus dem Wasser entfernt werden können. Diese Stoffe werden als gesundheitsgefährerdend angesehen weshalb die maximal zulässigen Konzentrationen im Trinkwasser sehr niedrig sind. Daraus resultiert ein großer Bedarf an neuen Techniken zur Entfernung von PFAS aus dem Wasser.

In dieser Arbeit werden optimierte Behandlungsmethoden und die Entwicklung neuer Verfahren beschrieben.In einer Trinkwasseraufbereitungsanlage wurde die Anwendung von Aktivkohlefiltern (GAC) untersucht, die ursprünglich zur Entfernung von geruchs- und geschmacksverändernden Stoffen eingesetzt wurden. Dabei konnten durch Anpassung der Flussrate und der Beschreibung mehrere Betriebsszenarien Schlussfolgerungen zur Kosteneffizienz gezogen werden (Artikel I). Membranfiltration wurde im Pilotmaßstab untersucht. Verschiedene Arten von Adsorptionsmaterialien für die Entfernung von PFAS direkt aus dem Rohwasser oder aus Membrankonzentrat wurden verglichen. Die Filtermaterialien GAC und Ionentauschen zeigten dabei eine bessere Leistung höheren eingehenden PFAS Konzentrationen. Die Filtration durch speziell entwickelte Ionentauscher stellt eine Option für das Management von Menbrankonzentrat dar (Artikel II und III).

Zudem konnte gezeigt werden, dass elektrochemische Behandlung PFAS in verschiedenen Wassertypen einschließlich Membrankonzentrat abbauen kann (Artikel IV). Heterogene Ozonierung wurde im Pilotmaßstab angewandt um kontaminiertes Leitungswasser zu behandeln. Bestimmte PFAS wurden dabei effizient entfernt (Artikel V). Insgesamt trägt die Arbeit zu einem erweiterten Wissen über Wasseraufbereitungstechniken zur Entfernung von PFAS bei und bietet neue Einblicke in Reinigungsmöglichkeiten zum Schutz von Mensch und Natur.

Schlüsselwörter: Trinkwasser, per- und polyfluorierte Alkylverbindungen, PFAS, PFC, Aktivkohle, moderne Oxidationsprozesse

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Dedication

To life. Thanks for being amazing! ☼

“The human race is challenged more than ever before to demonstrate our mastery, not over nature but of ourselves.”
Rachel Carson

Contents

List of publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I N. Belkouteb*, **V. Franke***, P. McCleaf, S. J. Köhler and L. Ahrens (2020). Removal of per- and polyfluoroalkyl substances (PFASs) in a full-scale drinking water treatment plant: long-term performance of granular activated carbon (GAC) and influence of flow-rate. *Water Research*, 182, pp. 115913.

II **V. Franke**, P. McCleaf, K. Lindegren and L. Ahrens (2019). Efficient removal of per- and polyfluoroalkyl substances (PFASs) in drinking water treatment: nanofiltration combined with active carbon or anion exchange. *Environmental Science: Water Research & Technology*, 5, pp. 1836–1843.

III **V. Franke**, M. Ullberg, P. McCleaf, M. Wålinder, S. J. Köhler and L. Ahrens. The price of really clean water - Combining nanofiltration with granular activated carbon and anion exchange resins for the removal of per- and polyfluoralkyl substances (PFASs) in drinking water production. *Under review in Environmental Science & Technology Water.*

IV **V. Franke**, W. Nassazzi and L. Ahrens (Manuscript). Electrochemical treatment for the removal of per- and polyfluoroalkyl substances (PFASs) in various water types using boron-doped diamond electrodes.

V **V. Franke**, M. D. Schäfers, J. J. Lindberg and L. Ahrens (2019). Removal of per- and polyfluoroalkyl substances (PFASs) from tap water using heterogeneously catalyzed ozonation. *Environmental Science: Water Research & Technology*, 5, pp. 1887–1896.

*shared first authorship

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The contribution of Vera Franke (VF) to the papers included in this thesis was as follows:

I VF was responsible for restructuring and reevaluating the data retrieved during N. Belkouteb's master thesis project. VF and N. Belkouteb shared the main responsibility for writing the manuscript with the support of all authors.

II VF performed extensive data evaluation of the data retrieved during K. Lindegren's master thesis project and was responsible for writing the manuscript with the support of all authors.

III VF had the main responsibility for the experimental design, performance of the experiment, and analysis of the data. Further, VF had the main responsibility for data evaluation and writing of the manuscript with support of all authors.

IV As main supervisor of master student W. Nassazzi, VF had the main responsibility for the experimental design of the experiment, as well as data evaluation and interpretation. VF had the main responsibility for the communication between involved parties and writing the manuscript with support of all authors.

V As main supervisor of master student M. Schäfers, VF was responsible for the design of the study, retrieval and interpretation of the data as well as communication with the involved company. VF prepared the manuscript with the help of all authors.

Additional papers

In addition to the papers included in the thesis, the author has contributed to the following peer-reviewed publications:

M. Sörengård, **V. Franke**, R. Tröger, L. Ahrens (2020). Losses of poly- and perfluoroalkyl substances to syringe filter materials, *Journal of Chromatography A*, 1609, pp. 460430

R. Tröger, S. J. Köhler, **V. Franke**, O. Bergstedt, K. Wiberg (2020). A case study of organic micropollutants in a major Swedish water source – Removal efficiency in seven drinking water treatment plants and influence of operational age of granulated active carbon filters, *Science of The Total Environment*, 706, pp. 135680.

Abbreviations

Technical terms

AFFF	Aqueous film-forming foam (also aqueous fire-fighting foam)
AIX	Anion exchange
BDD	Boron-doped diamond
DOC	Dissolved organic carbon
DWTP	Drinking water treatment plant
EBCT	Empty bed contact time
GAC	Granular activated carbon
HPLC	High-performance liquid chromatography
IS	Internal standard
LOQ	Limit of quantification
MS/MS	Tandem mass spectrometry
NF	Nanofiltration
PFASs	Per- and polyfluoroalkyl substances
PFCAs	Per- and polyfluoroalkyl carboxylic acids
PFSAs	Per- and polyfluoroalkyl sulfonic acids
$\sum_{11}$PFAS	Sum of 11 PFASs included in the Swedish drinking water guidelines
SPE	Solid-phase extraction
TOC	Total organic carbon
UPLC	Ultra-performance liquid chromatography
UV	Ultraviolet

Per- and polyfluoroalkyl substances

10:2 FTSA	10:2 Fluorotelomer sulfonic acid
4:2 FTSA	4:2 Fluorotelomer sulfonic acid
6:2 FTSA	6:2 Fluorotelomer sulfonic acid
8:2 FTSA	8:2 Fluorotelomer sulfonic acid
9Cl-PF2ONS	9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid
ADONA	Dodecafluoro-3H-4,8-dioxanonanoic acid
EtFOSA	N-ethyl perfluorooctane sulfonamide
EtFOSAA	N-ethyl perfluorooctane sulfonamido acetic acid
EtFOSE	N-ethyl perfluorooctane sulfonamidoethanol
FOSA	Perfluoroctane sulfonamide
HFPO-DA	Tetrafluoro-2-(heptafluoropropoxy)propanoic acid
MeFOSA	N-methyl perfluorooctane sulfonamide
MeFOSAA	N-methyl perfluorooctane sulfonamido acetic acid
PFCAs	MeFOSE N-methyl perfluorooctane sulfonamidoethanol
P37DMOA	Perfluoro-3,7-dimethyloctanoic acid
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid
PFDA	Perfluorodecanoic acid
PFDS	Perfluorodecanoic sulfonic acid
PFDoDA	Perfluorododecanoic acid
PFDoDS	Perfluorododecanoic sulfonic acid
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptane sulfonic acid
PFHxA	Perfluorohexaanoic acid
PFHxDA	Perfluorohexadecanoic acid
PFHxS	Perfluorohexane sulfonic acid
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoic acid
PFOcDA	Perfluorooctadecanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPeA	Perfluoropentanoic acid
PFPeS	Perfluoropentane sulfonic acid
PFTeDA	Perfluorotetradecanoic acid
PFTriDA	Perfluorotridecanoic acid
PFUnDA	Perfluoroundecanoic acid

1 Introduction

Access to clean and safe drinking water is a basic human right and essential to our health (UN, 2017; WHO, 2017). The vast majority of water sources require some form of treatment prior potable use. Historically, water treatment has focussed on filtration to clarify water and the implementation of disinfection processes to control pathogens. More recently, the potential harm of man-made organic chemicals has given rise to concern, leading to various regulations of these chemicals in drinking water (Crittenden et al., 2012). In this thesis different water treatment technologies were evaluated for the removal of a certain group of chemicals, per- and polyfluoroalkyl substances, from water.

1.1 Per- and polyfluoroalkyl substances

Per- and polyfluoroalkyl substances (PFASs) are a group of more than 4 700 different chemical compounds and have been produced since the 1950's (Kissa, 2001; OECD, 2018). Unique physico-chemical properties make PFASs suitable for all kinds of applications calling for oil- and water-repellency and durability.

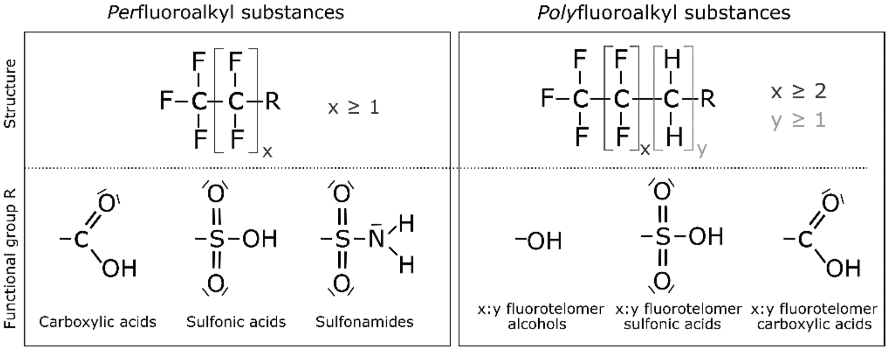


Figure 1: Examples of important non-polymeric PFAS classes frequently found in the aquatic environment.

Areas of application include industrial processes like metal-plating, photoresistors and anti-reflective coatings for semiconductors, medical devices as well as everyday consumer products like non-stick cookware, water-repellent textiles, sunscreens, dental floss or cosmetics (Paul et al., 2009; Herzke et al., 2012; Schultes et al., 2018; Goldenman et al., 2019). PFASs can be divided into polymer and non-polymer substances (Buck et al., 2011). PFAS polymers are characterized by the sequence of monomer units resulting in high molecular weight structures, which are generally regarded as substances of low concern (ECHA, 2012). Non-polymer PFASs are mobile in the environment and are commonly considered to have a larger impact on human health and the environment than polymeric PFASs and are therefore interesting for work related to aquatic systems (ITRC, 2020). Non-polymeric PFASs, for pragmatic reasons referred to as PFASs within this thesis, can be divided into perfluoroalkyl substances, containing a fully fluorinated carbon chain and polyfluoroalkyl substances, in which the carbon chain is partly fluorinated with at least one fluorine atom (Buck et al., 2011). PFASs frequently detected in aqueous systems comprise both a hydrophilic functional group and a hydrophobic fluorinated carbon chain of different length, see Figure 1 (Buck et al., 2011; Ahrens & Bundschuh, 2014). Several PFASs have been recognized as substances of very high concern, due to their persistent and bioaccumulative properties as well as existing evidence for various toxicological effects (DeWitt, 2015; ECHA, 2017). With very few exceptions, PFASs do not occur naturally. Given this, their global distribution in both the biotic and abiotic environment is astonishing. PFASs can be detected in the blood of nearly every human being as well as in extremely remote locations such as the Arctic, which highlights their persistence in the environment and their mobile nature enabling them to undergo long-range transport (Young et al., 2007; CDC, 2019). Emissions via e.g. wastewater and landfill leachate, sludge recycling or the use of PFAS containing aqueous film-forming foams (AFFFs) during fire-fighting can lead to widespread environmental contamination posing a problem for soil and water use in numerous places worldwide (Goldenman et al., 2019).

While exposure to PFASs via consumer products like make-up, water-repellent textiles or cookware presents an undisguised route of human exposure, more deceptive pathways include contaminated food or drinking water. Conventional drinking water treatment often remains ineffective for the removal of PFASs from a contaminated water source and PFAS uptake from drinking water can thus be a dominant exposure pathway (Vestergren & Cousins, 2009; Thompson et al., 2011; Eschauzier et al., 2012; Appleman et al., 2013; Rahman et al., 2014). In order to minimize human exposure to PFASs through drinking

Table 1: List of the 11 PFASs included in the current drinking water guideline issued by the Swedish Food Agency. It states that the sum of these 11 should not surpass a concentration of 90 ng L^{-1} in the final drinking water (Ankarberg & Lindberg, 2016). The number in parenthesis denotes the number of fully fluorinated carbon atoms (C^F_x). Note that in the aqueous phase, these compounds appear in their deprotonated (anionic) form.

	Name	**Formula (C^F_x)**
PFBS	Perfluorobutane sulfonic acid	$C_4F_9SO_3H$ (4)
PFHxS	Perfluorohexane sulfonic acid	$C_6F_{13}SO_3H$ (6)
PFOS	Perfluorooctane sulfonic acid	$C_8F_{17}SO_3H$ (8)
PFBA	Perfluorobutanoic acid	C_3F_7COOH (3)
PFPeA	Perfluoropentanoic acid	C_4F_9COOH (4)
PFHxA	Perfluorohexaanoic acid	$C_5F_{11}COOH$ (5)
PFHpA	Perfluoroheptanoic acid	$C_6F_{13}COOH$ (6)
PFOA	Perfluorooctanoic acid	$C_7F_{15}COOH$ (7)
PFNA	Perfluorononanoic acid	$C_8F_{17}COOH$ (8)
PFDA	Perfluorodecanoic acid	$C_9F_{19}COOH$ (9)
6:2 FTSA	6:2 Fluorotelomer sulfonic acid	$C_8H_4F_{13}SO_3H$ (6)

water, regulations have been introduced all over the world for single or several PFASs. In Sweden, a guideline value of 90 ng L^{-1} for the sum of 11 different PFASs ($\sum_{11}$PFAS) exists (see Table 1).

1.2 Drinking water treatment techniques

From simple filtration techniques for the removal of solids to disinfection methods to control threats arising from the presence of bacteria or viruses, modern drinking water treatment has developed to become a sophisticated combination of processes producing water that is safe to drink and aesthetically pleasing (Crittenden et al., 2012). Drinking water can be produced from surface water, natural and artificially recharged groundwater or from desalination of sea water. As drinking water treatment is dependent on the nature of the raw water source, there is no common technique applied by all waterworks. Treatment often involves the removal of particles through e.g. a combination of coagulation, sedimentation and filtration, and a disinfection step. Generally, conventional water treatment is not designed to remove organic micropollutants, such as PFASs (Stackelberg et al., 2007; Thompson et al., 2011). Even oxidation processes like ozonation are, under typical conditions applied in drinking water treatment plants (DWTPs), ineffective for PFAS removal and may on the other hand lead to the oxidation of precursor compounds to more stable PFASs

(Rahman et al., 2014). Treatment methods able to remove PFASs from water in large scale include filtration through fresh granular activated carbon (GAC) or anion exchange (AIX) resins and high-pressure membrane processes, like nanofiltration (NF) or reverse osmosis (Rahman et al., 2014; Merino et al., 2016), see Figure 2.

1.2.1 Separation processes

Treatment techniques based on physical separation of PFASs from the water phase can be divided into adsorption processes and techniques based on size-exclusion and diffusion (Figure 2).

Adsorption processes

While initial intentions were to remove compounds causing turbidity, odor, taste and discolouring of the water, adsorption processes today further act as chemical and microbiological barriers. Commercially available granular activated carbon (GAC) is the most frequently applied adsorbent for PFAS removal in common DWTP systems due to their low cost and ease of application (Espana et al., 2015). Carbon materials such as GAC and powdered activated carbon are produced from natural carbon based materials, which are heated in an oxygen deficient atmosphere and thereafter activated at temperatures >750°C in the presence of selected gasses (Mattson & Mark, 1971). Base materials include coconut shells, bituminous coal or bamboo.

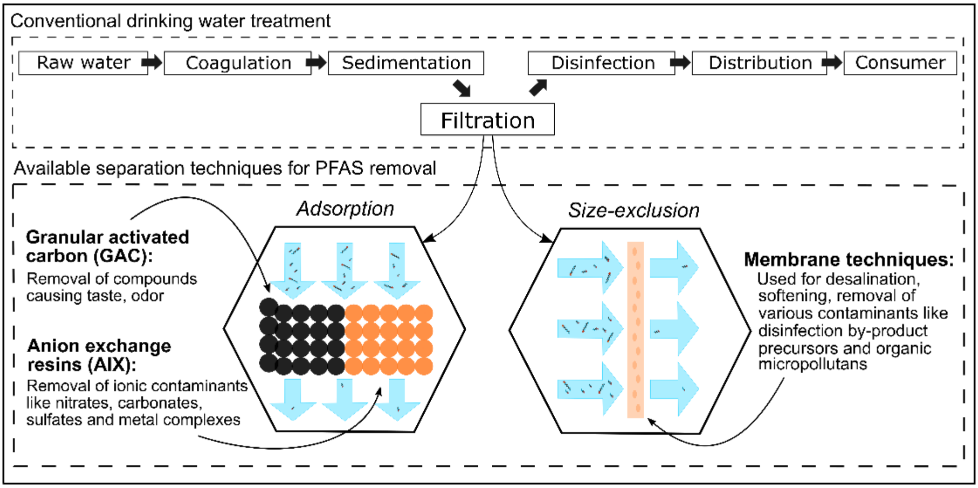


Figure 2: Illustration of conventionally available separation techniques capable of removing PFASs during drinking water treatment.

A complex combination of large surface area, the presence of macro and mesopores and certain functional groups able to undergo anion exchange processes or hydrophobic interactions are responsible for PFAS adsorption to activated carbons (Zhi & Liu, 2015; Zaggia et al., 2016; Kothawala et al., 2017). Fresh activated carbon shows a relatively high adsorption capacity for PFASs, especially for the longer chain homologues (Du et al., 2014; McCleaf et al., 2017). When applied for a range of different PFASs, however, GAC filters need to be replaced or reactivated frequently, as breakthrough of short-chain PFASs occurs rather fast (Rahman et al., 2014; McCleaf et al., 2017). Regeneration is most often accomplished thermally through heating at 800-1000°C in an oxygen deficient atmosphere, at which PFASs are destructed (Narbaitz & Karimi-Jashni, 2009).

AIX resins present another type of adsorption materials commonly applied in water treatment today. Synthetic AIX resins are very durable and generally have a more controlled distribution of pore sizes than GAC (Senevirathna et al., 2010). Resins are primarily used for water softening and demineralization, but applications can include the removal nitrate, barium, radium, arsenic, perchlorate, and chromate (Crittenden et al., 2012). Resins suitable for the adsorption of weak organic acids, such as perfluoroalkyl carboxylic acids (PFCAs) or perfluoroalkyl sulfonic acids (PFSAs), are strong base anion exchange resins with quaternary amine or dimethylethanolamine functional groups. Adsorption of PFASs on AIX resins is mainly based on electrostatic interactions. For the more hydrophilic short-chain compounds, such as PFBA and PFBS, electrostatic interactions play a more important role than for longer chain PFASs, such as PFOA and PFOS for which the hydrophobicity of the resin influences the adsorption capacity largely (Zaggia et al., 2016). AIX resins are usually regenerated by washing with concentrated solutions of inorganic salts. While this has proven inefficient for replacing PFASs on the AIX sorption sites, other regeneration methods include the application of organic alcohols in combination with dilute salt solutions (Deng et al., 2010; Zaggia et al., 2016). Proper management of the resulting salt brine, if laden with PFASs, presents a challenge until today and needs to be addressed.

Membrane filtration

Membrane processes applied in municipal water treatment today include microfiltration, ultrafiltration, NF and reverse osmosis and are based on differences in permeability of the water constituents (Crittenden et al., 2012). For objectives like solid particle removal and microorganism removal, low-

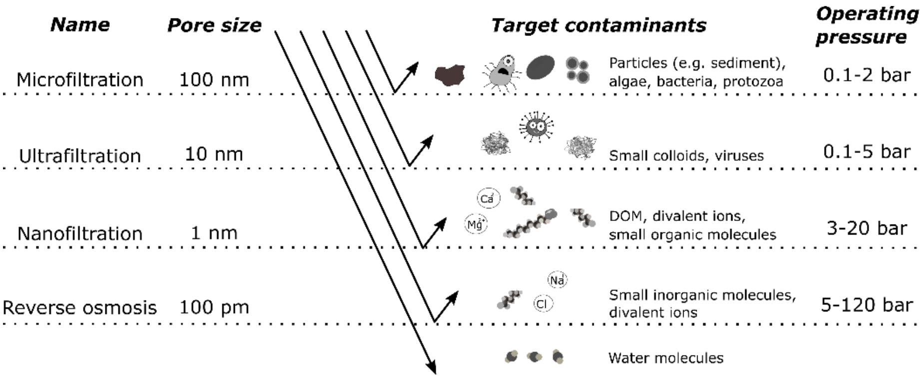


Figure 3: Illustration of the membrane processes applied in municipal water treatment. DOM = dissolved organic matter. Microfiltration and ultrafiltration are defined as low-pressure membrane processes, while nanofiltration and reverse osmosis are referred to as high-pressure techniques (Crone et al., 2019). Figure adopted from Crittenden et al. (2012). Typical operating pressures according to vVn der Bruggen et al. (2003).

pressure membrane processes like microfiltration or ultrafiltration can be applied. For the removal of dissolved solutes, such as PFASs, the high-pressure membrane techniques NF or reverse osmosis membranes are applicable (Figure 3). Next to size-exclusion, rejection mechanisms of high-pressure membranes include charge effects and diffusion (Van der Bruggen et al., 2003; Bellona et al., 2004). Membranes are thin sheets consisting of semipermeable, typically synthetic materials. Permeable constituents pass the membrane to result in what is called permeate, while impermeable components are retained on the feed side and collected in so-called concentrate (also referred to as retentate or brine; Crittenden et al., 2012). Recoveries of NF or reverse osmosis typically lays between 50-90 % depending on the raw water source, i.e. 10-50 % of the feedwater volume will be rejected by the membrane in the form of membrane concentrate. The concentrate is often disposed of by release into nearby watercourses, injection into non-potable aquifers or treatment in wastewater treatment facilities, however other management solutions need to be applied if the concentrate is laden with pollutants like PFASs to avoid environmental contamination (Van der Bruggen et al., 2003). Proper concentrate management presents a major challenge for the application of membrane filtration for PFAS removal until today (Crone et al., 2019).

1.2.2 Chemical transformation processes

In contrast to processes based on physical separation of contaminants from the water phase chemical transformation processes are used to treat and degrade unwanted constituents *in-situ*. Chemical transformation processes applied in drinking water treatment for the removal of organic matter, such as coagulation/flocculation, or the inactivation/disinfection of various types of microorganisms, like chlorine-based oxidation processes, ozonation or UV-treatment, are inefficient in removing PFASs at conventional conditions (Rahman et al., 2014, and references therein). Much research has been conducted on the application of advanced oxidation and reduction processes, however mostly in laboratory-scale.

Due to the complete substitution of fluorine for hydrogen in the carbon chain of PFASs they are recalcitrant towards oxidation (Schröder & Meesters, 2005). Fluorine is the strongest inorganic oxidant and it is therefore thermodynamically unfavourable to oxidize fluorine in oxidation state (-1) to its elemental state of $F_2(0)$ with any other one-electron oxidant (Vecitis et al., 2009). Oxidation processes based on the reactions with hydroxide radicals do not have any effect on PFAS concentrations (Dombrowski et al., 2017). The most promising advanced oxidation/reduction processes are treatments in which various highly active radical species are created or electrons are transferred directly. This includes persulfate oxidation, the combination of UV radiation with an oxidizing agent and/or catalyst as well as sonochemical and electrochemical treatment, photolysis or the application of electric discharge (Hori et al., 2005; Vecitis et al., 2009; Lee et al., 2012; Cheng et al., 2014; Chen et al., 2016; Fernandez et al., 2016; Merino et al., 2016; Niu et al., 2016; Qu et al., 2016).

1.2.3 Other methods

Plenty of studies have evaluated novel adsorption materials potentially useful for PFAS removal from water. This includes carbon nanotubes, certain mineral materials and biosorbents (e.g. Deng et al., 2012; Chen et al., 2011; Johnson et al., 2007; Tran et al., 2015). However, an application of these materials in full-scale remains to be evaluated. Additionally, much work has been done on novel degradation techniques like e.g. enzyme catalyzed oxidative humification reaction, ball-milling, the application of activated sewage sludge and γ- radiation or plasma reactions (Colosi et al., 2007; Zhang et al., 2013, 2014; Stratton et al., 2017).

While it is recognized that many of the mentioned techniques show potential for the removal of PFASs in drinking water treatment, the current thesis focusses on treatments involving adsorption to GAC and AIX resins, membrane filtration, electrochemical treatment and heterogeneously catalyzed ozonation.

2 Objective and research questions

Drinking water providers are obliged to deliver clean and safe water. The removal of PFASs is especially difficult, as many PFASs are persistent and highly water soluble and thus not effected by conventional drinking water treatment. Currently, adsorption to GAC is one of the most commonly applied treatment methods. The early break-through of certain PFASs, however, requires frequent exchange of the filters making this filtration technique cumbersome. As an alternative to activated carbon, specifically designed AIX resins are discussed as more efficient adsorption materials. Further, filtration through high-pressure membranes (i.e. NF or reverse osmosis) has been shown to remove a wide range of organic micropollutants, including PFASs, and other unwanted water constituents. However, the resulting contaminated waste stream of up to 10-50 % of the feedwater poses a challenge for the application of membrane techniques. The following research questions guided this thesis work and addressed the overall objective to identify integrated and cost-efficient methods for the removal of PFASs in drinking water treatment:

1. Can the adjustment of flow-rates through full-scale GAC filters improve the removal of PFASs in drinking water treatment for optimal cost-benefit? (Paper I)
2. How does the treatment method of NF perform for the removal for PFASs from groundwater and is filtration through GAC or AIX resins suitable for the treatment of the NF concentrate? (Paper II and III)
3. How do different water types influence PFAS removal during electrochemical treatment? (Paper IV)
4. Is the treatment method of heterogeneously catalyzed ozonation capable of removing PFASs from contaminated water at environmentally relevant concentrations? (Paper V)

3 Materials and methods

3.1 General study design

In **Paper I**, the performance of a full-scale DWTP was investigated for the removal of PFASs from an impacted groundwater source. The performance of the whole treatment process was evaluated, with a special focus on the GAC filters. The studies described in **Paper II and III** investigated NF membrane treatment as an alternative process in pilot-scale, while utilizing the adsorption materials GAC and AIX resins for the treatment of contaminated membrane concentrate in column tests. For comparison, both raw water and concentrate were treated by adsorption materials. Evaluated novel methods included electrochemical treatment (**Paper IV**), and heterogeneously catalyzed ozonation (**Paper V**). Electrochemical treatment was applied in bench scale batch tests for the removal of PFASs in three different water matrices, including NF concentrate. Heterogeneously catalyzed ozonation was applied in laboratory-scale where the addition of persulfate to the catalyzed ozone system was evaluated for a potential improvement of PFAS removal from ultrapure water. The commercially available technique was further tested in pilot-scale for the removal of PFASs in tap water.

3.2 Treatment techniques

Full-scale drinking water treatment plant Bäcklösa

The DWTP Bäcklösa is located in the south of the City of Uppsala, Sweden, and supplies ca. 80 000 people with drinking water producing 7 million m^3 yearly. Raw water is received from two different groundwater wellfields located in Sunnersta and Stadsträdgården in Uppsala. Both wellfields are impacted by a

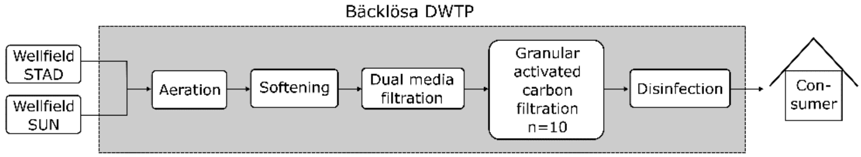


Figure 4: Schematic overview of the treatment train applied in the Bäcklösa drinking water treatment plant (DWTP). Ingoing water from the wellfields located in Stadsträdgården (STAD) and Sunnersta (SUN) is mixed and subsequently treated by aeration, softening and pH adjustment, dual media filtration before reaching the ten granular activated carbon (GAC) filters running in parallel. After filtration through GAC the water is disinfected and thereafter passed to the distribution network.

historical contamination with PFAS containing AFFFs in soil across the groundwater flow and show elevated detected P11PFAS concentrations of approximately 20 ng L^{-1} (Sunnersta) and 150 ng L^{-1} (Stadsträdgården), respectively. The treatment in Bäcklösa DWTP consists of aeration, softening (fluidized bed pellet reactor type), dual media filtration (granular carbon and sand), GAC filtration and disinfection with free chlorine, see Figure 4. For the evaluation of the full-scale treatment (**Paper I**), samples were taken for the incoming water and after each treatment step. In order to evaluate the performance of the different GAC filters, separate samples were taken for the water treated by each of the filters running in parallel. A change in flow-rates through two of the GAC filters of different age was conducted to evaluate the influence of flow-rates and GAC age on the removal of PFASs in the full-scale filters. Initial flow-rates of 35 L s^{-1} were first increased to 45 L s^{-1} and subsequently adjusted to 30 and 15 L s^{-1}, respectively. To ensure stable conditions, samples were taken and analyzed for UV light adsorption at 254 nm, which presents an accurate and quick measuring technique. PFAS samples were taken after stable values for UV were acquired (varying between 3-7 days). In a cost-analysis unit regeneration cost were determined for one of the full-scale filters evaluated during the study described in **Paper I**. The calculations were based on annual regeneration costs for the filter in the Bäcklösa DWTP and different treatment goals: $\sum_{11}$PFAS < 10 ng L^{-1}, 25 ng L^{-1}, 50 ng L^{-1}, and 85 ng L^{-1}, respectively.

Nanofiltration

Pilot-scale NF units were employed to treat groundwater received in the Bäcklösa DWTP. Experiments described in **Paper II** applied a NF plant consisting of spiral wound membranes with a molecular weight cut-off of 270 Da (NF 270-400; Dow Filmtech$^{\text{TM}}$ Membranes). Two membranes were operated

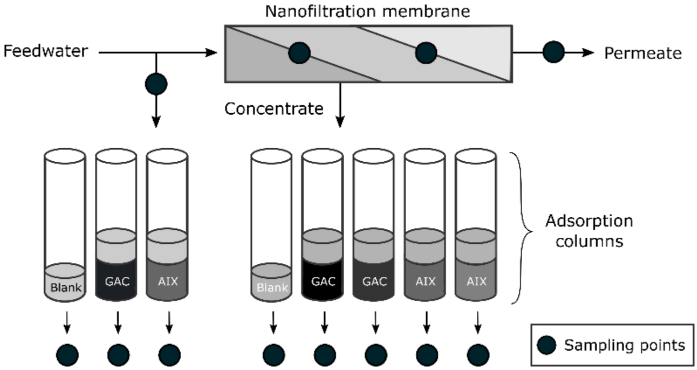


Figure 5: Conceptual figure illustrating the experimental setup in Paper II and III.

in series and a 30 µm patron filter was installed for the removal of potentially interfering particles while a feedwater pump provided simultaneous injection of antiscalant. The NF unit was operated at a feedwater flow-rate of 2.3 m^3h^{-1} with a targeted 78 % recovery rate. In experiments described in **Paper III** involved the application of a nanofiltration plant consisting of a two-stage membrane setup combining membranes with 90 Da (first stage) and 270 Da (second stage) molecular weight cut-offs. The pilot was operated at 8 m^3h^{-1} feedwater flow and anticipated 80 % recovery. A 5 µm pre-filter was installed to prevent solids from entering the NF unit.

As the NF process is under consideration for a new full-scale treatment plant for the City of Uppsala, the NF process was selected to achieve several treatment goals, such as the removal of mineral hardness, uranium-238, DOC, bromide and PFASs from the raw water and to serve as a microbiological barrier. For adsorption experiments membrane feedwater (i.e. raw water) and membrane concentrate were collected in 1000 L tanks, which were allowed to constantly overflow. Peristaltic pumps were used to transfer water from the tanks to columns containing adsorption material at a flow rate of 70-100 mL min^{-1}.

Adsorption materials in column experiments

Column experiments described in **Paper II and III** applied both GAC and AIX as adsorption materials. The GAC material Filtrasorb 400® (Calgon Carbon Corporation, Feluy, Belgium) and the AIX resin A600 (Purolite®, Llantrisant, Wales) were applied in both studies while the GAC material Norit 1240 W (Norit Nederland BV, Amersfoort, The Netherlands) and the AIX resin PFA694 (Purofine®, King of Prussia, Pennsylvania, USA) were applied additionally in the study described in **Paper III**. For more details on the specific properties of the materials it is referred to the manufacturers and the respective papers.

Columns were made of glass (**Paper II**) or acrylic glass (**Paper III**) with a sintered glass filter or stainless steel mesh at the bottom of each column, respectively, holding the adsorption material in place. Columns were supplied with water from the top. Due to clogging difficulties the materials had to be backwashed occasionally, which was done by introducing raw water at the bottom of the columns. Samples for PFAS analysis were taken once a week for a period of 35 weeks (**Paper II**) and approximately every two weeks for a period of 26 weeks (**Paper III**). Samples for analysis of organic matter and inorganic ion content were taken more frequently and only in the beginning of the experiments described in **Paper III**.

Electrochemical treatment

A custom-made electrochemical flow-cell provided by NOVA Diamant AB (Uppsala, Sweden) was applied in the study described in **Paper IV**. The cell consisted of six boron-doped diamond (BDD) electrodes, i.e. BDD coatings on 0.1 mm niobium sheets, with an effective area of 70 cm^2 each. Electrodes acted in a bipolar manner and were connected in series. A case made of polyvinyl chloride kept the electrode assembly in place. The cell was connected to a power supply providing up to 60 V. The studied solutions were circulated from a glass beaker to the flow-cell through compressible tubes with a peristaltic pump. The beaker was placed on a magnetic stirrer and kept under constant mixing. The solutions were allowed to circulate through the experimental system for > 10 hours before a first sample was taken as reference samples. Subsequently, the electrochemical treatment was started by applying a certain current to the solutions. Depending on the conductivity of the solutions, a certain amount of anhydrous sodium sulfate was added to ensure sufficient conductivity. Samples for PFAS analysis were taken in 5 mL aliquots throughout the course of each experiment.

Catalyzed ozonation

Catalyzed ozonation evaluated in the study described in **Paper V** applied a method developed by the company Ozone Tech Systems OTS AB (Hägersten, Sweden). The treatment is commercially available and frequently used for the removal of various organic micropollutants from e.g. hospital wastewater and has shown promising results for the treatment of water contaminated with PFASs (Joos Lindberg, 2016; Ozonetech, 2020). The treatment applies ozone in combination with a catalytic bed, consisting of iron oxide based catalyst granulates. In laboratory-scale trials, all possible combinations of 5 g catalyst material, 300 mg h^{-1} ozone and/or 187 mg ammonium persulfate were applied in batch experiments treating 500 mL of MilliQ water (Millipak$^{©}$ Express 20, 0.22 µm filter, Merck Millipore) fortified with each 1 µg L^{-1} of 14 different PFASs. In the pilot-scale experiment of the study, a system provided by Ozone Tech Systems was applied to treat 50 L tap water fortified with 17 PFASs. The system applied a 10 L catalytic bed, consisting of the same granulates as evaluated in the laboratory-scale experiments, and an introduction of 5 g h^{-1} produced by a Ozonetech ICT-5 ozone generator. In the first part (day 1) of this experiment, adsorption to the system walls was monitored by adding a low concentration of 100 ng L^{-1} per compound. The catalytic bed was excluded from the system at this stage. In a second step (day 2-5) PFAS adsorption to the catalytic material was monitored. PFAS concentrations were increased to a total concentration of 1.1 µg L^{-1} per compound and the water was circulated through the whole system, including the catalytic bed. On day 6, when it was ensured that ad- and desorption processes of PFASs were in equilibrium, ozone was introduced to the system for 8 hours and samples were taken in 5 mL aliquots after 0.5, 1, 2, 3, 4, 5, 6, 7, and 8 hours.

3.3 Chemical analysis

PFAS target analytes and quantification

Analysis of PFASs conducted at SLU as described in **Paper I, III, IV and V** included a list of up to 37 target PFASs, comprised of C^{F}_{3} - C^{F}_{13}, C^{F}_{15} and C^{F}_{17} PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA, PFOcDA), C^{F}_{4}- C^{F}_{8}, C^{F}_{10} and C^{F}_{12} PFSAs (PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFDS, PFDoDS), perfluorooctane sulfonamide, as well as methyl- and ethyl derivatives (FOSA, Me-FOSA, Et-FOSA), and several precursor compounds, such as C^{F}_{4}, C^{F}_{6}, C^{F}_{8}, C^{F}_{10} fluorotelomer sulfonic acids (4:2 FTSA, 6:2 FTSA, 8:2 FTSA, 10:2 FTSA), methyl and ethyl perfluorooctane sulfonamidoethanol (Me-FOSE, ET-FOSE), and methyl and ethyl perfluorooctane sulfonamide acetic acid (Me-FOSAA, ET-FOSAA). Further, dodecafluoro-3H-4,8-dioxanonanoic acid (ADONA), tetrafluoro-2-(heptafluoropropoxy)propanoic acid (HFPO-DA), perfluoro-3,7-dimethyloctanoic acid (P37DMOA) and 9-chlorohexadecafluoro-3-oxanonane-1-sulfonic acid (9Cl-PF2ONS, major component of F-53B) were included in the analysis in the studies described in **Paper III and IV**. For quantification purposes samples were fortified with a mixture of mass-labelled internal standards (IS) prior extraction. Depending on the study the IS mixture contained 16-26 compounds and target analytes were matched according to structural similarity. For details on analyte-IS pairs, IS concentration and limits of quantification it is referred to the respective studies included in this thesis.

Sample handling and sample preparation

For studies described in **Papers I and III**, samples were extracted using a solid-phase extraction (SPE) according to the ISO 25101:2009 method (ISO, 2008). Samples were collected in methanol-cleaned 1 L polypropylene bottles and stored in the dark at 8°C until further handling. Samples in **Paper I** were filtered through 0.45 µm glass microfiber filters and subsequently fortified with 100 µL of the IS mixture (see above). The extraction method itself is described in detail e.g. Ahrens et al. (2015). Briefly, extraction cartridges (Oasis® WAX cartridges, 6cc, 500 mg, 60 µm; Waters, Ireland) were preconditioned with ammonium hydroxide solution, methanol and MilliQ water. Subsequently, the samples were loaded onto the cartridges at one to two drops per second. After that the cartridges were washed by adding ammonium acetate buffer and dried through centrifugation. Samples were eluted from the cartridges before the extracts were concentrated to 0.5 mL by evaporating with nitrogen. Prior instrumental analysis 0.5 mL MilliQ water was added to each sample.

Instrumental analysis

Depending on the study, samples were analyzed as extracts prepared by off-line SPE, through direct injection into the instrument or through large-volume injections processed by on-line SPE. PFAS analysis was conducted on high- or ultra-performance liquid chromatography – tandem mass spectrometers (HPLC-MS/MS, UPLC-MS/MS) in which all Teflon® parts had been replaced to avoid background concentrations. Additionally, trapping columns were installed after the mixing chamber in order to distinguish sample peaks from possible mobile phase contamination. For details on instrument settings and parameters and analyte transitions see the respective papers. Parts of the samples evaluated in **Paper I, II and III** were sent to the commercial laboratory ALS Scandinavia for analysis.

Analysis of organic carbon and inorganic ions

Samples for the analysis of organic carbon were collected in pre-cleaned 25 mL glass flasks and analysed within one week during the study described in **Paper III**. Firstly, UV absorbance was measured using a photometer. Subsequently, the samples were acidified with hydrochloric acid and analysed with the help of a catalytic combustion analyser. Organic matter concentrations retrieved with this method describe the non-purgeable organic carbon content, which is referred to as the total organic carbon (TOC) content in **Paper III**. For determining the truly dissolved fraction of the organic carbon content (dissolved organic carbon, DOC), a pre-experiment was conducted and samples of each respective water type were filtered through a 0.45 µm filter. Differences between filtered and unfiltered samples were within measuring uncertainties and the filtering step was thus omitted on organic matter analysis. The analysis of inorganic ions in Paper III was conducted by the geochemical laboratory at SLU (accredited by SWEDAC). Certain samples were analyzed by the laboratories of Uppsala Vatten and Waste AB.

3.4 Quality control and quality assurance

All glassware was burnt at 400°C and rinsed with methanol prior contact with the samples. Polypropylene bottles were rinsed with methanol prior sample collection. For organic matter analysis flasks were dish washed and soaked in MilliQ water before use. Whenever feasible, samples were collected in duplicates (for PFAS analysis) or triplicates (analysis of organic matter). Field blanks consisted of pre-cleaned bottles filled with MilliQ water, which were opened at the experiment site during samples collection. Laboratory blanks were included during sample preparation through SPE, where one to three blanks were added to each extraction batch. Blank samples were handled in an identical manner as the actual samples and concentrations detected in the blanks were used to determine limits of quantification. For PFAS analysis, nine-point calibration curves were run in the beginning of the sequence and after every 12-15 sample injections. For a compound to be quantified, calibration curves had to be strongly linear ($R^2>0.99$). Limits of quantification (LOQ) were defined as the concentration of a compound detected in the blank samples plus 10 times the relative standard deviation. If a compound was not detected in the blanks, the lowest point in the calibration curve showing a signal-to-noise-ratio larger than 8 was reported as the LOQ. Branched PFAS isomers were quantified with the analytical standard corresponding to the linear isomer and are therefore considered to be analysed semi-quantitatively. In order to account for adsorption to surfaces of the experimental setups positive blank samples were evaluated in **Paper IV and V**.

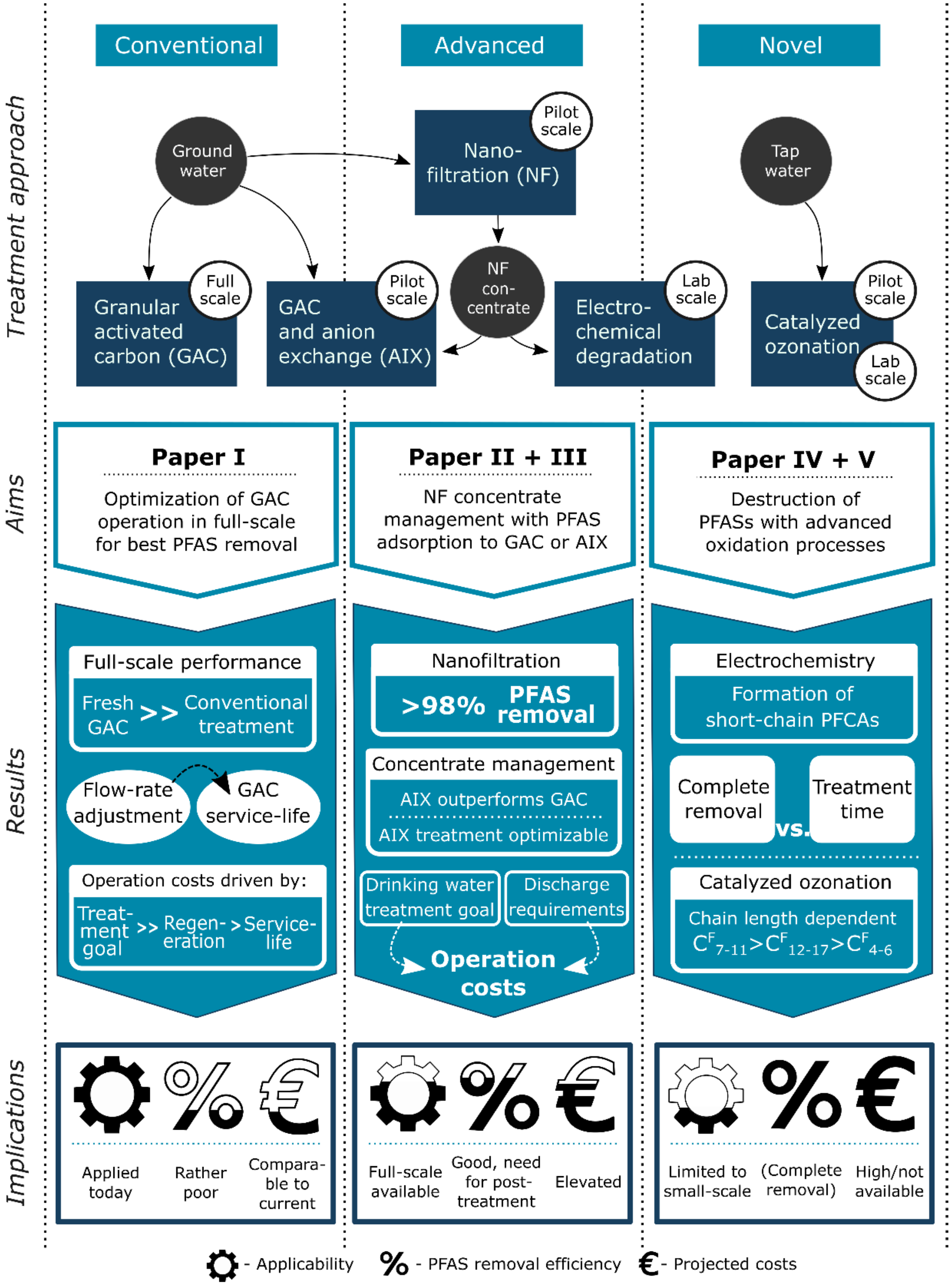
4 Results and discussion

Findings of chapters I-V are presented and discussed in the following section. Figure 6 summarizes the most important details of this thesis.

4.1 PFAS removal in full-scale drinking water treatment (Paper I)

A full-scale DWTP was evaluated for PFAS removal. As can be seen from Figure 7, satisfactory PFAS removal could only be observed for the treatment step involving GAC filtration. This observation confirmed results presented in earlier studies, in which it was found that many conventional water treatment techniques, such as the plants' treatment steps of aeration, softening, dual media filtration, and disinfection with free chlorine, are not able to remove PFASs efficiently (Eschauzier et al., 2012; Appleman et al., 2014; Rahman et al., 2014; Espana et al., 2015). PFAS removal was dependent on the GAC filters' age (i.e. number of bed volumes treated, $BV_{treated}$, as denoted in Figure 7), where young filters showed a higher removal efficiency than older filters. Generally, PFSAs were removed better than PFCAs and the filters showed lower removal efficiencies for homologues with shorter chain lengths (i.e. PFHxA and PFHpA) than for longer chain PFASs. Further, it was observed that linear (L) isomers of PFHxS and PFOS were removed better than the respective branched (B) isomers, which was especially apparent for older filters. For instance, L-PFHxS was removed to 66% compared to 37% removal (B-PFHxS) for a GAC filter with 360 days in operation (29 300 $BV_{treated}$). When comparing the two different carbon materials employed by the treatment plant in two filters of the same age and operation parameters, it was found that the filter comprised of Filtrasorb® 400 performed slightly better (outgoing $\sum_{11}$PFAS concentrations of 2.1 ng L^{-1}) than the AquaSorb® 2000 material (outgoing $\sum_{11}$PFAS= 4.9 ng L^{-1}).

Figure 6 (next page): Summary of this thesis work.


Treatment approach
Aims
Results
Implications
Conventional
Advanced
Novel
Ground water
Granular activated carbon (GAC)
Full scale
GAC and anion exchange (AIX)
Pilot scale
Nano-filtration (NF)
Pilot scale
NF con-centrate
Electro-chemical degradation
Lab scale
Tap water
Catalyzed ozonation
Pilot scale
Lab scale
Paper I
Optimization of GAC operation in full-scale for best PFAS removal
Paper II + III
NF concentrate management with PFAS adsorption to GAC or AIX
Paper IV + V
Destruction of PFASs with advanced oxidation processes
Full-scale performance
Fresh GAC >> Conventional treatment
Flow-rate adjustment
GAC service-life
Operation costs driven by:
Treat-ment goal >> Regen-eration > Service-life
Nanofiltration
>98% PFAS removal
Concentrate management
AIX outperforms GAC
AIX treatment optimizable
Drinking water treatment goal
Discharge requirements
Operation costs
Electrochemistry
Formation of short-chain PFCAs
Complete removal
vs.
Treatment time
Catalyzed ozonation
Chain length dependent
$C^F_{7-11} > C^F_{12-17} > C^F_{4-6}$
Applied today
Rather poor
Compara-ble to current
Full-scale available
Good, need for post-treatment
Elevated
Limited to small-scale
(Complete removal)
High/not available
⚙ - Applicability
% - PFAS removal efficiency
€ - Projected costs

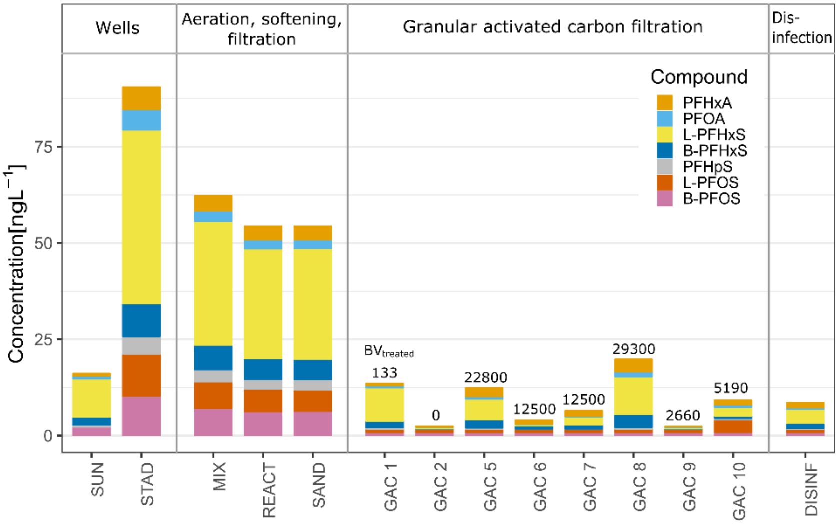


Figure 7: Average concentrations of detected PFASs for the various treatment steps applied in the Bäcklösa drinking water treatment plant. Denotation with L and B refers to the linear and branched isomers of PFHxS and PFOS. SUN = Sunnersta well field; STAD = Stadsträdgården wellfield; MIX = mixed water; REACT = aeration, softening and pH adjustment steps; SAND = dual media filtration; GAC = granular activated carbon; DISINF = disinfection step; $BV_{treated}$ *= amount of treated bed volumes at the time of sampling. Figure reprinted from Belkouteb et al. (2020) with permission.*

While the concentration profile summarized in Figure 7 can be regarded representative for the plants' full-scale performance, it should be noted that it presents the results of grab samples taken during one occasion, and does therefore not provide information on a change in PFAS concentrations over time. Monitoring the long-term performance of the filtration treatment showed a rapid decline in removal efficiency for the detected short chain PFASs (i.e. PFHxA and PFBS). A faster decline was observed for the removal efficiencies of PFSAs compared to PFCAs of the same perfluorcarbon chain length and removal generally followed the order PFOS > PFHxS > PFOA > PFBS > PFHxA, where PFOS was removed to > 80 %, even after 30 000 $BV_{treated}$. By gradually changing the flow-rates through filters of different age, it was observed that PFAS removal in the young filter (63 operation days, 5 725 $BV_{treated}$) was less sensitive to an adjustment in flow-rates than the older filter (264 operation days, 21 971 $BV_{treated}$). For instance, a 10 L s^{-1} flow-rate decrease from 39 to 29 L s^{-1} led to an average increase in total PFAS removal of 6.5 and 14 % for the young and the old filter, respectively. It was hypothesized that adopting a flow-rate strategy could be an economically beneficial option for plant operation. Practically, this would mean that the total life-time of filters could be extended by operating them at a lower flow-rate after a certain treatment goal could not be reached any longer. Different operating scenarios were evaluated, including a hypothetically longer overall life-time of the GAC filters, lower costs for regeneration or adopting the suggested flow-rate strategy. Costs for these scenarios were evaluated for several different treatment goals: total outgoing PFAS concentrations of 10, 25, 50 and 85 ng L^{-1}, respectively. Results highlighted the overwhelming impact of the treatment goal on operational costs. Annual GAC operation costs for reaching the treatment goals of 25, 50, and 85 ng L^{-1} were found to be 42, 70, and 76 % lower, respectively, as compared costs determined for the treatment goal of 10 ng L^{-1}. The results showed further, that adopting the proposed flow-rate strategy would result in the lowest estimated costs for the treatment goal of 50 ng L^{-1} PFAS and that regeneration costs generally had a larger impact on overall costs of the GAC filtration treatment compared to a 10-20 % longer service life or a 20 % lower purchasing price when considering a certain treatment goal. Future research should investigate the proposed flow-rate strategy further and verify the modelled behaviour of PFAS removal over time after changing the flow-rates in aged filters.

4.2 Nanofiltration combined with adsorption materials (Paper II and III)

The NF treatment removed PFASs to > 98 % in both of the studies applying NF considered in this thesis. Next to reduction in PFAS concentrations, a number of other treatment goals were reached with the NF treatment, such as the removal of mineral hardness, uranium-238, DOC and bromide from the raw water. Figure 8 illustrates the removal performance of the NF membrane treatment evaluated in Paper III, in which the NF process was operated at 80 % recovery. 20 % of the feedwater volume resulted in NF concentrate, which was treated further by filtration through adsorption materials in column experiments.

The treatment of NF concentrate by filtration through AIX or GAC highlighted the superior performance of AIX compared to GAC with regard to PFAS removal. In Paper II it was shown, that both materials adsorbed a larger total amount of PFASs from NF concentrate than from the more dilute raw water (Figure 9). The evaluated AIX resin and GAC material removed comparable amounts of PFASs from raw water (4.5 and 4.1 mg L^{-1} adsorption material, respectively). However, for the treatment of NF concentrate, the AIX resin outperformed GAC, where the AIX resin removed 4.1 times more PFAS mass

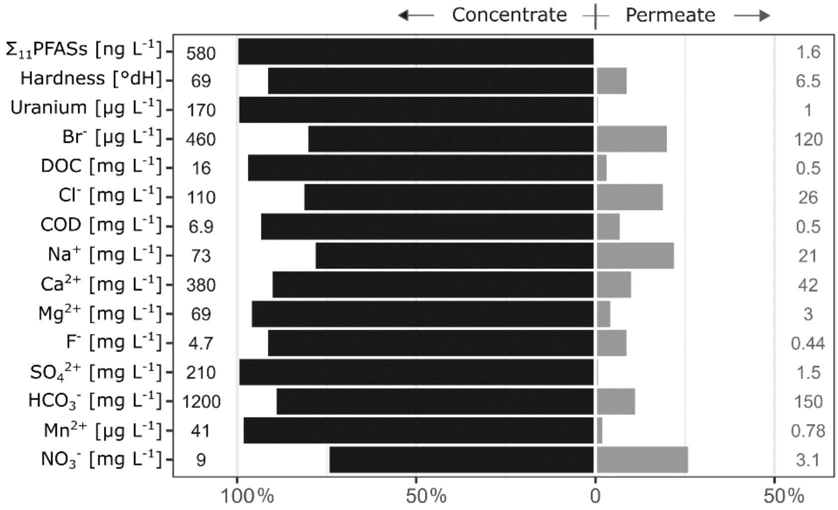


Figure 8: Summary of the removal of various constituents from raw water by the nanofiltration treatment applied in Paper III. The nanofiltration unit was operated at 80% recovery and thus 20% of the feedwater volume resulted in membrane concentrate, while 80% were recovered as permeate and could be used for drinking water production (Figure adopted from Paper III).

from the NF concentrate than from raw water (19 mg L^{-1} adsorption material), while GAC showed a 2.6-fold larger removal of PFAS mass from concentrate than from raw water (10 mg L^{-1} adsorption material). Similar differences between the performance of GAC and AIX were observed in the study described in Paper III. The anion exchange resins Purolite® A600 and Purofine® A694 removed a total PFAS mass of 23 and 37 µg g^{-1} adsorption material from NF concentrate, respectively, while the GAC materials Filtrasorb® 400 and Norit 1240 W removed 15 and 18 µg g^{-1} material, respectively. According to the manufacturers, the resin Purofine® A694, is specifically designed for the adsorption of PFASs. Indeed, results obtained in Paper III showed, that Purofine® A694 had a larger capacity for PFASs than the other AIX resin or the evaluated GAC materials, especially with regard to detected PFSAs.

In a cost calculation, operation costs for the removal of PFASs from the NF concentrate produced in the Bäcklösa DWTP were estimated for various discharge goals and the different adsorption materials evaluated in Paper III. Note, that in Sweden no legally binding regulatory values for the treatment of NF concentrate or the emission of PFAS laden water exist to this day. For the cost calculation, discharge goals were based on different drinking water guidelines issued by agencies throughout Europe and the USA, and some guidelines for PFAS concentrations in ground- and surface water established by

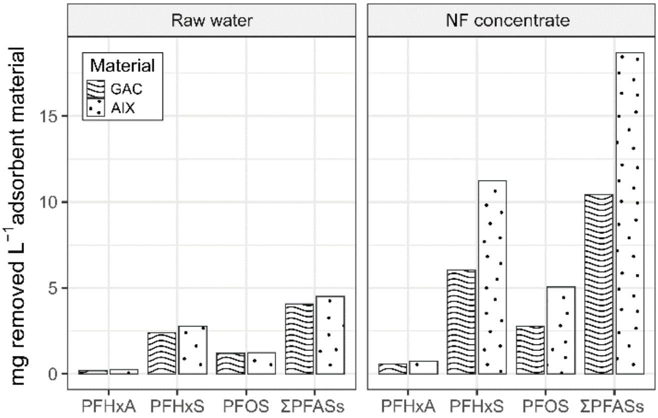


Figure 9: Total PFAS mass adsorbed by the anion exchange resin Purolite® A600 (AIX) and the granular activated carbon material Filtrasorb® 400 (GAC) in the column experiment investigated in Paper II. Total amounts of treated bed volumes were 23 100 for GAC and 18 600 for AIX treating raw water, and 23 100 for GAC and 21 600 for AIX treating nanofiltration (NF) concentrate. Figure adopted from Franke et al. (2019a).

the European Parliament and the Swedish Geotechnical Institute, respectively. Based on the results obtained during the column experiments in Paper III, the amount of bed volumes the absorbents could treat before the treated water reached the designated discharge goal were calculated. Upon reaching this bed volume limit the GAC was assumed to be regenerated, then placed back into operation while the AIX resin was assumed to be sent to incineration and new resin material would be purchased. Operational costs included costs for the purchase of the adsorption materials, transport, and electrical costs for pumping as well as regeneration (GAC only), and incineration (AIX only). Results showed, that the evaluated AIX resins Purolite® A600 and Purofine® PFA694 economically outperformed the GAC materials Filtrasorb® 400 and Norit 1240 W for most discharge goals (Table 2). Despite the differences in costs associated each materials' operation, the results of the cost-analysis thereby reflect observations made when evaluating PFAS adsorption of the materials applied in Paper III. Annual operations unit costs [euro m^{-3} treated drinking water] were determined for NF treatment combined with AIX for treatment of the resulting concentrate. Resulting unit costs were subsequently compared to unit costs connected to the treatment with GAC only, which is applied for PFAS removal at the Bäcklösa DWTP today. Results showed, that at drinking water treatment goals > 4 ng L^{-1} $\sum_{11}$PFAS GAC would provide a more cost efficient drinking water treatment if discharge goals for the NF concentrate are < 85 ng L^{-1} $\sum_{11}$PFAS. Costs for NF combined with AIX treatment of the NF concentrate were, however, found to be comparable to those determined for GAC treatment if discharge goals for NF concentrate are > 85 ng L^{-1} $\sum_{11}$PFAS. This highlights the impact and economic importance of discharge goals when selecting between GAC or NF for PFAS treatment. It should be noted that the NF provides additional economic value, aside from separating PFAS from the feedwater, including removal of i.e. hardness, DOC, and uranium. These benefits were not included in the economic analysis provided in Paper III and should be considered in future work.

Table 2: Estimates of annual operation costs [kiloeuro] arising in the Bäcklösa drinking water treatment plant for the treatment of nanofiltration concentrate when considering the maximum amount of bed volumes the studied adsorption materials could treat before different discharge goals were reached. The cost analysis regards the treatment of 7 million m^3 permeate and a resulting production of 1.8 million m^3 concentrate. No estimations could be made in case respective PFAS concentrations in the water leaving the adsorption columns did not reach the certain discharge goal.

Discharge goal [ng L^{-1}]	**Filtrasorb® 400**	**Norit 1240 W**	**Purolite® A600**	**Purofine® PFA694**
$\sum_{11}$PFAS < 900[a,b]	-	-	-	-
$\sum_{11}$PFAS < 90[a]	398	452	419	357
$\sum_{11}$PFAS < 85[c]	421	475	425	366
$\sum_{11}$PFAS < 50[c]	654	728	799	445
$\sum_{11}$PFAS < 25[c]	1 080	4 210	1 120	1 455
$\sum_{11}$PFAS < 10[c]	1 795	10 600	1 474	2 133
$\sum_{11}$PFAS < 4[d]	5 954	26 064	1 687	2 621
$\sum$ (PFOA, PFOS) < 70[e]	98	95	261	-
$\sum$ (PFOA, PFOS) < 50[f]	216	230	290	-
Any PFAS < 100[g]	98	286	384	-
$\sum$PFAS < 500[g]	14	27	-	-
PFOS < 45[h]	119	198	283	-
PFOS < 0.65[i]	16 919	64 288	543	628

[a] Ankarberg & Lindberg (2016); [b] The nanofiltration concentrate did not reach this concentration; [c] Belkouteb et al. (2020); [d] Lowest concentration produced by the NF in Paper III; [e] USEPA (2016); US EPA (2016); [f] UBA (2019); [g] ECHA (2019), frequently detected PFASs in Paper III were PFHxA, PFOA, PFBS, PFPeS, PFHxS, PFHpS and PFOS; [h] SGI (2015); [i] Annual Average Environmental Quality Standard (AA-EQS) of the EU Water Framework Directive (European Parliament, 2013) .

4.3 Electrochemical degradation (Paper IV)

Electrochemical treatment in bench scale revealed a detrimental effect of DOC on the removal efficiency of PFASs. Over the course of 120 min experiment time concentrations of added PFASs decreased by an average of 77 % (0.95 mA cm^{-2} current density applied), 78 % (4.8 mA cm^{-2}) and 88 % (11.8 mA cm^{-2}) in MilliQ water, as compared to 52 %, 68 % and 80 % in MilliQ water fortified with 11 mg L^{-1} DOC (DOC water), respectively. The impaired removal efficiency of the treatment in DOC water can be explained by competitive reactions between PFAS and DOC molecules on the electrode surface (Sun et al., 2017; Guo et al., 2019; Cui et al., 2020). Longer chain PFASs were removed faster than shorter chain homologues in both MilliQ water and DOC containing water. The observed formation of PFPeA and PFHpA, which were not initially present in the solutions, suggested the degradation of PFASs via a reaction circle of direct electron-transfer on the anode and subsequent decarboxylation / desulfonation, followed by defluorination and hydrolysis leading to a step-wise unzipping of the fluorinated carbon chain and formation of shorter carboxylates (Zhuo et al., 2011; Niu et al., 2016). The removal of PFASs followed first-order reaction kinetics. First-order reaction rate constants *k* were determined to be between -0.26 h^{-1} (PFBA in MilliQ water) and 11 h^{-1} (PFHxDA in DOC water), see Figure 10. Rate-constants followed a trend towards increasing values for *k* (i.e. faster decline in concentrations) with increasing perfluorocarbon chain length. One exception was PFHxDA, which did not follow this trend consistently.

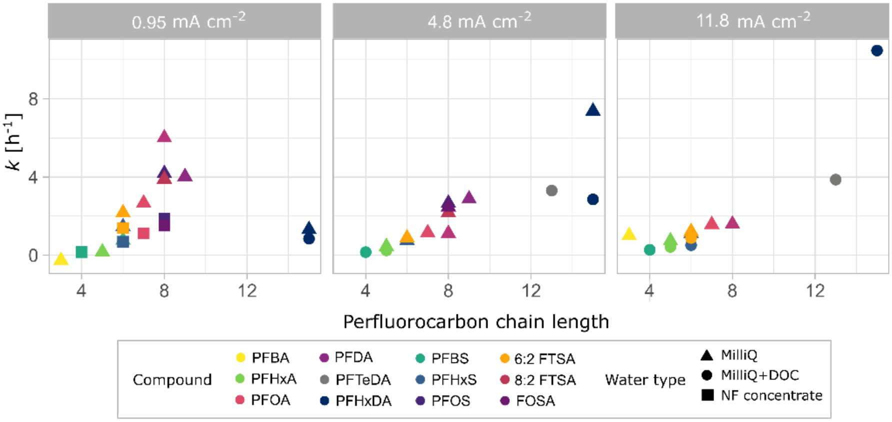


Figure 10: First-order rate constants k [h^{-1}] as a function of perfluorocarbon chain length for the treatment of MilliQ water and MilliQ water containing 10.8 mg L^{-1} dissolved organic carbon (MilliQ+DOC) and nanofiltration (NF) concentrate (Figure adopted from Paper IV).

Shorter chain PFASs were observed to have slower reaction rates, which is in line with results reported earlier (Zhuo et al., 2012; Schaefer et al., 2017). A comparison of the obtained rate constants with other studies remains difficult, since rate constants are dependent on initial PFAS concentrations, water quality parameters and experimental setup (Zhuo et al., 2012).

NF concentrate was treated at a current density of 0.95 mA cm^{-2}. Highest and fastest removal (> 98 % removed after 180 minutes of treatment) was observed for the detected precursor compounds (6:2 FTSA and FOSA) followed by PFOS (98 % removal) and PFOA (94 %). L-PFHxS (85 %), B-PFHxS (66 %) and PFBS (26 %) were effected to a lesser extent and PFHxA and PFHpA concentrations increased during the 180 minute experiment. Generally, removal of PFASs from NF concentrate at 0.95 mA cm^{-2} current density showed similar trends as observed in the experiments treating MilliQ water and DOC containing MilliQ water. Note, that the NF concentrate contained lower PFAS concentrations than the PFAS fortified water in the other experiments. This may have caused a limited mass-transport to the electrodes in the experiment treating NF concentrate compared to the experiments treating fortified MilliQ and DOC water leading to lower observed rate constants k as illustrated in Figure 10. Additionally, the water quality differed substantially between the evaluated water matrices, and direct comparisons should therefore be made with care.

An estimation of the energy consumed per ng PFAS removed revealed a beneficial effect of the DOC content on the treatment, with less energy consumed per ng PFAS removed in the experiments considering DOC water as compared to MilliQ water.

4.4 Heterogeneously catalyzed ozonation (Paper V)

Laboratory scale experiments applying ozone, the catalytic material and persulfate in all possible combinations on fortified MilliQ water resulted in significant PFAS removal only for ozone applying trials. Observed removal was independent of the PFAS functional group and increased with increasing perfluorocarbon chain length of the compounds. This, despite the fact that isotherms determined prior the experiments showed the strongest adsorption to the catalytic material for both short fluorinated chains $C^F{}_3$-$C^F{}_4$ and longer chain PFASs $C^F{}_{10}$-$C^F{}_{11}$ compared to the compounds with medium long chains $C^F{}_5$-$C^F{}_9$. As argued by the manufacturers, the treatment system is designed to degrade organic micropollutants mainly with the help of ozone and hydroxyl radicals (Joos Lindberg, 2016). Previous research has shown, that both ozone and hydroxyl radicals are unable to degrade PFASs (Dombrowski et al., 2017). Any degradation must therefore involve other reactive species. It is recognized that

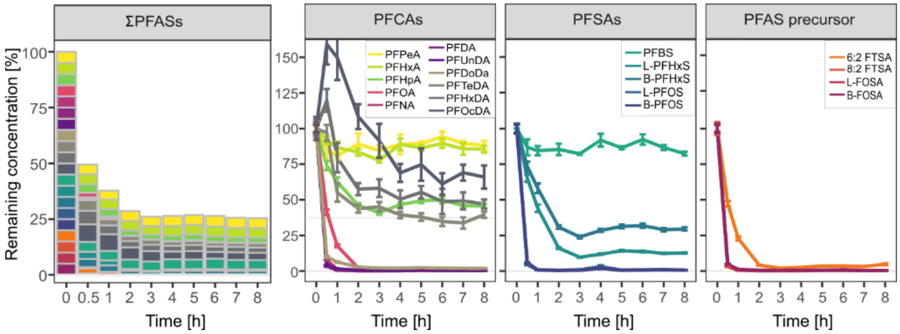


Figure 11: Remaining concentrations [%] of the individual PFASs in tap water in the pilot-scale plant applying heterogeneously catalyzed ozonation. Figure reprinted from Franke et al. (2019b) with permission.

indirect free radical reactions participate in a pool of reactions assumed to occur in the system, where ozone reacts with hydroxide ions to form different reactive oxygen species including radical species such as $O_2^{\bullet-}$, $O_3^{\bullet-}$, HO_3, and HO_4 (Lim & Chidsey, 2005). In persulfate systems highly oxidative persulfate radicals are produced, which have been shown to effectively degrade certain PFASs (Hori et al., 2005, 2008; Wang et al., 2010). Indeed, of the ozone applying trials evaluated in laboratory-scale the combination of all three parameters (ozone, catalyst and persulfate) was more successful in PFAS removal than applying ozone only (47 % vs. 22 % total PFAS removal after 120 min treatment time, respectively). The combination of ozone and persulfate or ozone and catalyst, led to 24 % and 26 % total PFAS removal after 120 min, respectively.

In the pilot-scale system rapid removal was observed for ∑PFASs in fortified tap water within the first 30 minutes of the ozonation experiment (Figure 11). Removal of the total PFAS concentration of up to 70 % was observed within the first three hours of the experiment, after which the concentrations remained stable throughout the remaining five hours. Highest removal was observed for PFASs with medium chain lengths C^F_7-C^F_{11} (> 98 %), followed by longer chain compounds C^F_{12}-C^F_{17} (64 %) and shorter chain homologues C^F_4-C^F_6 (55 %). This trend was consistent for all PFASs evaluated and independent of PFAS functional group. The high removal observed for the PFASs with medium long chains, C^F_7-C^F_{11}, did not correspond to the adsorption behaviour observed in the isotherm studies. It is therefore hypothesized that PFAS removal is not dependent on the binding strength of the compounds to the catalytic material but rather on the type of interaction with the reactive species created in the water phase.

Overall, the obtained results suggest that the existing commercially available treatment using heterogeneously catalyzed ozonation can be an effective treatment techniques for PFASs in water, especially if PFAS contamination dominantly consists of PFASs with perfluorinated chain-lengths of C^F_7-C^F_{11}. Further studies should investigate degradation products, including the formation of potentially harmful by-products, the feasibility of applying the treatment on various water types, and the occurrence of PFASs and degradation products in the gas-phase.

5 Conclusions and outlook

The main conclusions of this thesis are:

1. Filtration through young GAC material is one of the few treatment techniques in conventional drinking water treatment capable of removing a broad range of PFASs from water. Filtration efficiency and cost-effectiveness can be improved by adopting a flow-rate strategy.
2. Filtration through NF membranes presents one of the safest water treatment technique today and was able to remove more than 98 % of the detected PFASs from contaminated groundwater. Membrane concentrate can be efficiently treated with adsorption to GAC or AIX resins and the cost-effectiveness of each material is dependent on the treatment goal for the concentrate.
3. Electrochemical degradation is a powerful treatment technique for PFAS contaminated water, including NF concentrate. Treatment efficiency is highly dependent on the water type and future research is needed to evaluate the process on a larger scale for optimal cost-benefit.
4. Heterogeneously catalysed ozonation presents a potential technique for the treatment of water contaminated with PFASs containing fluorinated carbon chains with 7 - 11 carbon atoms.

The adopted treatment goal for both the final drinking water and the treatment of NF concentrate was shown to be the key factor determining overall treatment costs, which highlights the impact of a change in PFAS regulatory levels on operation costs. Further, it should be emphasized that existing regulatory guidelines focus on a specific subgroup of PFASs, often only a single compound, which can be problematic as PFAS contamination often occurs with complex mixture of compounds and many of the previously disregarded PFASs have been shown to be connected to adverse health effects at a later stage.

As emphasized in the different studies included in this thesis, the definite effectiveness of the investigated treatment techniques needs to be evaluated for each specific source water before application in full-scale drinking water treatment, which can clearly be regarded a disadvantage. However, the general working principles and tools provided in this thesis give drinking water providers and researchers valuable insights in the performance of the treatment methods. Trends observed in the presented studies can be expected to coincide for different source waters.

There are numerous ways to continue the search for integrated, cost-efficient treatment methods for the removal of PFASs in drinking water treatment. While the techniques discussed in this thesis show high potential, there is much left to investigate regarding the improvement of their treatment efficiency and cost-effectiveness. The pressure on drinking water providers to control an ever increasing number of chemical contaminants with ever decreasing guideline values can be expected to become more apparent in the future. NF presents an exceptionally safe treatment option, which does not rely on the addition of difficult-to-handle chemicals and is easy to operate. NF concentrate can be treated by adsorption materials like GAC or specifically designed AIX resins or by degradation techniques. Apart from an extended investigation of suitable treatment techniques, different analytical methods than the ones applied in this thesis could provide more comprehensive information regarding the initial composition of the samples as well as intermediate and final reaction products. With total organic fluorine measurements one could gain conclusions on the initial composition of natural samples regarding their organofluorine content. Ion chromatography and gas chromatography could be applied to confirm PFAS mineralization. The analysis of samples with high-resolution mass spectrometry presents another powerful method for a much more extended investigation of the chemical composition of natural samples. Analysing the content of evolved gasses and inorganic ions could benefit analysis further. Today, many of the aforementioned analytical techniques comprise comparatively high limits of quantification, which hampers the analysis of raw and treated drinking water with these methods. However, as technological achievements have occurred rapidly during the past decades, it is likely that these analytical techniques evolve to become more applicable for the analysis of drinking water in the near future. Additional to monitoring PFASs and other compounds in chemical analysis, one should consider the application of integrated strategies for the measurement of toxicological response, such as *in vivo* and *in vitro* methods. This applies especially to degradation techniques and it is strongly encouraged that future work on degradation methods applied on natural waters investigates the formation and effects of potentially toxic by-products.

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Popular science summary

Have you ever thought about how luxurious it is to get drinking water right from the tap?

Clean and safe drinking water is something everybody should have access to. But in our modern world natural waters become more and more influenced by human activities and we need to keep an eye on how clean our water actually is. We need to make sure the water is free from color, bad smells or taste but also from invisible things like viruses, bacteria and toxic chemicals. There is a group of chemicals called PFASs (per- and polyfluoroalkyl substances), which is of special concern for our drinking water. Even if you haven't heard of PFASs, you most certainly have been in contact with them, since they are used in all kinds of products, like rain jackets, Teflon pans, microwave popcorn bags, cosmetics and fire-fighting foams. They are actually quite amazing chemicals, since they can give all these products their convenient (and even life-saving) properties. But what happens when they reach the environment? Then they become a big problem, especially because they tend to end up in our drinking water. Methods we usually use to clean our water are not designed to remove PFASs. Researchers have found that many PFASs cause cancer or contribute to other health issues in animals and humans, which is why many countries regultare how much PFASs are allowed in drinking water. Drinking water providers need to know more about the methods they can use to remove PFASs from water.

In my studies I looked at different treatment methods, both in small experiments in the laboratory and in large installations in a real drinking water treatment plant. Filtration through certain carbon materials or specially designed resins can remove PFASs because they attach to the surface of those materials. After the filter materials are full, they can be burnt, which at the same time destroys PFASs. Other components in the water, however, also stick to the materials and compete with the PFASs, which leads to that they are not being removed very efficiently. The filtration materials have to be exchanged quite often. A technique called membrane filtration can remove all kinds of things we do not want in the water, even small molecules like PFASs.

Membranes (like the ones used in nanofiltration or reverse osmosis) are thin sheets with tiny holes in them, so tiny that only molecules like water can pass and anything bigger than the size of the holes will be filtered out. Nanofiltration can remove PFASs and other unwanted components from water and the water passing the membrane can be used for making drinking water. But there is a disadvantage - the materials that are filtered out have to end up somewhere and nanofiltration produces roughly 20 % of "rejected" water containing everything that did not pass the membrane. Usually, if this water only contains natural material, it can be left into a nearby river or lake. But if the water contains PFASs, as is the case in Uppsala, we do not want to send out into the environment. I therefore looked into the possibility to combine nanofiltration with the adsorption materials mentioned above. It turns out that the materials are better at taking up PFASs from this rejected nanofiltration water than when filtering raw water directly. Even though nanofiltration is rather energy consuming, we should definitely consider a switch to membrane filtration a good investment, as it filters out all kinds of things we don’t want in the water (maybe even harmful chemical compounds we have no clue about yet).

I also tested some methods that are supposed to destroy PFASs. The nice thing with those is that one could destroy the PFASs on the spot without creating any waste. One of the destruction methods I tested is called electrochemical treatment, where water is treated with an electrical current. This can start chemical reactions in the water leading to the destruction of PFAS molecules. I tested this for the nanofiltration reject water and it showed that you can actually remove quite a good amount of PFASs. In another destruction method ozone gas is introduced to the water. In combination with a solid iron material, the ozone molecules can create very reactive chemical species that can attack PFASs and destroy them. This worked for a certain group of PFASs, which also happens to be the PFAS molecules we most often find in the environment meaning this treatment, could be useful for some areas. This seems promising but these destruction methods are still in the starting phase of being developed for larger scale. More work needs to be done on building and testing them in a large drinking water treatment plant. All in all, it is possible but demanding to make sure our water is as perfectly clean as we expect it to be when we open our tap.

So I hope next time you enjoy a nice glass of water you will be extra happy. Knowing that it actually is quite a lot of work to produce tap water free from harmful chemicals, we should really all put a smile on our faces when we enjoy this fantastic luxury.

Populärvetenskaplig sammanfattning

Har du någonsin funderat på hur lyxigt det är att få dricksvatten direkt från kranen?

Rent och säkert dricksvatten är naturligtvis något alla borde ha tillgång till. Men i vårt moderna samhälle påverkas vattendrag och grundvatten mer och mer av mänskliga aktiviteter och vi måste hålla ett öga på hur rent vårt vatten faktiskt är. Vi måste se till att vattnet är fritt från färg, dålig lukt och smak men även från osynliga saker som virus, bakterier och giftiga kemikalier. Det finns en grupp kemikalier som kallas PFAS (per- och polyfluoralkylämnen), som är särskilt knepigt för vårt dricksvatten. Även om du inte har hört talas om PFAS tidigare så har du säkert varit i kontakt med dem, eftersom de används i alla möjliga typer av produkter, så som regnjackor, teflonpannor, micropopcornpåsar, kosmetika och brandbekämpningsskum. De är faktiskt ganska fantastiska kemikalier eftersom de kan ge alla dessa produkter sina praktiska (och till och med livräddande) egenskaper. Men vad händer när de kommer ut i miljön? Plötsligt blir de till ett stort problem, speciellt för att de tenderar att också hamna i vårt dricksvatten. Metoder som vi brukar använda för att rena vårt vatten är inte utformade för att ta bort PFAS. Forskare har funnit att många PFAS är cancerframkallande eller bidrar till andra hälsoproblem hos djur och människor, vilket har lett till att många länder över hela världen börjat reglera hur mycket PFAS som får vara i dricksvatten. Dricksvattenleverantörer behöver därför veta mer om de metoder de kan använda för att ta bort PFAS från vatten.

I mina studier tittade jag på olika behandlingsmetoder, både i små experiment i laboratoriet och i stora installationer i ett riktigt dricksvattenreningsverk. Filtrering genom vissa kolmaterial eller specialdesignade hartser kan ta bort PFAS eftersom de fäster på ytan på dessa material. När filtermaterialet är fullt kan de brännas, vilket samtidigt förstör PFAS. Andra komponenter i vattnet håller dock också fast vid materialen och konkurrerar med PFAS, vilket leder till att de inte tas bort särskilt effektivt. Filtreringsmaterialen måste bytas ut ofta.

En teknik som kallas membranfiltrering kan ta bort alla olika slags ämnen vi inte vill ha i vattnet, även små molekyler som PFAS.

Membran (som de som används vid nanofiltrering eller omvänd osmos) är tunna ark med små hål i sig, så små att endast de små vattenmolekylerna kan passera och allt större än porernas storlek filtreras bort. Nanofiltrering lyckas ta bort PFAS och andra oönskade komponenter från vatten. Vattnet, som passerar nanofiltreringsmembranet, kan användas för att göra dricksvatten. Men det finns en nackdel - ämnena som filtreras bort måste såklart ta vägen någonstans. Nanofiltrering producerar ungefär 20 % ”avvisat” vatten som innehåller allt som inte passerar membranet. Om detta koncentrerade vattnet bara innehåller naturligt material kan det vanligtvis lämnas i en närliggande flod eller sjö. Men om detta vattnet innehåller PFAS, som i Uppsala, är det här vattnet fullt med PFAS som vi inte vill släppa ut i miljön igen. Jag undersökte därför möjligheten att kombinera nanofiltrering med adsorptionsmaterialen som nämnts ovan. Det visar sig att materialen är bättre på att ta upp PFAS från koncentratvatten än om man filtrerar grundvatten direkt. Även om nanofiltrering är ganska energikrävande, bör vi definitivt betrakta en övergång till membranfiltrering som en bra investering, eftersom den filtrerar bort alla slags saker vi inte vill ha i vattnet (kanske till och med skadliga kemikalier som vi ännu inte har någon aning om) . Jag testade också några metoder som ska förstöra PFAS. Det fina med dessa är att man kan förstöra PFAS på plats utan att skapa något avfall. En av nedbrytningsmetoderna jag testade kallas elektrokemisk behandling, där man behandlar vatten med en elektrisk ström. Detta kan starta kemiska reaktioner i vattnet som leder till att PFAS förstörs. Jag testade detta för bland annat koncentratvatten från membranprocessen och det visade att man efter en viss tid kan ta bort en hel del PFAS. En annan nedbrytningssmetod använder sig av ozongas, som man tillför vattnet. I kombination med ett järnmaterial kan ozonmolekylerna skapa mycket reaktiva kemiska föreningar som kan reagera med PFAS och förstöra dem. Detta fungerade för en viss grupp av PFAS, vilket också råkar vara de PFAS-molekyler som vi oftast hittar i miljön. Det betyder att denna behandling kan vara användbart i vissa områden. De utvärderade nedbrytningssmetoderna verkar lovande men de befinner sig fortfarande i utvecklungsfasen. Mer arbete behövs för att kunna installera dem i ett stort vattenverk.

Allt som allt är det möjligt men krävande att se till att vårt vatten är lika perfekt som vi förväntar oss när vi öppnar kranen. Så jag hoppas du blir extra glad nästa gång du njuter av ett gott glas vatten. När vi vet att det faktiskt är en hel del jobb att producera kranvatten utan skadliga kemikalier, så borde vi alla sätta ett leende på läpparna när vi njuter av denna fantastiska lyx.

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ACTA UNIVERSITATIS AGRICULTURAE SUECIAE

DOCTORAL THESIS NO. 2020:40

Per- and polyfluoroalkyl substances (PFASs) are a group of persistent chemicals polluting water sources worldwide. Within this thesis work conventional and novel treatment methods were evaluated for the removal of PFASs in drinking water production. Filtration through granular activated carbon could be optimized by adopting flow-rates. Combining nanofiltration with filtration through adsorption materials presented a robust and secure treatment. Electrochemical treatment and catalyzed ozonation might be a solution for destroying PFASs on-site, while technical feasibility and by-product formation need to be investigated further.

Vera Franke received her doctoral education at the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences. She received her master's degree in environmental chemistry at Stockholm University and her bachelor's degree in chemistry at the Karlsruhe Institute of Technology.

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