Thesis for the Degree of Doctor of Philosophy

PFAS in Managed Aquifer Recharge

Solution or hidden risk in drinking water production?

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To mom & dad, thank you for always encouraging me.

"[PFAS sind] wie Glitzer:
erstmal ganz toll, aber landen auch
überall, wo man es nicht braucht
- und das für immer."

Mai Thi Nguyen-Kim, 2026

Abstract

Managed Aquifer Recharge (MAR) is a globally implemented, cost-effective method to mitigate groundwater depletion while simultaneously meeting the need for higher quantities of groundwater during drinking water production with increasing populations and climate change effects. Through MAR, surface waters from rivers and lakes artificially infiltrate into groundwater systems before being pumped up for further drinking water treatment. However, surface waters contain variable concentrations of the harmful per- and polyfluoroalkyl substances (PFAS). The amphiphilic, persistent, and highly mobile nature of PFAS distinguishes their behavior from that of many traditional persistent organic contaminants, raising concerns that PFAS end up in ground- and drinking water systems during MAR, thus posing a risk for carcinogenicity and infertility to humans. The aim of this thesis was therefore to assess whether MAR acts as a source or sink for PFAS into groundwater systems and eventually drinking water supplies by (1) identifying periods of increased risk of PFAS infiltration by monitoring surface water PFAS variations, (2) understanding PFAS fate and transport in the subsurface at low ambient concentrations, as well as (3) assessing possible optimization strategies to meet upcoming stricter PFAS drinking water guidelines. Therefore, a two-year monitoring campaign at two operating Swedish MAR sites, saturated column experiments, as well as a pilot study of foam fractionation as potential pre-treatment to MAR, were conducted.

The key finding of this thesis is that MAR can act both as a source and temporary sink for PFAS. This thesis further shows that other environmental proxies in surface waters cannot predict PFAS variations that are instead highly site-specific and depend on local conditions and PFAS point sources. PFAS transport and retardation within MAR systems are governed by complex interactions between sorption processes, aquifer heterogeneity, and groundwater level fluctuations, which can promote both temporary retardation and later remobilization of PFAS. Here, long-term monitoring of stable water isotope ($\delta^{18}O$) signals infiltrated via MAR provide valuable insight into water flow velocities, mixing processes, and PFAS transport mechanisms.

The findings highlight the need for integrated management approaches that combine appropriate (pre-)treatment technologies with strategic MAR design and operation to produce drinking water that meets increasingly stringent current and upcoming PFAS guidelines. The results therefore support the design, optimization, and expansion of MAR systems while minimizing the input of PFAS into artificially recharged groundwater systems and drinking water supplies.

Keywords: Per- and polyfluoroalkyl substances (PFAS), Managed Aquifer Recharge (MAR), drinking water, fate and transport, sorption, groundwater, infiltration

Svensk summering

Konstgjord grundvattenbildning (MAR) är en metod som tillämpas globalt och som på ett kostnadseffektivt sätt motverkar grundvattenbrist, samtidigt som den möter behovet av större mängder grundvatten till dricksvattenproduktion i samband med en växande befolkning och större klimatförändringar. MAR innebär att ytvatten från vattendrag och sjöar infiltreras i grundvattensystemen innan det pumpas upp för dricksvattenproduktion. Ytvattnet innehåller dock varierande koncentrationer av de skadliga per- och polyfluorerade alkylsubstanserna (PFAS). PFAS amfifila, svårnedbrytbara och mycket mobila egenskaper skiljer deras beteende från många traditionella persistenta organiska föroreningar, varför PFAS antas hamna i grund- och dricksvattensystemet under MAR, vilket utgör en risk för cancer och infertilitet hos människor. Syftet med denna avhandling var därför att utvärdera om MAR fungerar som en källa eller en sänka av PFAS i grund- och dricksvattensystem genom att (1) identifiera perioder med ökad risk för PFAS infiltration genom övervakning av variationer i PFAS-halter i ytvatten, (2) förstå öde och transport av låga PFAS-koncentrationer i marken och grundvattensystem, samt (3) utvärdera möjliga optimeringsstrategier för att uppfylla kommande striktare PFAS-gränsvärden för dricksvatten. Därför genomfördes en tvåårig övervakningskampanj vid två svenska MAR-anläggningar, mättade kolonnexperiment samt en pilotstudie av skumfraktionering som potentiell förbehandling inför MAR.

Den viktigaste slutsatsen i denna avhandling är att MAR kan fungera både som källa och tillfällig sänka för PFAS. Avhandlingen visar också att andra miljöindikatorer i ytvatten inte kan förutsäga variationer i PFAS-halterna, som i stället beror på lokala förhållanden och punktkällor. Transport och fördröjning av PFAS inom MAR-system styrs av komplexa interaktioner mellan sorptionsprocesser, akvifers heterogenitet och fluktuationer i grundvattennivån, vilket kan leda till både tillfällig fördröjning och senare remobilisering av PFAS. Långsiktig övervakning av signaler från stabila vattenisotoper ($\delta^{18}O$) som infiltreras via MAR ger värdefulla insikter i vattenflödeshastigheter, blandningsprocesser och transportmekanismer för PFAS.

Resultaten belyser behovet av integrerade strategier som kombinerar lämpliga (för-) behandlings- och reningstekniker med strategisk design och drift av MAR-system för att producera dricksvatten som behöver uppfylla allt striktare PFAS riktlinjer. Resultaten kan användas till att bättre utforma, optimera och utvidga MAR-system samtidigt som att minimerar tillförseln av PFAS-koncentrationer till konstgjord grundvatten och dricksvattenresurser.

Keywords: Per- och polyfluorerade alkylsubstanser (PFAS), konstgjord grundvattenbildning (MAR), dricksvatten, öde och transport, sorption, grundvatten, infiltration

List of Publications

This doctoral thesis is based on the following studies, referred to in the following by their Roman numbers.

I Mumberg, T., Ahrens, L. and Wanner, P. (2024). Managed aquifer recharge as a potential pathway of contaminants of emerging concern into groundwater systems – A systematic review, *Chemosphere* 364:143030. doi: 10.1016/j.chemosphere.2024.143030.

II Mumberg, T., Ahrens, L., Kothawala, D., McCleaf, P., Pott, B.-M., Rehnstam, S. and Wanner, P. (2026). Seasonal trends of PFAS concentrations in surface waters and fate during Managed Aquifer Recharge (MAR). *Journal of Hazardous Materials Advances.* 23:101340. doi: 10.1016/j.hazadv.2026.101340.

III Mumberg, T., McCleaf, P., Ahrens, L., Franke, V. and Wanner, P. (in review). Resolving PFAS transport in groundwater requires resolving hydrology: Evidence from Managed Aquifer Recharge systems. *Environmental Science & Technology.*

IV Mumberg, T., Nivall, F., Ahrens, L., Pott, B.-M., McCleaf, P., Chakraborty, S., Franke, V., Kothawala, D. and Wanner, P. (manuscript in preparation). Concentration-dependent PFAS transport during managed aquifer recharge (MAR) - insights from laboratory column experiments.

V Mumberg, T., McCleaf, P., Ahrens, L., Ekesiöö, O., Franke, V., Arebratt, R., Rang, S. and Wanner, P. (manuscript in preparation). Treatment of low ambient PFAS concentrations in surface water by foam fractionation prior to managed aquifer recharge (MAR).

The contribution of Tabea Mumberg to the papers in this thesis was as follows:

I Development of the search string and conduction of the literature research, data retrieval and analysis, and compiling of the results. Writing of the original manuscript as well as editing and revision of the manuscript.

II Coordination of the monitoring program and sampling, as well as pre-concentration, extraction and analysis of the PFAS samples. Analysis, compiling and visualization of the data and statistical analysis. Writing of the original manuscript as well as editing and revision of the manuscript.

III Coordination of the sampling outside of the waterworks standard monitoring program, as well as pre-concentration, extraction and analysis of the PFAS samples. Analysis, compiling and visualization of the data and statistical analysis. Writing of the original manuscript as well as editing and revision of the manuscript.

IV Planning and set-up as well as conducting the column experiments together with the bachelor student Frida Nivall (acting as her co-supervisor). Sampling, and pre-concentration, extraction and analysis of the PFAS samples. Analysis, compiling and visualization of the data and statistical analysis. Writing of the original manuscript as well as editing and revision of the manuscript.

V Involvement in planning the pilot study together with the waterworks. Pre-concentration, extraction and analysis of the PFAS samples. Analysis, compiling, and visualization of the data. Writing of the original manuscript as well as editing and revision of the manuscript.

Other publications not included in this thesis:

1 Moghadasi, R., Mumberg, T. and Wanner, P (2023). Spatial prediction of concentrations of per-and polyfluoroalkyl substances (PFAS) in European soils. *Environmental Science & Technologies Letters* 10, 11, p. 1125–1129. doi: 10.1021/acs.estlett.3c00633.

2 Mumberg, T., Beck, A. J., Martínez-Cabanas, M., Gosnell, K. J., Gledhill, M. and Achterberg, E. P. (2026). Low levels of munition compounds in commercially-available seafood from the North Sea and Baltic Sea pose limited risk to human seafood consumers. *Marine Pollution Bulletin* 225, 119281.
doi: 10.1016/j.marpolbul.2026.119281.

Acronyms

AFFF	Aqueous film forming foam
CECs	Contaminants of emerging concern
δ^{18}O and δ^{2}H	Stable water isotope ratios
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
ECHA	European Chemicals Agency
EEA	European Environment Agency
EU	European Union
FF	Foam fractionation
GAC	Granular activated carbon
HDPE	High-density polyethylene
IS	Internal Standard
K_d	Soil adsorption distribution coefficient
K_{oc}	Soil-water partitioning coefficient
K_{ow}	Octanol-water partitioning coefficient
LC-MS/MS	Liquid chromatography tandem mass spectrometer
MAR	Managed aquifer recharge
MDL	Method detection limit
NMDS	Non-metric Multi Dimensional Scaling
OECD	Organisation for Economic Co-operation and Development
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluoroalkyl carboxylic acid
PFSA	Perfluoroalkyl sulfonic acid
PVC	Polyvinyl chloride
R	Retardation factor
REACH	Registration, Evaluation, Authorisation, and Restriction of Chemicals
US EPA	United States Environmental Protection Agency
VSMOW	Vienna Standard Mean Ocean Water

List of PFAS (by group and chain length)

PFBA	Perfluorobutanoic acid
PFPeA	Perfluoropentanoic acid
PFHxA	Perfluorohexanoic acid
PFHpA	Perfluoroheptanoic acid
PFOA	Perfluorooctanoic acid
PFNA	Perfluorononanoic acid
PFDA	Perfluorodecanoic acid
PFUnDA	Perfluoroundecanoic acid
PFDoDA	Perfluorododecanoic acid
PFTriDA	Perfluorotridecanoic acid
PFTeDA	Perfluorotetradecanoic acid
PFBS	Perfluorobutanesulfonic acid
PFPeS	Perfluoropentanesulfonic acid
PFHxS	Perfluorohexanesulfonic acid
PFHpS	Perfluoroheptanesulfonic acid
PFOS	Perfluorooctanesulfonic acid
PFNS	Perfluorononanesulfonic acid
PFDS	Perfluorodecanesulfonic 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
FOSA	Perfluorooctanesulfonamide
MeFOSAA	N-Methylperfluorooctanesulfonamido acetic acid
EtFOSAA	N-Ethylperfluorooctanesulfonamido acetic acid
HFPO-DA	Hexafluoropropylene oxide-dimer acid (GenX)
NaDONA	4,8-dioxa-3H-perfluorononanoic acid
9Cl-PF3ONS	9-chloro-hexadecafluoro-3-oxanonane sulfonic acid
11Cl-PF3OUdS	11-chloro-eicosafluoro-3-oxaundecane-1-sulfonic acid
PFECHS	Perfluoroethyl-cyclohexane sulfonic acid

Contents

Part I

Overview

CHAPTER 1

Introduction

1.1 Context & objectives

Drinking water is an important good and about half of the global drinking water is produced from groundwater (Svenskt Vatten, 2019; UNESCO World Water Assessment Programme, 2022). However, the need for drinking water increases with rising populations and global development, while groundwater levels decrease due to higher water use and warmer climate (Taylor et al., 2013). Additionally, even though groundwater makes up 96-99% of the globally available freshwater sources (Cross et al., 2016; Gleeson et al., 2016; Langbein and Office, 1947; Postigo and Barceló, 2015), it is not always available where drinking water sources are needed. In Sweden, among other countries, local geology results in few aquifers and only 25% of the drinking water production originates from natural groundwater (Svenskt Vatten, 2019). Due to these factors, using surface water resources for recharging groundwater

reservoirs or for direct drinking water production has become more relevant during recent years (Dillon et al., 2019; Taylor et al., 2013). A common process that produces “artificially produced groundwater” (Svenskt Vatten, 2019) from surface water, is called managed aquifer recharge (MAR). During MAR, surface water is infiltrated into groundwater systems to recover, secure, and maintain groundwater systems artificially or to improve water quality. It has become increasingly recognized since the 1970s and will likely be even more relevant for future water management strategies (Dillon et al., 2019).

However, every day, about 15 000 new chemical compounds are entering the market with a variety of purposes and characteristics (Escher et al., 2020; Wang et al., 2024), and many chemical contaminants eventually end up in surface and groundwater systems leading to only 41.5% of the European Union (EU) water bodies being considered of good chemical status and 14.5% of the EU groundwater bodies being labeled as "poor" in chemical status (European Environment Agency (EEA), 2022). Only a fraction of these compounds is regulated throughout one year due to regulations being based on single compound risk assessments and time-consuming processes (Escher et al., 2020). Many of these chemicals therefore classify as so-called contaminants of emerging concern (CECs), which, according to the United States Environmental Protection Agency (US EPA), are *"pollutants not currently included in routine monitoring programs and [that] may be candidates for future regulation depending on their (eco)toxicity, potential health effects, public perception, and frequency of occurrence in environmental media"* (Smith, 2008). CECs include a variety of compounds such as plastics, certain pesticides, pharmaceuticals and personal care products, endocrine disrupting compounds, or per- and polyfluoroalkyl substances (PFAS), among others. CECs are often anthropogenic compounds that are widely-spread in the environment, very persistent and hardly degrading (Ahmed and Marhaba, 2017; Evich et al., 2022; Gobelius et al., 2018; Malnes et al., 2022; Scheringer et al., 2022), as well as can often, already at ng L^{-1} concentrations, cause severe health effects in humans and to the environment (Smith, 2008; Wang et al., 2024).

Only a few CECs, such as certain pesticides or some PFAS, have been regulated in the past decade (Wang et al., 2024) and drinking water guidelines currently approach low ng L^{-1} levels, especially for contaminants within the group of PFAS (Gobelius et al., 2018; Livsmedelverket, 2022; Miljøministeriet,

2024; United States Environmental Protection Agency, 2024). However, an understanding of the fate and transport of these low ng L^{-1} PFAS concentrations is lacking and recent evidence indicates that surface waters used for MAR may contain elevated concentrations of PFAS. These could then enter groundwater systems and drinking water supplies through MAR, particularly because these systems were not originally designed to remove such (very) mobile and (very) persistent contaminants. In addition, the recent lowering of drinking-water limits for PFAS to the low ng L^{-1} range changes the risk assessment of MAR for drinking-water production and creates a growing trade-off between water-supply security and the protection of drinking-water quality. In light of this revised risk profile, research is urgently needed to quantify the transport of PFAS in MAR systems, evaluate their attenuation and removal capacity, and develop appropriate monitoring, treatment, and management strategies.

This thesis therefore had the objective of understanding the fate and transport of PFAS under field conditions in surface waters used for drinking water production as well as the subsurface and groundwater systems at MAR sites to eventually assess whether MAR acts as a source or sink for PFAS and advance the optimization of MAR systems for PFAS removal.

1.1.1 Outline of the thesis

The following chapter 2 will provide the background by giving an overview on the definition of PFAS, their classification and regulation, especially in drinking water, as well as an overview on current knowledge of their fate in the environment. Furthermore, a brief outline of MAR as a part of drinking water production will be given. Chapter 3 will then give further insights into the aims and questions addressed in this thesis. The sampling, experimental, and analytical methods are presented in Chapter 4, while Chapter 5 summarizes the five papers included in this thesis and Chapter 6 discusses the results as well as provides an outlook on future studies. The conclusion then answers the research question and summarizes the results.

CHAPTER 2

Background

2.1 Per- and polyfluoroalkyl substances (PFAS)

PFAS are CECs that comprise a group of several thousand to 6.5 million anthropogenic compounds that have been used since the 1950s in a variety of products due to their unique chemical features, consisting of a hydrophobic, fluorinated chain and a hydrophilic functional head group (Evich et al., 2022; Glüge et al., 2020; Padhye et al., 2026; Schymanski et al., 2023). This makes PFAS both oil and water resistant as well as extremely durable and PFAS are therefore greatly used for stain-resistant materials or coatings, such as outdoor gear and clothing, waterproof shoes, or impregnation sprays, as well as for non-stick pans or food-container coatings (Glüge et al., 2020). But PFAS are, among other examples, also used in laboratory and hospital tubing and equipment as Teflon®, in paint, dental floss, ski wax, climbing gear, pesticides, pharmaceuticals and personal care products, or solar cells (Glüge et al., 2020; Nordic Council of Ministers, 2019).

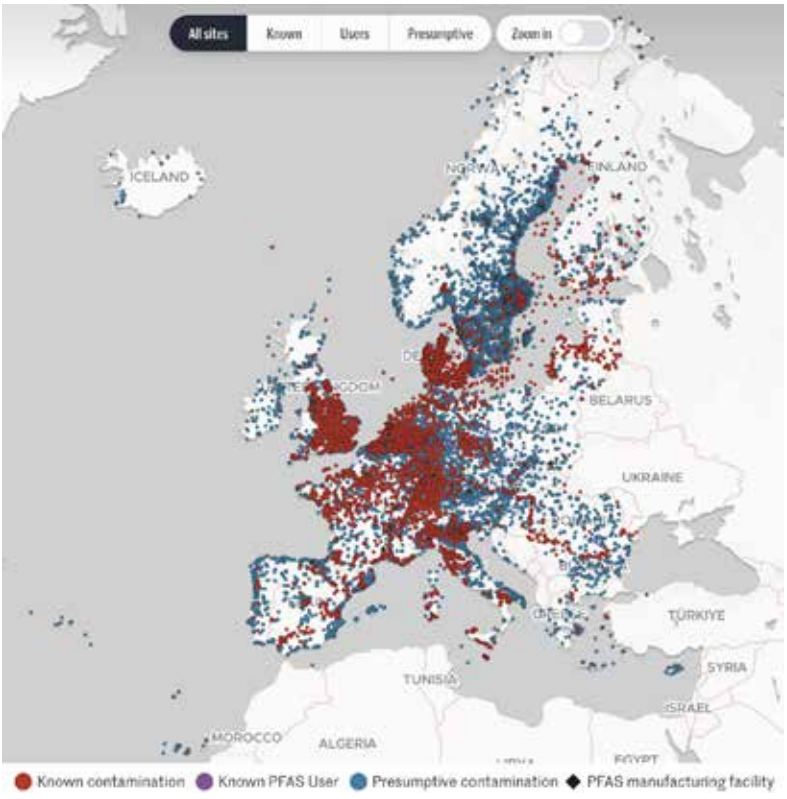

Figure 2.1: Forever pollution map generated by Le Monde and The Forever Pollution Project (Dagorn et al., 2023). Forever pollution map showing the wide-spread known and presumptive PFAS contamination locations detected in Europe in soil and water until 2023. The map is reprinted from Le Monde with the permission of Stéphane Horel.

In total, this spans across 200 applications in 64 categories (Evich et al., 2022; Glüge et al., 2020). However, PFAS are released not only during the use of these products, but predominantly during their production and the incineration of waste after use, leading to a large fraction of the PFAS emissions to the environment (Ahrens and Bundschuh, 2014; Evich et al., 2022; Sörengård

et al., 2022b). One of the most prominent examples of a PFAS containing product that emits PFAS during usage, on the other hand, is aqueous film forming foam (AFFF), which has been used for decades for firefighting as well as firefighting training purposes due to the great ability of AFFF to extinguish heavy flames and electrical fires (Evich et al., 2022; Glüge et al., 2020; Sörengård et al., 2022a). These foams contain extremely high concentrations of PFAS in the μg L^{-1} to mg L^{-1} range and have led to a majority of the today known PFAS hot-spots as shown in Figure 2.1 (Ahrens and Bundschuh, 2014; Dagorn et al., 2023). The environmental concern surrounding PFAS is further compounded by their severe health effects to humans and the environment already at low ng L^{-1} levels, including testicular and kidney cancer as well as immunotoxic effects and hormonal disruptions, increases in cholesterol levels, liver damage or developmental effects during pregnancy (Evich et al., 2022; Fenton et al., 2021; Steenland and Winquist, 2021; Sunderland et al., 2019). In humans, their half-life for example for PFOA is between 2.3 years and 3.8 years and in similar ranges depending on the compound (Seals et al., 2011), however, the PFAS intake is constant and 30% of the general population's PFAS exposure stems from drinking water (Padhye et al., 2026; Tefera et al., 2026). Therefore, PFAS can, with their great persistence and widespread use, be found in various concentrations in every person's blood as well as across all environmental matrices (Bartell Scott et al., 2010; Dagorn et al., 2023; Fenton et al., 2021; Tefera et al., 2026).

2.1.1 Definition and chemical characteristics of PFAS

When defining PFAS as a group of compounds, various definitions exist. The most commonly used definitions are those from Buck et al. (2011) defining PFAS as *"aliphatic substances containing one or more C atoms on which all the H substituents present in the nonfluorinated analogues from which they are notionally derived have been replaced by F atoms, in such a manner that [they] contain the perfluoroalkyl moiety* C_nF_{2n+1}*-"* (Buck et al., 2011) and that of the OECD - Organisation for Economic Co-operation and Development (2021) defining PFAS more inclusively as *"fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it)"* (OECD - Organisation for Economic Co-operation and Development, 2021). Following the most recent standards in

Figure 2.2: Display of the chemical structure of PFAS. Left: Polyfluoroalkyl substance with a partially fluorinated, hydrophobic chain and a hydrophilic functional group. Right: Two legacy perfluoroalkyl substances with each an eight-carbon fully fluorinated chain and a carboxylic acid (PFCA) as functional group of perfluorooctanoic acid (PFOA; top) and a sulfonic acid functional group (PFSA) of perfluorooctanesulfonic acid (PFOS; bottom). PFCAs will be color-coded brown with darker colors for increasing chain length, while PFSAs will be color-coded blue with similarly darker colors for increasing chain length throughout the thesis and papers.

the field, this thesis follows the OECD definition (Evich et al., 2022; OECD - Organisation for Economic Co-operation and Development, 2021).

Classification of PFAS

PFAS are classified by their functional group, with the most well-known groups being the carboxylic acids (PFCAs) and the sulfonic acids (PFSAs) as shown in Figure 2.2. They are then named by the functional group and the chain length of the alkyl chain denoted in Latin numbers as well as the fluorinated degree with fully fluorinated PFAS being denoted as perfluoroalkyl substances and PFAS where one or several fluorine atoms have been replaced by a hydrogen atom being denoted as polyfluoroalkyl substances (Buck et al., 2011). Due to differences in sorption behavior and mobility, PFCAs and PFSAs are often further grouped into short-chain PFAS (C_4-C_7 PFCAs and C_4-C_5 PFSAs) or long-chain PFAS including PFCAs and PFSAs with chain lengths of up to 14 carbon atoms (Interstate Technology Regulatory Council (ITRC), 2020; OECD - Organisation for Economic Co-operation and Development, 2013).

The traditionally used, most commonly known PFAS are often denoted as "legacy PFAS" (Buck et al., 2011; Mahoney et al., 2022), which mostly refer to the longer-chained PFCAs and PFSAs. The most prominent examples are the C8-compounds perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) shown in Figure 2.2, a carboxylic acid and a sulfonic acid with an eight carbon, fully fluorinated chain each. Due to them being banned nowadays, they are in many applications replaced by their shorter-chained C4- or C6-equivalents (Mahoney et al., 2022). Additionally, shorter-chained compounds, which are neither PFCAs nor PFSAs, such as hexafluoropropylene oxide-dimer acid (HPFO-DA or GenX) or 4,8-dioxa-3H-perfluorononanoic acid (NaDONA), are used as replacements and often referred to as next-generation PFAS (Mahoney et al., 2022).

In the environment, precursor compounds can break down into legacy or shorter-chained PFAS, which further complicates the mass balance and can lead to increasing PFAS concentrations detected in target analysis over time (Ersan et al., 2024; Gómez-Ávila et al., 2026). Those precursors include fluorotelomer alcohols, fluorotelomer acrylates, perfluroalkyl sulfonamides such as perfluorooctanesulfonamide (FOSA), perfluoroalkyl sulfonamido ethanols or acetic acids such as N-methylperfluorooctanesulfonamido acetic acid (MeFOSAA) or N-ethylperfluorooctanesulfonamido acetic acid (EtFOSAA), polyfluoro ether carboxylic acids, fluorotelomer phosphate esters and diesters, fluorotelomer carboxylic and sulfonic acids such as 4:2 fluorotelomer sulfonic acid (4:2 FTSA), 6:2 fluorotelomer sulfonic acid (6:2 FTSA), and 8:2 fluorotelomer sulfonic acid (8:2 FTSA), as well as fluorotelomer sulfonamido n-alkyl betaines (Evich et al., 2022).

When conducting target analysis, a fraction of the several thousand PFAS is analysed, which includes the most well-known and studied PFAS. Since this thesis focuses on current and upcoming drinking water guidelines, the PFAS included in this thesis are 29 of the most well-known PFAS included in drinking water guidelines globally as well as a few of their precursors that can break down into the regulated PFAS and some upcoming replacement compounds that are used instead of the traditionally used, regulated PFAS. The full list can be found in the acronym list at the beginning of this thesis.

2.1.2 "Forever chemicals" - PFAS sources, occurrence, and environmental release pathways

PFAS enter the environment originating from point sources such as industrial production sites and their wastewater, firefighting training sites, military airbases or other sites with fire incidents, where AFFF has been applied, landfills or sludge application as well as wastewater treatment plants (Dagorn et al., 2023; Evich et al., 2022). They further stem from diffuse sources such as surface and stormwater runoff or atmospheric deposition, and long-range transport (Banzhaf et al., 2016; Hartz et al., 2024; Ruffle et al., 2024). When looking at the distribution of PFAS, as for example shown on the Forever Pollution Map shown in Figure 2.1 (Dagorn et al., 2023), hot-spots with higher PFAS concentrations in the μg L^{-1} to mg L^{-1} are emphasized. They can be found around the point sources such as PFAS manufacturers, military sites and airports that have used AFFF for fire-extinguishing training or incidents, or close to industrial and waste handling sites (Dagorn et al., 2023; Moghadasi et al., 2023). Depending on the PFAS source, different PFAS are present, such as for example perfluorobutanoic acid (PFBA) and PFOA at landfills or industrially impacted sites and perfluorohexanesulfonic acid (PFHxS) and PFOS at AFFF contaminated sites (Li et al., 2023). Even though legacy PFAS and some next generation PFAS are largely restricted today, many next generation PFAS are still used in a large variety of products (Glüge et al., 2020; Mahoney et al., 2022). Legacy PFAS such as PFOA or PFOS are, due to their persistence, still frequently detected in the environment (Dagorn et al., 2023; Evich et al., 2022; Li et al., 2019; Yang et al., 2026), especially when at concentrations within the ng L^{-1} range (Evich et al., 2022; Gobelius et al., 2018; Kurwadkar, 2017; Ruffle et al., 2024; Sörengård et al., 2022b; Wang et al., 2024; Yang et al., 2026), and therefore reflect the total PFAS release volume since the start of their production.

PFAS transport behavior in the environment

Once in the environment, PFAS can migrate further through the environmental compartments depending on their physio-chemical characteristics as well as complex sorption patterns. Due to the wide diversity of compounds clas-

sified as PFAS, this group shows highly variable water solubility as well as different sorption and transport patterns due to their molecular composition of a hydrophilic functional group coupled to a hydrophobic, fluorinated tail (Ahrens et al., 2011b; Brusseau, 2018; Li et al., 2023; Mishra et al., 2021; Page et al., 2019). Especially highly fluorinated PFAS with a ratio of fluorine to carbon ratio above 1.5 or a perfluorinated carbon chain with four or more carbon atoms are more hydrophobic and therefore show partitioning behavior that is different from other organic contaminants, by strongly sorbing to air-water interfaces (Padhye et al., 2026).

PFAS mobility is therefore largely dependent on the chain length spanning a range of octanol-water partitioning coefficients (log K_{ow}) and soil-water partitioning coefficients (log K_{oc}) (Geosyntec Consultants NC PC, 2019; Gomis et al., 2015; Yang et al., 2026). The higher the hydrophobicity, the lower are the solubility and mobility of the respective compound (Mishra et al., 2021). Consequently, higher hydrophobicity generally leads to better sorption of long-chained PFAS, that then outcompete the shorter-chained PFAS at the sorption sites leading to shorter-chained PFAS often breaking through faster during groundwater transport through an aquifer and being more mobile in the subsurface (Gagliano et al., 2020; Mishra et al., 2021). Similarly, PFSAs tend to be less mobile than the corresponding chain length PFCAs, since PFSAs contain one more fluorinated carbon compared to the carboxylic acid group carbon in the PFCAs, which leads to higher hydrophobicity of PFSAs. Therefore, PFSAs tend to sorb stronger than a PFCA of the same chain length (Gagliano et al., 2020; Maimaiti et al., 2018).

Since the strength of the hydrophobic interactions is correlated to the PFAS chain length, electrostatic interactions are the major sorption process for shorter-chained PFAS. Electrostatic interactions of the anionic head-group with positively charged sorbents can then be further enhanced by increased concentrations of major cations in the water, while major water anions in the solution possibly compete for the same sorption sites (Maimaiti et al., 2018). Long-chained PFAS sorption, however, is mainly driven by hydrophobic interactions (Patterson et al., 2010). PFAS can also sorb via hydrogen bonding or covalent bonding (Gagliano et al., 2020).

Soil adsorption distribution coefficients (K_d values) describe the ratio of the contaminant concentration on the solid phase to its concentration in the aque-

ous phase at equilibrium and similarly increase with PFAS chain length (Higgins and Luthy, 2006; Sima and Jaffé, 2021), with 0.06 ± 0.02 L kg^{-1} for shorter-chained PFAS like perfluoropentanoic acid (PFPeA) and ranging up to 1.2 – 3.2 L kg^{-1} for the longer-chained PFOS (Ferrey et al., 2012; Kolade et al., 2025; Nguyen et al., 2020; Silva et al., 2021). These distribution coefficients are commonly based on solid-phase adsorption, where sorption of these organic compounds is strongly influenced by organic matter. However, due to the surfactant nature of PFAS, sorption at the air-water interface in the unsaturated zone can also be a major attenuation pathway (Brusseau, 2025; Kolade et al., 2025; Lyu et al., 2018) and their sorption behavior is complex. Long-chained PFAS generally sorb primarily to the humin fraction of the soil organic matter, while shorter-chained PFAS preferentially sorb to the more hydrophilic humic or fulvic acid fractions (Campos Pereira et al., 2018; Higgins and Luthy, 2006; Sima and Jaffé, 2021). Moreover, variations in the water content of the unsaturated zone can lead to two to four times higher PFAS concentrations entering groundwater systems during wet seasons or high precipitation events (Liu et al., 2018) due to changes in the air-water interfaces and subsequent remobilization of PFAS (Guo et al., 2020; Liu et al., 2018; Newell et al., 2023).

This is also reflected by the substantial variability of reported retardation factors (R), which describe the ratio between the water flow velocity and the contaminant flow velocity. R values for PFAS vary between 1.2 for PFOA and 1.7 for PFOS in saturated sand with low f_{oc} values around 0.02% to 0.04% (Ferrey et al., 2012; Xiao et al., 2015) and up to over 1000 in the unsaturated zone (Guo et al., 2020; Kolade et al., 2025; Xiao et al., 2015). Further, PFAS desorption hysteresis means that PFAS desorption is slower than adsorption, leading to sorbed PFAS in the unsaturated zone posing a risk to underlying groundwater systems and drinking water supplies for up to decades (Sima and Jaffé, 2021; Sörengård et al., 2022a; Xiao et al., 2015).

These variations and effects are often underestimated in laboratory experiments (Sima and Jaffé, 2021) and are highly complex due to various influencing factors e.g., the specific compound's chain length, the environmental medium, groundwater levels (saturated vs. unsaturated zones), hydrological conditions like flow velocity and recharge rate, as well as geochemical conditions like pH and ionic strength (Das et al., 2024).

2.1.3 Analytical challenges

Throughout time, analytical capabilities and the possibility to detect very low concentrations of contaminants have improved considerably now reaching down to ng L^{-1} levels or even further. To illustrate this, imagine dissolving one grain of salt in an Olympic-sized swimming pool and then finding it again: assuming one grain of salt weighing 0.02 g, this corresponds to a concentration of 8 ng salt per liter in the swimming pool. But despite these analytic possibilities, detecting and quantifying PFAS within the low ng L^{-1} concentration range remains challenging. Stronger sorption effects to analysis equipment or sampling containers, contamination of blank samples from the surroundings during analysis, or challenges associated with using PFAS-free equipment (Ahrens et al., 2010) as well as previously higher drinking water guideline thresholds and "famous" cases of highly polluted areas have led to a large majority of studies focusing on highly contaminated sites or experiments with high spiking concentrations of PFAS (Høisæter et al., 2019). Therefore, PFAS at low ambient concentrations at sites that do not have any known "contamination case" are rarely part of monitoring programs and little is known about the temporal and seasonal variations of PFAS in surface waters. Further, ambient or background concentrations of contaminants in general are often considered as being "non-problematic" concentrations, while for PFAS already those low ng L^{-1} concentrations can cause serious health effects. This is increasingly reflected in sharpened drinking water guidelines in the low ng L^{-1} range (Livsmedelverket, 2022; Miljøministeriet, 2024; United States Environmental Protection Agency, 2024), which makes PFAS quite prominent compared to other persistent organic contaminants as well as leads to more widespread areas being classified as "contaminated" (Padhye et al., 2026).

2.1.4 Regulation of PFAS

The exponential increase in newly produced chemicals (Escher et al., 2020; Wang et al., 2024) means that regulations are not able to keep up with the increasing number of potential contaminants released to the environment (Escher et al., 2020). This leads to many local point and diffuse sources releasing contaminants to the environment and eventually reaching groundwater systems and drinking water supplies. PFAS have been increasingly produced

since the 1940s and today, global fluoropolymer production volumes are above 230 000 tonnes year^{-1} (Evich et al., 2022; Glüge et al., 2020). However, a few legacy PFAS are banned since 2009, 2019, and 2022 by the Stockholm Convention, a regulatory framework addressing a variety of persistent organic pollutants. Those are PFOA (2019), PFHxS (2022), and PFOS (2009) and their respective precursors (Padhye et al., 2026; Rudin et al., 2023). Even though not longer being produced, these compounds have been emitted to the environment for decades and are still detected at high frequencies and concentrations due to their persistence (Cousins et al., 2022; Dagorn et al., 2023; Evich et al., 2022). This is why these compounds are still included in different countries' drinking water guidelines.

Since 2007, the EU chemical regulation REACH (registration, evaluation, authorisation, and restriction of chemicals) is based on six criteria to classify and regulate a compound as a "substance of very high concern": (1) carcinogenicity, (2) mutagenicity, (3) toxicity to reproduction, (4) persistence, bioaccumulation, and toxicity, (5) very high persistence and very high bioaccumulation, and (6) substances of equivalent concern (Rudin et al., 2023; The European Parliament and The Council of the European Union, 2020). If one of the criteria is met, the compound can qualify for the REACH candidate list, but the endpoints for these criteria are based on acute evidence (The European Parliament and The Council of the European Union, 2020). Since the health effects of PFAS are chronic, long-term, and similar to effects of other compounds or diseases, this assessment is very challenging for PFAS (Andrews and Walker, 2015; Boden, 2020; Frisbee et al., 2010; Rudin et al., 2023). PFAS therefore pose a threat to humans and the environment and can be linked to certain diseases (Evich et al., 2022; Fenton et al., 2021; Steenland and Winquist, 2021), but often do not meet the regulatory toxicity and carcinogenicity criteria. While it is debated, whether contaminants which classify as (very) mobile and (very) persistent, should be regulated (Cousins et al., 2020; Rudin et al., 2023), these criteria are not yet included in regulations such as the EU REACH regulation.

Additionally, these assessments are very time-consuming and require a large quantity of evidence for each compound, which is why it would take decades if an assessment was to be made for every of the several thousand to several million PFAS (Cousins et al., 2020; Rudin et al., 2023). Therefore, five coun-

tries within the EU (Denmark, Germany, Norway, Sweden, and the Netherlands) have put forward an EU restriction proposal to the European Chemicals Agency (ECHA) at the end of 2023 to propose a ban of all PFAS as a single group (European Chemicals Agency (ECHA), 2024). This proposal is one of the first of its kind trying to regulate a contaminant class as one group and is currently assessed by the ECHA (Cousins et al., 2020; European Chemicals Agency (ECHA), 2024). However, due to over 5 000 comments from stakeholders and researchers, this assessment is expected to require an elongated time frame and derogations will be proposed based on the public consultation (European Chemicals Agency (ECHA), 2024).

PFAS drinking water regulations

Globally, many countries adjust PFAS drinking water guidelines to low ng L^{-1} concentrations as shown in Figure 2.3, and often regulate a sum of several well-known PFAS, such as the sum of four PFAS, namely PFOA, perfluorononanoic acid (PFNA), PFHxS, and PFOS. These regulations are often a result of sharpening the drinking water guidelines by several orders of magnitude in recent or upcoming years in compliance with the analytical limitations of commercial laboratories as well as health concerns (EFSA Panel on Contaminants in the Food Chain (EFSA CONTAM Panel) et al., 2020; Ruffle et al., 2024). A recent example are the Canadian drinking water guidelines that in 2024 proposed a change from 600 ng L^{-1} for the sum of PFAS to a limit of 30 ng L^{-1} for the sum of 25 PFAS (Health Canada, 2024). These drastic changes (Figure 2.3) show the urgency for addressing and understanding the fate and effects of the low-contaminated PFAS sites. Similarly, the EU has an overall drinking water limit of 500 ng L^{-1} for the sum of all PFAS and a lower limit of 100 ng L^{-1} for the sum of 20 specific PFAS (The European Parliament and The Council of the European Union, 2020). Individual countries within the EU can then have lower drinking water guidelines, with Denmark having the lowest limit of 2 ng L^{-1} for the sum of 4 PFAS (PFOA, PFNA, PFHxS, PFOS) (Miljøministeriet, 2024) and Sweden has implemented a similar limit of 4 ng L^{-1} for the same 4 PFAS in January 2026 (Livsmedelverket, 2022). The US EPA also introduced a similar limit in 2023 with 4 ng L^{-1} for PFOA and PFOS, respectively, as well as 10 ng L^{-1} for each of PFNA, PFHxS, and GenX (United States Environmental Protection Agency, 2024).

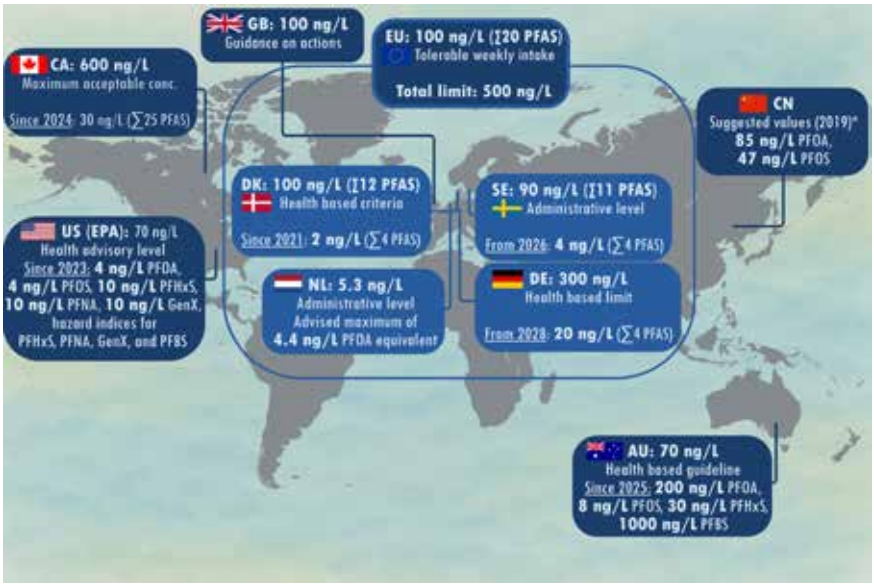

Figure 2.3: Current and upcoming drinking water guidelines for PFAS. Values were obtained from literature: Australian Government - National Health and Medical Research Council (NHMRC) (2025); Council of the European Union (2025); Drinking Water Inspectorate Guidance to water companies (DWI)) (2025); European Comission (2020); Gobelius et al. (2018); Health Canada (2024); Lange and Sacher (2025); Livsmedelverket (2022); Miljøministeriet (2024); Smith et al. (2025); United States Environmental Protection Agency (2024); van der Aa et al. (2021). Figure modified and reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

With drinking water guidelines currently approaching these low levels for PFAS, not only the "hot-spots", but also the sites showing low ambient PFAS concentrations become relevant. According to Evich et al. (2022); Gobelius et al. (2018); Jarvis et al. (2021); Ruffle et al. (2024), ubiquitously detected PFAS concentrations are between 1 and 10 ng L^{-1} and Jarvis et al. (2021) even classifies any PFOS concentration in surface waters below 30 ng L^{-1} as "very low" (Jarvis et al., 2021), which, depending on the local regulations, pose a challenge when used for drinking water production. Therefore, detailed moni-

toring and an in-depth understanding of PFAS fate as well as their impacts on human and environmental health are urgently needed (Padhye et al., 2026), especially for sites with low ambient PFAS concentrations.

2.1.5 PFAS treatment technologies

While PFAS sorption patterns are complex, those characteristics can be utilized in engineered PFAS removal or treatment strategies (Padhye et al., 2026). When producing drinking water, waterworks that handle highly PFAS contaminated waters often have a combination of respective adsorption, enrichment, and treatment systems in place, such as reverse osmosis, nanofiltration, anion exchange resins, granular activated carbon (GAC) filters, foam fractionation (FF), or electrochemical oxidation (Baresel et al., 2022; Chen et al., 2022; Padhye et al., 2026; Stackelberg et al., 2007).

Membrane filtration, such as reverse osmosis or nanofiltration, can reach PFAS removal rates of up to 99% (Baresel et al., 2022), but at the same time results in a retentate waste stream that contains high PFAS concentrations and needs further destructive treatment such as electrochemical oxidation (Baresel et al., 2022; Xu et al., 2021). FF is considered a relatively inexpensive and environmentally friendly PFAS concentration technique, but similarly generates a retentate waste stream with highly contaminated foam (Buckley et al., 2023; Smith et al., 2022, 2023c). Moreover, shorter-chained PFAS are less efficiently removed and require the addition of a co-surfactant (McCleaf et al., 2021, 2023; Smith et al., 2022). Anion exchange resins and GAC filters also show high removal rates of especially long-chained PFAS and GAC filters have been used widely. However, shorter-chained PFAS can break through early and the regeneration of the material via incineration at extremely high temperatures is costly (Belkouteb et al., 2020). Anion exchange resins show better PFAS removal rates than GAC, especially for shorter-chained PFAS, but the media costs are higher (McCleaf et al., 2017; Murray et al., 2021).

Since many of these treatment methods are cost-intensive and require regular change and maintenance, especially when treating for PFAS, targeting PFAS concentrations in the low ng L^{-1} range substantially increases the drinking water treatment costs (Belkouteb et al., 2020; Franke et al., 2021). Waterworks

that have so far not had to treat for PFAS, possibly need to rethink their treatment systems with lower drinking water thresholds approaching (European Comission, 2020; Livsmedelverket, 2022; Miljøministeriet, 2024) and an optimized MAR system acting as natural pre-treatment barrier could be one approach to complement these technologies.

2.2 Managed Aquifer Recharge (MAR)

MAR is the process of artificially infiltrating surface water into groundwater systems. Globally, the highest proportion of water is infiltrated in India (31%), the USA (26%), and Germany (9%) (Sinshaw et al., 2025). The earliest prototype of a MAR system was built in 221 B.C. in China (Sinshaw et al., 2025; Wang et al., 2014). In Sweden, MAR has been widely applied since the end of the 19th century (Svenskt Vatten, 2019). Today, Europe has 224 active MAR sites across 23 countries. After MAR passage, the surface water or artificially recharged groundwater is, together with the natural groundwater, retrieved at the drinking water treatment plant to undergo aeration, hardness removal, pH adjustment, rapid sand filtration, sometimes further treatment via activated carbon filters or similar, and disinfection before distribution of the final drinking water (Yu et al., 2022). The MAR process is schematically shown in Figure 2.4.

MAR is a very effective technique to ensure a stable water supply in groundwater systems used for drinking water production as well as to reduce turbidity and dissolved organic matter (DOM) from surface water (Ahmed and Marhaba, 2017; Balke and Zhu, 2008; Fakhreddine et al., 2021; Henzler et al., 2014). It can aid to increase aquifer storage and thereby reduce water scarcity, act as prevention measure for saltwater intrusion, reduce environmental risks such as land subsidence or flooding, enhance agricultural efficiency, and improve water quality (Sinshaw et al., 2025). MAR further ensures a lower and more constant water temperature throughout the year, which is associated with more palatable drinking water (Ahmed and Marhaba, 2017; Lee et al., 2009). During infiltration, natural purification processes further result in complete or partial retention of nutrients, metallic or organic pollutants in the subsurface as well as the significant reduction of DOM. Thereby, MAR can improve color, taste, and odor, and thus is an alternative for chemically treat-

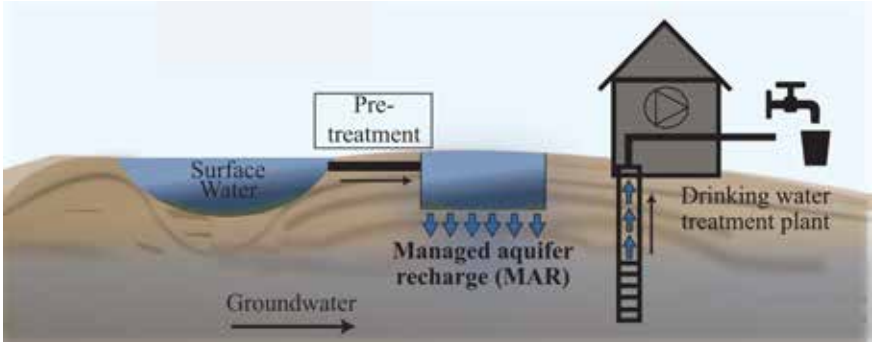

Figure 2.4: Schematic overview of the artificial infiltration in a MAR scheme using basin filtration.

ing surface water during drinking water production (Balke and Zhu, 2008; Fakhreddine et al., 2021; Jokela et al., 2017). MAR can also be combined with other pre-treatment technologies to remove suspended solids, salinity, or microbial risks and to increase the recharge efficiency. However, MAR can become expensive at contaminated sites if the water has to be pre-treated with expensive techniques (Sinshaw et al., 2025). But with decreasing groundwater levels due to climate change and greater needs for sufficient drinking water quantity and quality for growing populations, MAR will likely become even more relevant in the future and especially in countries with few or small aquifers, MAR can account for a large fraction of the drinking water production (Ahmed and Marhaba, 2017; Hägg, 2020; Sweden Water Research, 2024).

2.2.1 PFAS in MAR systems

While MAR has many benefits, these systems are not designed to treat organic CECs and unintentional desorption or dissolution might cause the opposite effect introducing toxic metals, pesticides, industrially produced compounds, microorganisms, natural toxins or other micropollutants into the subsurface (Fakhreddine et al., 2021; Yu et al., 2022). The infiltration during MAR

could thus act as a shortcut for mobile and persistent CECs such as PFAS into groundwater systems and eventually drinking water supplies, but research regarding this process is scarce, especially for PFAS (Díaz-Cruz and Barceló, 2008; Postigo and Barceló, 2015; Sörengård et al., 2022b). Once infiltrated, PFAS can be accumulated and eventually be remobilized from the subsurface (Gómez-Ávila et al., 2026), but the processes are not well understood. Furthermore, due to the chronic toxicity of some PFAS at low concentrations and their complex transport behavior as well as the limited understanding of PFAS fate in MAR systems, together with increasingly stringent drinking water regulations as well as the expense of available treatment technologies (Franke et al., 2021; Sinshaw et al., 2025), monitoring efforts to track changing PFAS concentrations are highly valuable.

To address this research gap, this thesis investigates the transport and fate of PFAS in MAR systems at sites with low PFAS concentrations. The aim is to gain a detailed process understanding as well as mechanistic and operational insights to eventually be able to provide the basis for the optimization of MAR systems for PFAS removal to meet upcoming drinking water guidelines with cost-efficient drinking water treatment systems.

CHAPTER 3

Specific aims

To answer the research question of this thesis, *"Does MAR act as a source or sink for ambient PFAS concentrations into groundwater systems and drinking water supplies?"*, the following aims were addressed:

1. Understanding drivers of seasonal variations and sources of PFAS concentrations in surface waters (Papers **I** & **II**) and groundwater systems (Papers **I** & **III**) at ambient background level concentrations.

2. Assessing the fate and transport of PFAS in the subsurface including detailed process understanding of the factors affecting the mobility, retention, accumulation, as well as sorption and release processes of ambient PFAS concentrations in MAR systems (Papers **II** & **III** & **IV**).

3. Exploring the possibilities to optimize MAR systems for effective PFAS removal by looking at the buffering capacity for seasonal peaks and strategic operation plans (Papers **II** and **III**) and pre-treatment steps prior to MAR (Papers **IV** and **V**).

Included are five studies investigating what drives the fate of ambient PFAS concentrations in MAR systems. This includes a review paper (Paper **I**) giving an overview of the relevance of addressing CECs in MAR systems, a two-year monitoring program at two full-scale MAR sites (Papers **II** and **III**), saturated column experiments (Paper **IV**), and a pilot study on PFAS treatment via FF (Paper **V**) prior to infiltration via MAR (Figure 3.1). The papers thus give insights into PFAS concentrations prior, during, and after MAR as well as possible modifications to reduce the input of PFAS into groundwater systems and eventually drinking water supplies and to meet upcoming drinking water guidelines (Figure 3.1). Especially data on sites with only ambient background levels of PFAS outside of PFAS hot-spots (Dagorn et al., 2023) are scarce, as shown in chapter 2, which is why this thesis especially focuses on low ambient PFAS concentrations to understand possible differences compared to highly contaminated sites.

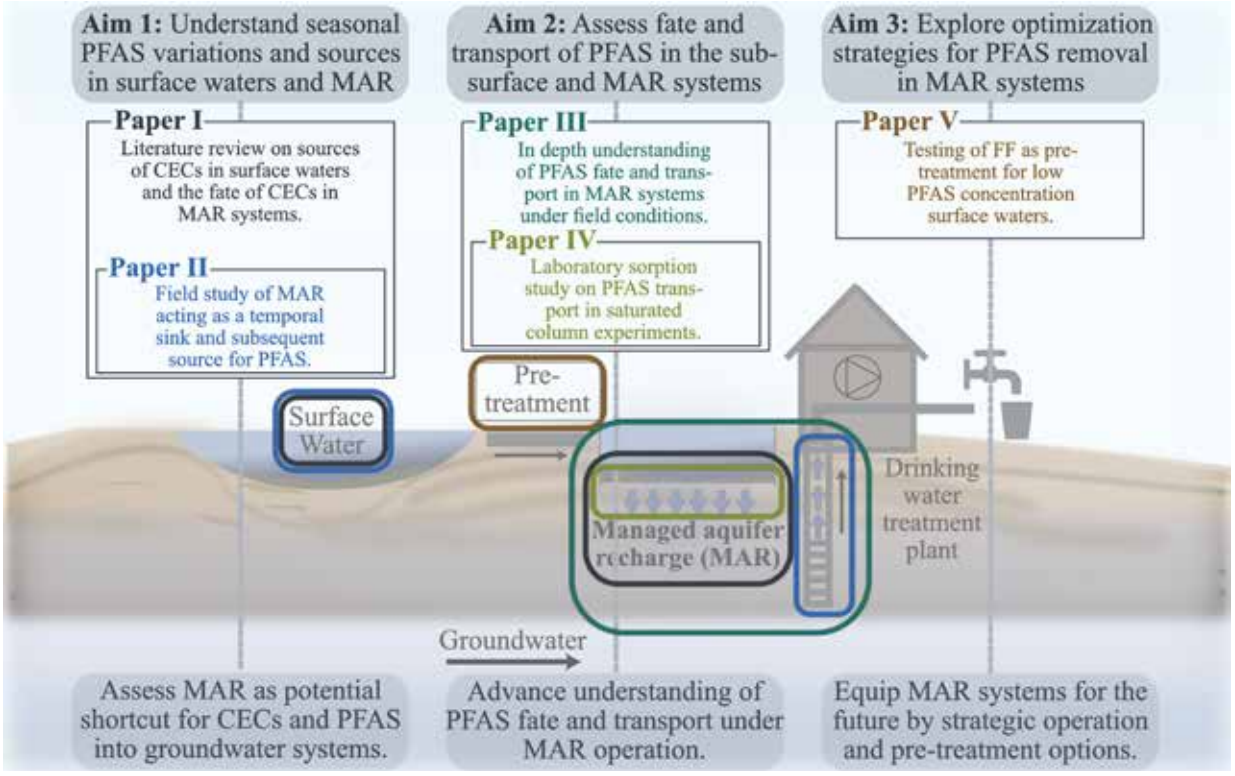

Figure 3.1: Overview of the aims of this thesis and how the five papers address these. The colored boxes indicate which paper (color-coded text) focuses on the various parts of the MAR system show in Figure 2.4.

CHAPTER 4

Methodology

The methodology section of this thesis is structured into four major parts describing (1) the field sampling conducted at two hydrogeologically contrasting Swedish MAR sites (A and B) between September 2022 and June 2024 to provide the data shown in papers **II** and **III**, (2) a laboratory column experiment conducted for paper **IV**, (3) a foam fractionation pilot at site B for paper **V** and (4) the chemical analysis of the samples collected for (1), (2), and (3).

4.1 Field site description

For the seasonal monitoring data, monthly samples were taken at two Swedish waterworks in Southern Sweden at the respective MAR sites in addition to their own standard water quality sampling. Due to confidentiality reasons, the sites will be denoted as site A and site B throughout this thesis and the respective papers. For an understanding of the systems and their characteristics in this thesis, the exact location is not relevant and schematics are provided to illustrate groundwater flow directions and sampling points in the respective papers. Site A is a large MAR site in a shallow glaciofluvial aquifer

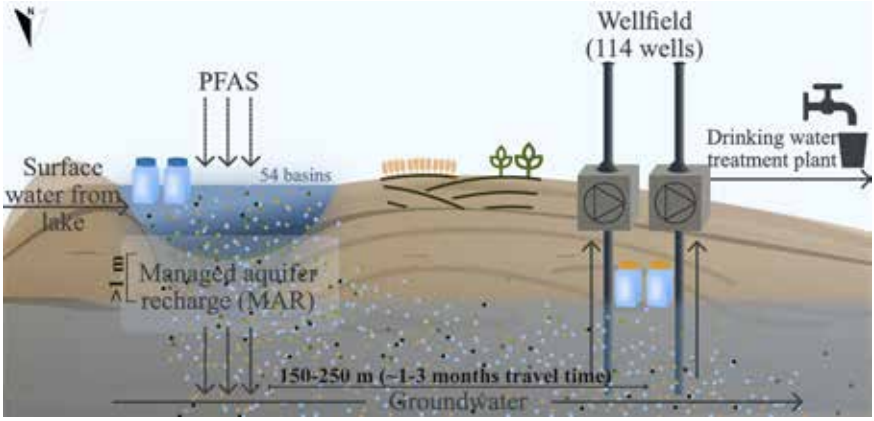

Figure 4.1: Schematic drawing of sampling site A located in a remote, agricultural area. Samples were taken monthly over two years at the inlet to the infiltration basins and at a well that retrieves the artificially recharged groundwater. The figure was modified and reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

supplied by lake water and operated continuously, whereas Site B is a smaller system that utilizes a deep esker system supplied by river water and operated intermittently. These differences provide the basis for investigating PFAS transport under contrasting hydrogeological and operational conditions.

Some of the papers show an overlap or comparison of the data sampled and analysed for this thesis and the waterworks standard water quality samples acquired prior to fall 2022 or during the sampling period of this thesis. Additionally, the waterworks provided surface water from their MAR sites as inlet water for the column experiments and pilot study. The following information and details of the systems at the two sites were kindly provided through personal communication with the waterworks.

4.1.1 MAR site A

Site A is a large MAR system using a lake with a surface area of 12 km^2 and a catchment area of 447 km^2 as the source water for the MAR system. The lake

is located in a remote, agricultural area without any known PFAS "hot-spots" in the vicinity.

MAR system and operation

The MAR system is located in a shallow glaciofluvial aquifer and uses 54 artificial infiltration basins with a surface area of about 9000 m^2 each and a sandy bottom, utilizing the natural deposit of sand from the area (Figure 4.1). Of the 54 basins, about half of the basins are simultaneously in operation and the cumulative infiltration rate is about 1000 L s^{-1}. The lake water is passed through a 40 μm sieve prior to distribution to the infiltration basins and recovered after one to three months of travel time at one of the 114 wells. The unsaturated zone below the basins comprises about four to six meters. Sampling of the surface water took place at the inlet to the infiltration basins before the water is distributed to the different basins for papers **II** and **IV** and of the artificially recharged groundwater at a recovery well that has the lowest impact of natural groundwater and therefore primarily recovers the artificially infiltrated groundwater for paper **II**.

Further drinking water treatment

After recovery from the MAR system, the water is aerated and passed through a softening reactor. Afterwards, particles are removed by coagulation with iron chloride followed by a rapid sand filter. Finally, the water is UV treated and, if necessary, chlorinated, before distribution to the drinking water network or a temporary water reservoir. The MAR system, as the first step in this treatment chain, acts as a natural and cost-effective filter for organic matter, as described in Chapter 2.

4.1.2 MAR site B

The MAR system at site B utilizes surface water from a river with an average flowrate of 500 L s^{-1}. The river passes through a mid-sized Swedish city downstream of the infiltration site and is therefore more anthropogenically impacted. River water is passed through a sand filter before entering the MAR system.

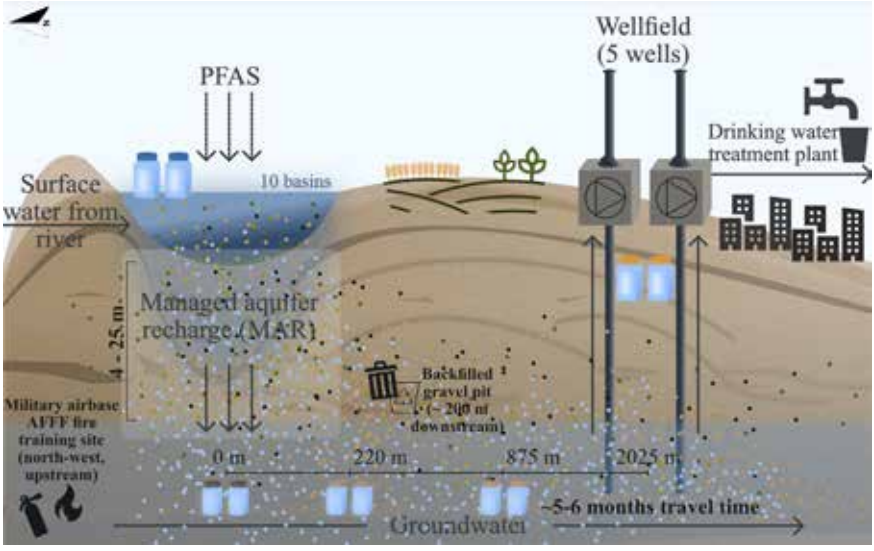

Figure 4.2: Schematic drawing of sampling site B located in vicinity to a mid-sized Swedish city. Samples were taken bi-weekly to monthly over two years at the inlet to the infiltration basins and at the wellfield that retrieves the artificially recharged groundwater. Additional samples between these two sampling points were taken bi-weekly to monthly in fall 2023 and spring 2024. The figure was modified and reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

MAR system and operation

The MAR system at site B consists of ten infiltration basins with a surface area of about 950 m^2 each and a sandy bottom (Figure 4.2). However, due to a lack of natural sand deposits at this site, the sand is obtained from other sites and added artificially to the infiltration basins. About half of the infiltration basins are simultaneously in operation resulting in an average infiltration rate of about 135 L s^{-1}. Since the MAR system is located on top of a deep esker, the unsaturated zone comprises up to 25 m and the travel time for the distance of about 2 km until recovery of the artificially recharged water is five to six months. The composition of the recovered groundwater is estimated by the waterworks to be about 90%-95% artificially infiltrated groundwater and 5%-

10% natural groundwater. In contrast to site A, the MAR system at site B is not always in operation, since the infiltration is turned off when the natural groundwater is sufficient during periods of high precipitation or when the river water quality criteria are not met. Samples were taken at the sand filters prior to the MAR system for paper **V**, before distribution to the MAR system for papers **II**, **III**, and **IV** and at the recovery wells for papers **II**, and **III**. Additional samples for paper **III** were taken in fall 2023 and spring 2024 at a reference sample north of the infiltration site, below one of the infiltration basins, 220 m downstream from the MAR system, and 875 m downstream from the MAR system, respectively.

Further drinking water treatment

After recovery, the water is passed to the drinking water treatment plant, where it is aerated, softened, pH adjusted and passed through two-stage rapid sand filters. Finally, the water enters the drinking water distribution network after disinfection.

4.1.3 Sampling

Overall, the monitoring campaign comprised two years of monthly sampling at the two contrasting MAR sites, supplemented by a high-resolution three-month campaign at site B during fall 2023. For the monitoring in papers **II** and **III**, surface- and groundwater samples were taken monthly by the waterworks as grab samples from September 2022 to March 2024 at site A and until June 2024 at site B. In fall 2023, an additional three months of bi-weekly sampling, including additional sampling points between the infiltration site and the wellfield with detailed analysis of PFAS and stable oxygen water isotope ratios, was conducted at site B for paper **III**. The column experiments for paper **IV** were conducted in September and October 2024, and the inlet water for the experiments was collected weekly at the two sites. Pilot tests of a foam fractionation for PFAS treatment in paper **V** at the sand filters prior to the MAR system at site B were conducted in summer 2025. The samples were collected in 1L high-density polyethylene (HDPE) bottles for PFAS, in 250 mL HDPE bottles for cation and anion analysis, and in 60 mL HDPE bottles for stable water isotope analysis. Dissolved organic carbon (DOC) and DOM samples were collected in 60 mL HDPE bottles until August 2023

and afterwards as triplicates in 17 mL muffled glass vials. During sampling, nitrile gloves were worn, the bottles were rinsed three times with the sampling water, and an air blank was taken for PFAS every other month. Surface water samples from the lake and the river, respectively, were taken 10 cm below the surface, while the standing water was pumped out and discarded prior to taking groundwater samples from a well. PFAS samples were stored frozen and all other water samples were stored refrigerated. Soil samples for PFAS analysis were collected in plastic bags and stored frozen, while foam samples for PFAS analysis were collected with a squeegee or a vacuum pump and collapsed into a 1L HDPE bottle using a hot air gun before being frozen.

Further details about the sampling can be found in the respective papers (**II** - **V**).

4.2 Column experiment design

The experimental design was intended to evaluate the influence of infiltration material, prior MAR operation, sorbent amendments, and inlet PFAS concentrations on PFAS transport. To study the effects in a controlled environment, laboratory flow-through column experiments filled with infiltration sand or crushed rock were set up under saturated conditions using new sand and sand that had been used previously in the operating infiltration basins from both MAR sites as well as testing different levels of PFAS inlet concentrations.

The column experiments were conducted using 16 polyvinyl chloride (PVC) columns with a length of 30 cm and a diameter of 5.3 cm (see Figure 4.3) to ensure a length to diameter ratio above four as recommended in Lewis and Sjöstrom (2010) and were run in duplicates. The columns were operated at room temperature with top-down flow in a closed, fully saturated system driven by a hydraulic gradient. The inlet water originated from the two MAR sites and was replaced daily. The sand used in the columns originated from the two MAR sites, and a grain size analysis was conducted showing well-sorted, medium sand. The columns contained fresh, clean sand (columns 1, 2, 6, 7, and 11-16 in Figure 4.3) and sand that had been used in the operating MAR infiltration basins and therefore contained a biofilm (columns 3, 4, 8, and 9 in Figure 4.3). Columns 11-16 additionally contained PFAS sorbent materials (GAC, biochar or wood bark; Figure 4.3) on top of the sand using

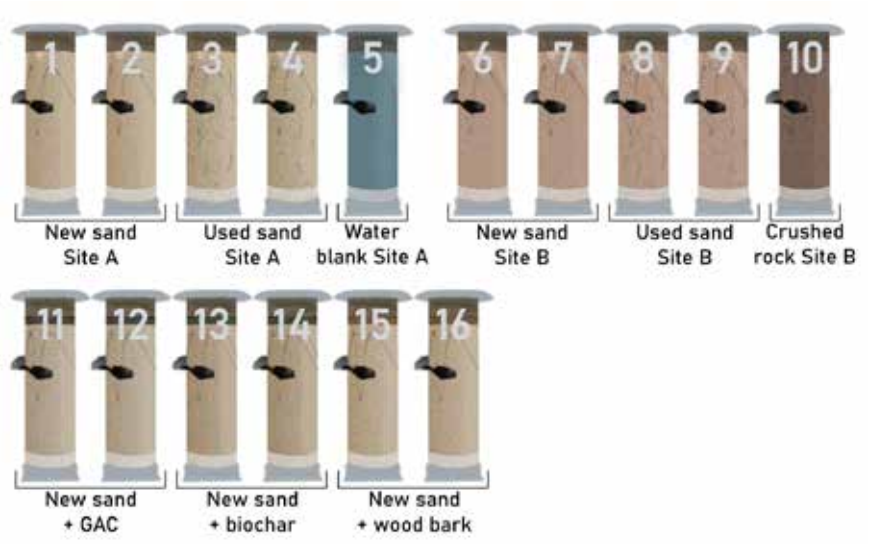

Figure 4.3: Schematic drawing of the set-up of the 16 columns and their numbers. The columns were filled with sand or crushed rock from the respective sites (A and B) of different ages testing new, pristine sand against sand that had been operated in the infiltration basins before. Column 5 was a water blank. Columns 11-16 additionally contained a PFAS sorbent material on top of the sand column. The figure was reprinted from paper **IV** (manuscript in preparation).

a dry weight ratio of sorbent:sand of 1:100. A blank column without sand containing water from site A (column 5 in Figure 4.3) as well as a column containing crushed rock from site B (column 10 in Figure 4.3) with a similar grain size distribution as the sand were included as well. Details regarding the setup and packing as well as the individual conditions of the columns and flowrates can be found in paper **IV**.

To compare differences in sorption behavior, three PFAS spiking concentration levels were tested over two periods, with (1) testing unspiked inlet water from the two respective sites over 8 days, and (2) testing two different spiking solutions: a realistic environmental peak PFAS concentration of approximately 40 ng L^{-1} using a PFAS spiking mixture (PFBA, PFPeA, PFHxA, PFHpA,

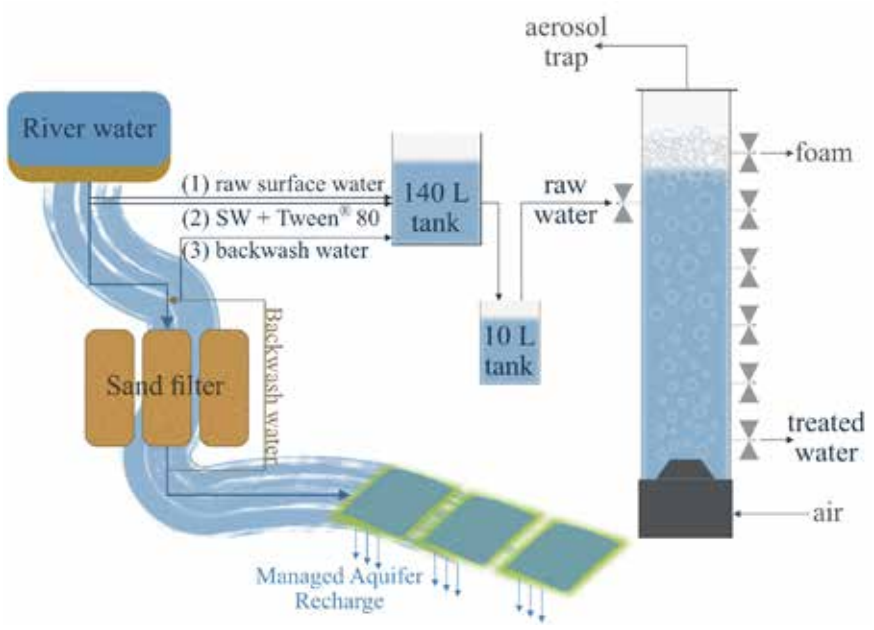

Figure 4.4: Schematic drawing of the set-up of the foam fractionation pilot and its placement in the water treatment train. Three different water types as well as contact times of 10, 20, and 82 minutes were each tested in triplicates. The figure was modified and reprinted from paper **V** (manuscript in preparation).

PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA, PFBS, PFHxS, PFOS, FOSA, 6:2 FTSA) for one hour before returning to unspiked inlet water, and a very high spiking concentration of of approximately 65 000 ng L^{-1} of the same PFAS spiking mixture for one hour before returning to unspiked inlet water. The sampling frequency was highest during the initial breakthrough period and gradually reduced over time.

4.3 Pilot test of foam fractionation at site B

As a possible cost-efficient and environmentally friendly PFAS pre-treatment step prior to MAR, a continuous FF pilot column was tested at the site with

the higher surface water PFAS concentrations (site B). Three water types were selected to investigate treatment performance under different particle loads and surfactant conditions. The FF pilot was therefore operated in triplicate runs using (1) inlet water (from a river) for the rapid sand filters prior to the MAR system, (2) the same water as in (1), but spiked with 10 mg L^{-1} of a biodegradable surfactant (Tween® 80), and (3) backwash water from the rapid sand filters (Figure 4.4). The column was filled to a level of 1.5 m and air was continously introduced through a nozzle at the bottom of the column. Three different contact times were tested to determine stabilization, removal efficiencies, and enrichment factors for each water type: 10 minutes, 20 minutes, and 82 minutes. Samples of the raw water, the inlet water, and the treated water, as well as of the acquired foam and of an aerosol trap were then taken at the beginning and the end of each run. Details regarding the experimental setup and sampling can be found in paper **V**.

4.4 Analytical methods and data evaluation

The analysis of the samples was conducted at the department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (SLU) in Uppsala for PFAS samples, while DOC and DOM samples were analysed by Dolly Kothawala at the department of Ecology and Genetics - Limnology at the Evolutionary Biology Centrum at Uppsala University. All other water quality parameters were analysed by external commercial laboratories (see Section 4.4.3).

4.4.1 PFAS target analysis

Target analysis of 29 PFAS (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, PFDS, 4-2 FTSA, 6-2 FTSA, 8-2 FTSA, FOSA, Me-FOSAA, Et-FOSAA, HFPO-DA, NaDONA, 9Cl-PF3ONS, 11Cl-PF3OUdS, PFECHS; full names can be found in the acronym list in the beginning of the thesis) was conducted using liquid chromatography coupled to tandem mass spectrometry (LC-MS/MS; SCIEX Triple Quad 3500 UPLC-MS/MS) as described in detail in papers **II** and **III**.

Sample pre-concentration

Due to sufficiently high PFAS concentrations in the highly spiked column experiment, the samples could be directly analysed on the LC-MS/MS (without pre-concentration) as described in paper **IV**. PFAS samples were pre-concentrated for water and foam matrices for all other samples to reach sufficiently high concentrations for detection. Solid-phase extraction was conducted following the protocol of Ahrens et al. (2010) as described in Belkouteb et al. (2020) and Smith et al. (2023c) using Oasis WAX cartridges (6 cc, 500 mg, 60μm, Waters Corporation, USA). Each solid-phase extraction included two solvent blanks.

Water samples for papers **II**-**V** were sonicated and filtered using 1.2 μm Whatman® glass microfiber filters (binder free, grade GF/C) to remove particles that could possibly clog the solid phase extraction cartridges. The samples were then split into duplicates or triplicates and each spiked with internal standard (IS; 0.05 μg mL^{-1}; Wellington Laboratories, MPFAC-24ES mixture with $^{13}C_3$-HFPO-DA added individually) prior to solid-phase extraction and evaporation under constant nitrogen flow followed by LC-MS/MS analysis.

For the foam samples, 100 mL of foam were filtrated, spiked with IS, and extracted via solid-phase extraction including evaporation under constant nitrogen flow as described for the water samples. One third of the water and foam samples from the pilot study were analysed in triplicates. Details can be found in paper **V**.

Soil samples for papers **II** and **IV** were extracted following the protocol of Nassazzi et al. (2022) and started by freeze drying the samples in pre-cleaned 50 mL polypropylene tubes at -90°C for four days. 1 g of homogenized sample was spiked with IS. The samples were then extracted in three cycles with 3 mL of methanol (LC-MS grade; LiChrosolv, Merck, Darmstadt, Germany) under sonication followed by centrifugation. The combined extract was run through an ENVIcarb cartridge (12 cc, 1g, Sigma-Aldrich, St. Louis, MO, USA) and evaporated under constant nitrogen flow before transfer to the vials for LC-MS/MS analysis.

The GF/C filters were extracted following the protocol of Ahrens et al. (2011a) using the dried GF/C filters from the water and foam extractions in paper **V**.

The filters were weighted into a pre-cleaned 15 mL Falcon tube and spiked with IS. The samples were then extracted three times with 3-5 mL of LC-MS grade methanol shaking before being evaporated under constant nitrogen flow and transfer to the vials for LC-MS/MS analysis.

Air field blank samples for papers **II** and **III** were extracted by rinsing the container with three times 2 mL of LC-MS grade methanol into a pre-cleaned 15 mL Falcon tube. The extract was then spiked with IS and evaporated under a constant nitrogen stream before LC-MS/MS analysis.

LC-MS/MS analysis

A volume of 10 μL of extract was injected into the LC-MS/MS operated in scheduled multiple reaction monitoring mode and with negative electrospray ionization. The sample was injected at 40°C on a Phenomenex Gemini 3 μm C18 column including a guard column (Phenomenex KJ0-4282) and a precolumn (Phenomenex Kinetex 1.7 μm C18). The details regarding LC-MS/MS operation and evaluation can be found in papers **II** and **III**.

4.4.2 DOC concentration and DOM composition analysis

DOC samples were analysed using a total organic carbon analyser (Sievers M9 Laboratory Analyzer, BE Analytical Instruments, Boulder, USA), while the DOM molecular composition was analysed using bulk optical properties including absorbance and fluorescence spectroscopy, whereof fluorescence intensity and standard fluorescence indexes were calculated. MilliQ® water blanks were used to correct fluorescence spectra. Details on the DOC and DOM analyses as well as the interpretation of the results can be found in paper **II**.

4.4.3 General water chemistry

Stable water isotope ratios (δ^{18}O and δ^{2}H) as well as cations (sodium, potassium, magnesium, calcium) and anions (fluoride, chloride, nitrate, sulfate, carbonates) were analysed by the commercial laboratory JR-AquaConSol GmbH

in Graz, Austria. For paper **III**, stable water isotope analysis prior to 2022 was conducted as part of the waterworks monitoring program and analysed at the Environmental Isotope Laboratory of the University of Waterloo, Ontario, Canada. Delta notations in per mil were calculated in relation to the Vienna Standard Mean Ocean Water (VSMOW) reference using the following formula:

$$\delta^{18}O[‰] = \left(\frac{\left(\frac{\delta^{18}O}{\delta^{16}O}\text{sample}\right) - \left(\frac{\delta^{18}O}{\delta^{16}O}\text{VSMOW reference}\right)}{\left(\frac{\delta^{18}O}{\delta^{16}O}\text{VSMOW reference}\right)} \right) * 1000$$

4.4.4 Quality control

Quality control was ensured by air field blanks, solvent laboratory blanks, as well as duplicate or triplicate sample analyses for PFAS, DOC, and DOM samples. For the direct injection samples of paper **IV**, MilliQ® from a Milli-Q IQ 7000 Ultrapure Water purification system including a 0.22 µm Millipak Express membrane and a LC-PAK polishing unit (Merck Millipore, Billerica, MA, USA) was used to dilute the samples and respective MilliQ® blanks were included. All glassware and glass-fiber filters were cleaned at 400°C overnight and the glassware was rinsed six times with LC-MS grade methanol prior to analysis. All HDPE bottles, PVC columns and other equipment were cleaned with LC-MS grade methanol before sampling. Nitrile gloves were worn throughout the sampling, experiments, and analysis, and as much PFAS-free equipment as possible was used throughout.

Method detection limits (MDLs) and recoveries were calculated based on a nine-point standard calibration curve (from 0.01 to 100 ng mL^{-1}; $R^2 > 0.98$) and the laboratory blanks. The mean of the laboratory blanks for each PFAS plus 3.3 times the standard deviation of the laboratory blanks were used to calculate the MDLs. If no PFAS were detected in the laboratory blanks, the lowest detected standard calibration curve point was used instead. Details about detection limits, calibration standards, and results of the blanks and recoveries can be found in the respective papers (papers **II**-**V**).

4.5 Data handling and analysis

All data was treated and processed in R Studio® 2024.04.1 Build 748 (R Core Team, 2024). For graphical presentation, non-detects were displayed as zero, whereas for statistical analyses they were replaced by half the respective MDL. Details on specific calculations and respective MDLs can be found in papers **II**-**V**).

4.5.1 Statistical analysis

The statistical methods were selected according to the objectives of each study and are therefore summarized separately below. Detailed descriptions of the statistical analyses can be found in the respective papers, but in brief, in paper **II**, a combined RLQ method and 4th corner analysis with a Monte-Carlo significance test was conducted to assess the relationships between the PFAS concentrations, compound characteristics, as well as different environmental parameters, such as temperature, precipitation, wind speed, or seasonality. Furthermore, a cross-correlation lag-lead analysis was conducted to assess correlations of environmental variables prior or after the sampling against the detected PFAS concentrations. For paper **III**, nonparametric Wilcoxon and Spearman correlation tests were conducted to assess the correlations between the stable water isotope measurements and the detected PFAS concentrations and composition. Non-metric Multi Dimensional Scaling (NMDS) was used to assess the PFAS composition under different conditions. For paper **IV**, nonparametric Wilcoxon and Kruskal Wallis tests were conducted to assess differences in PFAS retardation between new and used sand as well as the two different sites.

CHAPTER 5

Paper Summaries

To answer the research question of this thesis, *"Does MAR act as a source or sink for ambient PFAS concentrations into groundwater systems and drinking water supplies?"*, five studies were conducted and are summarized in the following sections. The full papers can be found in part II of this thesis.

5.1 Overview of CECs in MAR systems

Paper I: Managed aquifer recharge as a potential pathway of contaminants of emerging concern into groundwater systems – A systematic review

Motivation and aim

Groundwater is an often overlooked and important resource. To protect and ensure stable quantity and quality, a common method is to artificially recharge groundwater supplies with surface water via MAR. However, surface waters often contain anthropogenic CECs, such as plastics, pesticides, pharmaceuticals and personal care products, or PFAS. When infiltrating this surface water, MAR might thus act as a shortcut for CECs into groundwater sys-

tems and eventually drinking water supplies. This systematic review assesses the transport of CECs through MAR systems by addressing (1) typical CEC concentrations in surface waters, (2) different factors affecting CEC transport and retention in MAR systems, and (3) major contaminant groups passing through MAR into groundwater systems, as well as possible pre-treatment options.

Methodology

A systematic literature review on CECs in surface waters and MAR was conducted using a targeted search string, and resulting in 1460 screened articles as well as 131 additional papers during an updated screening for new articles towards the end of the writing period. The review and screening was conducted using the PRISMA flow-chart for systematic reviews using databases and registers (Page et al., 2021). This led to a total number of 89 articles included in the review. The details on the methodology of the systematic literature review can be found in paper **I**.

Key findings

- The use of MAR and the number of CECs in surface waters are both likely to increase in upcoming years, which is why regular, site-specific monitoring and seasonal operational planning are likely to become more relevant in the future.

- MAR can act as a shortcut for CECs into groundwater systems. Especially PFAS are an example of (very) mobile and (very) persistent contaminants showing atypical transport patterns during MAR and thus posing a risk for ground- and drinking water contamination. The literature review thus identified PFAS as one of the contaminant groups for which significant knowledge gaps remain, thereby motivating the subsequent studies in this thesis.

- Redesign or adjustment of MAR systems is needed to capture CECs before entering groundwater systems while keeping the system as cost-efficient as possible.

5.2 MAR as a shortcut for PFAS from surface waters into groundwater systems?

Paper II: Seasonal trends of PFAS concentrations in surface waters and fate during Managed Aquifer Recharge (MAR)

Motivation and aim

If surface waters often contain elevated concentrations of PFAS, using these surface waters for direct drinking water production or MAR risks PFAS entering groundwater systems and eventually drinking water supplies. However, there is a lack of research on the variations and sources of PFAS in surface waters to identify periods of increased infiltration risk, as well as on the fate of PFAS during MAR. This study therefore (a) assesses seasonal variations in surface water PFAS concentrations and their correlation with water quality, and (b) determines the annual PFAS load entering groundwater systems via MAR by monitoring two MAR systems in Sweden over two years.

Methodology

Monthly monitoring was conducted at two hydrogeologically contrasting Swedish MAR sites between September 2022 and June 2024, as explained in Section 4.1, combining PFAS, major water ion, DOC and DOM, and stable water isotope analyses. Surface water samples were collected 10 cm below the surface at the inlet to the infiltration ponds, and samples of the artificially recharged groundwater were collected at the wellfield, where the standing water was pumped out prior to sampling. Laboratory analyses and statistical methods were performed as described in Chapter 4.

Key findings

- DOC concentrations at the two investigated sites decrease by approximately 50% during MAR, while PFAS behave fundamentally different with their concentrations staying constant or increasing at the recharge wells compared to the infiltrated water.
- PFAS surface water concentrations at the two MAR sites vary by an order of magnitude and cannot be associated with other seasonal environmental proxies, which makes strategic operation of MAR systems

to mitigate the amount of PFAS infiltrated challenging. MAR is highly site-specific, and there is thus an urgent need for frequent and standardized monitoring for CECs to optimize MAR systems accordingly.

- Possible reasons for increased PFAS concentrations in the artificially recharged groundwater could be accumulation in the aquifer material and seasonal re-mobilization, or additional contaminated groundwater input from point sources.

5.3 Sources and factors affecting PFAS transport under field conditions

Paper III: Resolving PFAS transport in groundwater requires resolving hydrology: Evidence from Managed Aquifer Recharge (MAR) systems

Motivation and aim

PFAS transport in the subsurface remains poorly constrained, due to most studies being conducted under simplified laboratory conditions at unrealistically high PFAS concentrations, as well as because of limited hydrological characterization of groundwater systems. Under field conditions, stable oxygen water isotope ratios ($\delta^{18}O$) can be used as a tool for tracking the movement of surface and groundwater, as well as to determine the origin of groundwater abstracted long distances from the infiltration site (Mook, 2006; Stichler et al., 1986). To assess the movement of contaminants such as PFAS in the subsurface as well as their origin in detail, tracking the infiltrated water using stable water isotopes in MAR systems, combined with detailed monitoring can then help to understand movement of these persistent and mobile contaminants with the water at environmentally relevant concentrations. The aims of this study are therefore to estimate (a) water and PFAS travel times in the subsurface, (b) the relative contribution of the artificially infiltrated water to the natural groundwater system in a thick Esker and (c) the effect of intermittent MAR operation on PFAS levels in the groundwater.

Methodology

A 10-year sampling and analysis campaign for stable oxygen water isotope ratios ($\delta^{18}O$) conducted by the waterworks was combined with two years of

monthly PFAS measurements in surface- and groundwater to determine the dynamics of water recharge, movement, and quality in the Esker. In fall 2023, an additional three months of bi-weekly sampling at additional sampling points between the infiltration site and the wellfield, including detailed analysis of PFAS and stable oxygen water isotope ratios, was conducted. Water travel times and mixing ratios were determined based on the detected $\delta^{18}O$ signals according to Schudel et al. (2002) and Wanner et al. (2011).

Key findings

- Over 500 $\delta^{18}O$ analyses show a unique $\delta^{18}O$ "signature" of the infiltrated water, with enriched heavy stable oxygen isotope ratios in September or October every year caused by evaporation of the surface water. This signature could be traced throughout the infiltration process until the groundwater recharge wells, allowing estimation of water travel times of 6-7 months over approximately 2 km.

- The infiltrated water mixes with and is diluted by the natural groundwater. The stable water isotope data allows quantification of these mixing ratios at the different sampling locations, which aids in identifying local point sources and understanding the impact of the MAR system itself on PFAS concentrations at the wellfield.

- The various analysed sampling locations show a complex picture of PFAS mobility and retention in the subsurface. Intermittent MAR operation leads to remobilization of sorbed PFAS from the aquifer material, while heterogeneous flow pathways in the aquifer and the resulting reduction in air-water interfaces result in low PFAS retardation despite passage of a thick unsaturated zone.

- Paper **III** demonstrates that understanding PFAS transport in MAR systems requires understanding groundwater hydrology, providing the mechanistic basis for interpreting the field observations from paper **II**. The results aid in planning future operation of the existing artificial infiltration scheme through the detailed process understanding and thereby help minimize contaminant concentrations in the artificially produced groundwater.

5.4 Are we simplifying PFAS sorption processes? - A column study

Paper IV: Concentration-dependent PFAS transport during managed aquifer recharge (MAR) - insights from laboratory column experiments

Motivation and aim

PFAS often behave differently from classical persistent organic pollutants and traditional transport and sorption models often do not apply (Sima and Jaffé, 2021). According to Ruffle et al. (2024), increased knowledge on the fate and partitioning of PFAS in aquatic environments, as well as sufficient data on surface water PFAS concentrations in non-hot-spot-areas, are urgently needed to improve the basis for regulations and quality criteria (Ruffle et al., 2024; Sörengård et al., 2022b). The field observations from papers **II** and **III** suggested complex sorption patterns at these low PFAS concentrations. This study therefore tests a range of PFAS spiking concentrations in saturated sandy columns to assess whether PFAS sorption behavior varies at different concentration levels. The aim is to study (a) three different concentrations of PFAS in the inlet water to test for any changes in sorption and release behavior of low, non-spiked inlet water compared to low or high spiking concentrations, respectively, (b) differences in transport of PFAS through completely pristine sand and sand that has already been used in operating MAR basins from both MAR sites, compared to emerging materials as alternatives for sand, such as artificially crushed rock, and (c) the effect of adding different sorbent materials, such as GAC, biochar, and wood bark, on top of the infiltration sand as possible PFAS treatment step in MAR systems.

Methodology

Laboratory flow-through column experiments were conducted under saturated conditions using 16 PVC columns filled with infiltration sand from the two MAR sites (see Figure 4.3). The columns were operated at room temperature with top-down flow in a closed, fully saturated system driven by a hydraulic gradient. The inlet water originated from the two MAR sites and was replaced daily. The experiment was conducted in two time periods first testing unspiked inlet water over a period of eight days, while the second time period included two different spiking tests (1 hour of approximately 40 ng L^{-1} per PFAS and

1 hour of 65 000 ng L^{-1} per PFAS, respectively) using a PFAS spike mixture containing 15 PFAS.

Key findings

- No significant differences in PFAS breakthrough and retardation could be observed between sites, emerging materials, and used or new sand indicating that, under saturated conditions, inlet concentrations and flow velocity have a greater influence than infiltration material.
- At low PFAS concentrations, primarily shorter-chained PFCAs break through, while at the highest spiking concentrations, also short- and long-chained PFSAs break through rapidly. Calculated PFAS retardation factors are highly increased indicating complex sorption behavior during the low-spiking experiment, but decrease to values between 1 and 3 at the high spiking concentrations, showing classical PFAS sorption patterns.
- Long-chained PFCAs and PFSAs are primarily retained in the top layer of the column sand. The results align well with the field observations shown in papers **II** and **III**.
- A nylon mesh used to separate the sorbent from the sand sorbed highest PFAS concentrations followed by GAC, and biochar. Adding a thin charged membrane on top of the infiltration sand could be a treatment option that is easy to remove and regenerate for sites with low ambient PFAS concentrations, but has to be tested further.

5.5 Foam fractionation as pre-treatment coupled to MAR

Paper V: Treatment of low ambient PFAS concentrations in surface water by foam fractionation prior to managed aquifer recharge (MAR)

Motivation and aim

Pre-treating the infiltrated surface water prior to MAR can be a strategy to adapt MAR systems for increasing contaminant concentrations and to meet stricter drinking water quality criteria. FF is an efficient PFAS concentration

technique that utilizes the surfactant properties of many non-polymeric PFAS such as PFCAs and PFSAs. Especially the long-chained PFAS sorb to the air-water interface of the air bubbles in the column and thereby concentrate in the foam by several orders of magnitude. The foam can then be skimmed off and the treated water remains with very low PFAS concentrations. This technique is commonly tested to be effective for highly contaminated water, such as landfill leachate or AFFF plumes (Padhye et al., 2026; Smith et al., 2023c), but has, to our knowledge, not been previously applied to low PFAS concentrations in surface waters. This study therefore aims at testing (1) FF removal efficiencies and feasibility at low ng L^{-1} PFAS concentrations, (2) the impact of a biologically degradable surfactant (Tween® 80) on FF at low ambient PFAS concentrations, and (3) the impact of high particle loads in the water by testing FF on backwash water from rapid sand filters

Methodology

The FF pilot was operated at site B in continuous triplicate runs with (1) river water abstracted prior to rapid sand filters and the MAR system, (2) the same river water as in (1), but with the addition of 10 mg L^{-1} Tween® 80 surfactant, and (3) backwash water from the rapid sand filters. Each water type was tested at 10, 20, and 82 minutes, respectively. PFAS foam, water, and aerosol trap samples were taken at the beginning and the end of each run.

Key findings

- FF is an efficient pre-concentration technique, especially for long-chained PFCAs and PFSAs, and achieves to reach removal efficiencies of 75%-100% even at low ambient PFAS concentrations. In combination with MAR, this represents a cost-effective and promising treatment train for low PFAS concentrations to meet newer drinking water guidelines in the low ng L^{-1} range.
- Generally, intermediate contact times of 20 minutes and addition of the biodegradable surfactant Tween® 80 increase the removal efficiencies, even though highest enrichment factors are reached with the highly particulate backwash water.
- Continuous extraction of the foam from the water surface likely improves removal efficiencies of shorter-chained PFAS.

CHAPTER 6

Results & Discussion

The following sections address the three specific aims outlined in Chapter 3 based on the five conducted studies and their results to provide insights into the fate of PFAS from surface waters into groundwater systems during MAR. The chapter ends with a synthesis and recommendations for meeting current and upcoming PFAS drinking water guidelines, as well as an outlook for future research.

6.1 Aim 1: Drivers of seasonal variability and sources of PFAS in surface waters and groundwater systems

Research on the sources and fate of PFAS during MAR remains limited, as shown in the literature review (paper **I**), especially regarding their seasonal variability in the infiltrated surface water and their removal efficiency during MAR. Within the literature review, a 78% removal efficiency of PFOS during MAR was reported by only one study (Hrkal et al., 2023) within the search string of paper **I**. Most other studies included in the literature review con-

clude that the behavior of CECs, and especially PFAS, in MAR systems is complex and depends on hydrogeology, residence time, and physio-chemical properties of the infiltration layer, as well as degradation processes, redox conditions, and organic matter composition and concentration (Ahmed and Marhaba, 2017; Mishra et al., 2021; Regnery et al., 2017). However, several studies state that MAR can act as a shortcut for (very) mobile and (very) persistent contaminants, such as PFAS, into groundwater systems and drinking water supplies (Collard et al., 2023; Eschauzier et al., 2010; Page et al., 2019; Pramanik, 2015), but understanding of the exact processes is lacking. The literature in paper **I** and the results of paper **II** further emphasize that "classic" contaminants such as nutrients, metals, or natural organic matter can be retained partially or completely in MAR systems (Balke and Zhu, 2008; Fakhreddine et al., 2021; Jokela et al., 2017). The DOC concentrations in paper **II** confirm the typically observed approximately 50% decrease and subsequent stabilization during MAR, while PFAS concentrations remained constant or were increased at the recharge wells compared to the infiltrated water (Figure 6.1). These atypical transport patterns of PFAS during MAR result from their amphiphilic properties, mobility, and persistence and vary considerably among the large group of compounds, as shown in papers **I** and **III**.

However, a detailed understanding of temporal variations in PFAS surface water concentrations can aid in identifying periods of increased contamination risk in MAR systems, which is why an extensive monitoring study of PFAS in surface waters and MAR systems was conducted for papers **II** and **III**. Paper **II** shows that even in surface waters affected primarily by diffuse PFAS sources and with no known "hotspots" in the vicinity, PFAS concentrations varied by an order of magnitude, with occasional peaks of up to 63 ng L^{-1} $\sum_{29}$PFAS (Figure 6.1), which is in the range of concentrations reported in previous studies that typically detect concentrations of about 100 ng L^{-1} (Behrens et al., 2025; Liu et al., 2025; Wu et al., 2025; Zhang et al., 2025). However, previous studies have focused on spatial differences in PFAS concentrations among various surface waters (Ore et al., 2025; Wu et al., 2025; Xiao et al., 2026) or differences between different years to show the gradual shift in composition profiles from primarily legacy PFAS to newer alternatives such as shorter-chained PFCAs and PFSAs or GenX (Celma et al., 2025; Liu et al., 2025; Wu et al., 2025; Xiao et al., 2026), rather than on seasonal monitoring. The

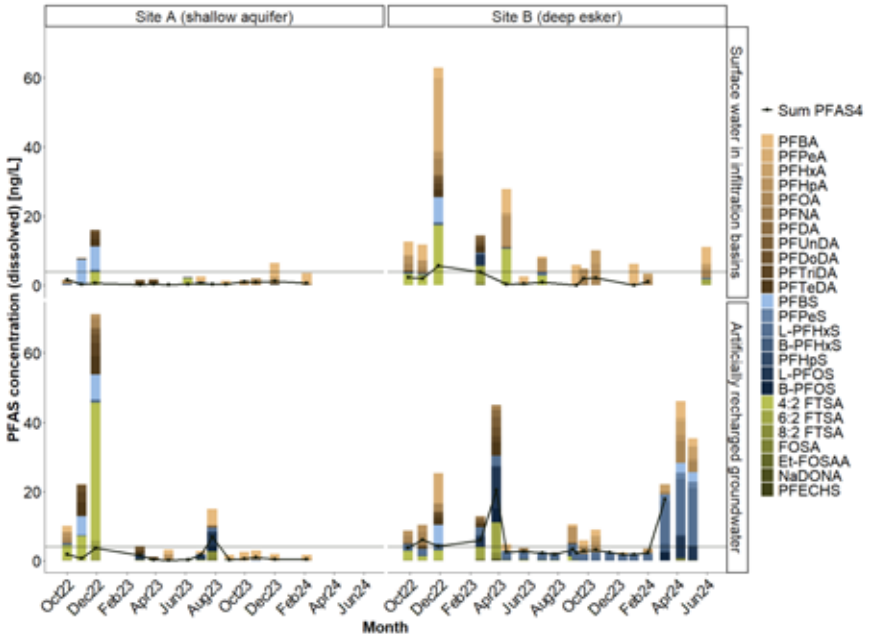

Figure 6.1: PFAS concentrations [ng L^{-1}] in surface waters used for MAR (top) and artificially recharged groundwater (bottom) over the two year monitoring campaign at site A (left) and site B (right). PFCAs are displayed in brown, PFSAs in blue, and precursors, NaDONA, and PFECHS in green, with darker colors showing longer-chained PFAS. $\sum_4$PFAS results are shown as black line with the new Swedish drinking water limit of 4 ng L^{-1} for $\sum_4$PFAS as a horizontal line in black. The figure was reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

monthly to bi-weekly monitoring resolution in papers **II** and **III** further shows that the occasional peaks did not correlate with other measured environmental parameters or seasonality (Figure 6.2 and paper **II**), which indicates that increased PFAS concentrations in surface waters at ambient background sites cannot be predicted by other environmental proxies and instead behave somewhat independently. Moreover, the results in paper **II** do not allow for specific PFAS sources to be identified by fingerprinting the detected PFAS composi-

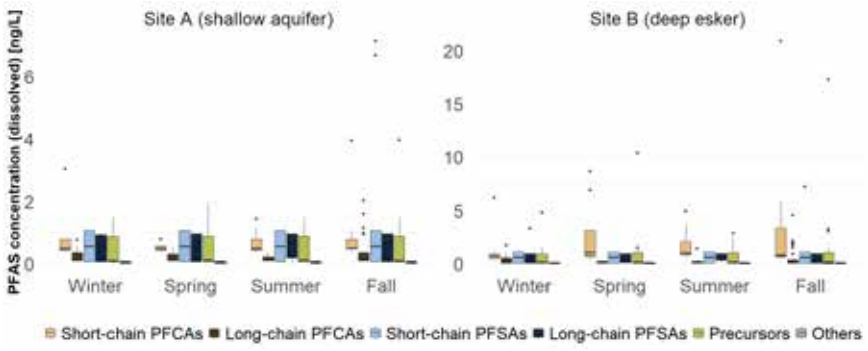

Figure 6.2: Seasonal differences of PFAS concentrations [ng L^{-1}] in surface waters used for MAR at site A (left) and B (right) over the two year monitoring campaign. Grouping the observations by season did not reveal a consistent seasonal pattern at either site. PFCAs are displayed in brown, PFSAs in blue, precursors in green, and NaDONA and PFECHS in grey, with darker colors showing longer-chained PFAS. The figure was reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

tion profiles, since the occasional peaks mostly originated from a variety of diffuse sources, in contrast to studies that identify clear PFAS point sources and "hotspots" based on such fingerprinting (Houtz et al., 2013; Sörengård et al., 2022a; Zhao et al., 2022). The difficulty in predicting PFAS concentration peaks in low-impacted surface waters therefore makes it challenging to operate MAR systems strategically to reduce the amount of PFAS infiltrated.

However, paper **II** indicates possible underlying release mechanisms of previously sorbed PFAS from the subsurface into the groundwater. One such mechanism could be the possible competition for sorption sites between PFAS and surfactant-producing algae blooms at the end of summer at site A, potentially leading to the release of previously sorbed PFAS from the infiltration layer to the artificially recharged groundwater in late summer (Figure 6.1). This was identified as one possible explanation for increased PFAS concentrations at the wellfield. For site B, transport and release processes appeared to be more complex, with possible freeze-thaw cycles and groundwater level changes affecting the amount of air-water interfaces available for PFAS sorption and thereby

potentially releasing pulses of previously sorbed PFAS. Together, these site-specific processes complicate the identification and prediction of PFAS sources and fate, which is why estimating PFAS concentrations entering groundwater systems and eventually drinking water supplies via MAR requires both extremely high-resolution, long-term monitoring with daily to weekly sampling (paper **II**) as well as assessment of the local groundwater travel times and retardation factors (paper **III**). Therefore, paper **III** includes a higher resolution monitoring of site B and is discussed in detail in Section 6.2.

Overall, PFAS concentration peaks at the two investigated sites cannot be predicted using the measured environmental proxies, and their sources and subsequent fate are controlled by site-specific combinations of diffuse surface-water inputs and local seasonal or operational release processes.

6.2 Aim 2: The fate of low PFAS concentrations in MAR systems - towards a detailed process understanding

Paper **II** shows that MAR can act both as a source and a (temporary) sink for PFAS and that PFAS fate in MAR is extremely site-specific, emphasizing the need for high-resolution monitoring at low ambient PFAS concentrations to eventually understand the major pathways for PFAS ending up in potential drinking water sources. The field monitoring in papers **II** and **III** together with the saturated column experiments in paper **IV** provide further insights on the processes controlling transport, fate, and sorption of environmentally relevant PFAS concentrations during MAR. Together, they demonstrate that PFAS fate at ambient background concentrations is governed by the complex interaction between PFAS physio-chemical and sorption properties, competition for sorption sites, hydrogeology, and MAR operation rather than by contaminant chemistry alone.

6.2.1 Mobility of ambient PFAS concentrations in MAR systems

As shown in paper **II**, PFAS concentrations are comparable or even higher in the artificially recharged groundwater compared to the surface water concentrations, indicating that PFAS are mobile in MAR systems. Even though not

all individual PFAS are expected to pass the subsurface at the same speed as the groundwater due to different functional groups, chain length, and resulting sorption characteristics (Kolade et al., 2025; Silva et al., 2021; Xiao et al., 2015; Zhao et al., 2022), $\sum_{29}$PFAS could be placed on the same timeline as groundwater travel times calculated from a high-resolution stable water isotope (δ^{18}O) dataset (Figures 6.1 and 6.3), as shown in paper **III**.

The observations in paper **III** indicate that, $\sum_{29}$PFAS appear to be highly mobile with retardation factors close to one, even at low PFAS concentrations and while passing a thick unsaturated zone at site B, due to preferential flow paths and therefore reduced air-water interfaces in the heterogeneous aquifer (Guo et al., 2020; Zeng and Guo, 2023). Fast PFAS breakthrough could also be observed for both sites in the saturated column experiments presented in paper **IV**, especially for the shorter-chained, more mobile PFCAs and PFSAs. Already at low PFAS concentrations, PFAS were shown to be highly mobile with fast breakthrough curves, but increased retardation factors. Even though the column experiments cannot fully reproduce the hydrogeological heterogeneity, preferential flow paths, and fluctuating groundwater conditions observed in full-scale MAR systems, they isolate individual transport processes under saturated conditions. The column experiments therefore complement the field observations showing that ambient PFAS concentrations remain highly mobile during MAR despite partial sorption of long-chained compounds.

Another contributor to increased legacy PFAS concentrations in the artificially recharged groundwater is contaminated groundwater input from point sources at site B impacting the natural groundwater that mixes with the artificially recharged water prior to extraction at the wellfield (paper **III**). Hereby, δ^{18}O signals (Figure 6.3) provide great insights into local water travel times and surface water-groundwater mixing ratios to identify locations with higher contributions of the natural groundwater, where local point sources could have a greater effect. One such location is a capped, back-filled gravel pit located downstream of the infiltration basins at site B (Figure 4.2). The calculated water transit times (Figure 6.3) and the mixing ratios calculated from the long-term δ^{18}O analysis results in paper **III** thus strongly support source identification by locating sampling points with the highest groundwater contribution (paper **III**).

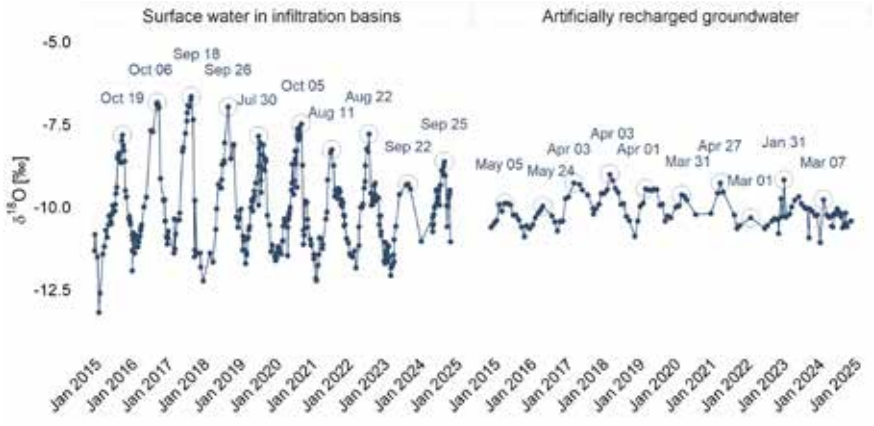

Figure 6.3: Ten years of stable oxygen water isotope signals (δ^{18}O) in the surface water used for MAR (left) and the artificially recharged groundwater at the wellfield (right) at site B. Circles mark the sampling dates with the annual peak of enriched δ^{18}O signal. The figure was reprinted from paper **III** (in review) with permission from the American Chemical Society, copyright 2026.

6.2.2 Retention and accumulation of PFAS in the subsurface

A critical part for the retention of PFAS in MAR systems appears to be the clogging layer, the so-called "Schmutzdecke". This layer is built up by various factors, such as the microbial and biologically active community within the Schmutzdecke, organic matter type and content as well as residence time (Bertelkamp et al., 2012; Regnery et al., 2017), since the Schmutzdecke is scraped off in regular intervals at operating MAR sites to prevent clogging of the infiltration basins. As shown in papers **II**, **III** and **IV**, the Schmutzdecke and the air-water interface in the unsaturated zone mostly sorb the more hydrophobic, long-chained PFCAs and PFSAs and especially PFOS (Figure 6.4), while the shorter-chained PFCAs pass through. When all sorption sites are taken, competitive sorption of for example other surfactants produced by algae blooms (paper **II**) or long-chained PFAS (papers **III** and **IV**), can lead to subsequent release of previously sorbed PFAS to the subsurface (Niarchos

et al., 2023). The column experiments further demonstrate that PFAS retardation cannot simply be transferred between concentration ranges (paper **IV**). Classical sorption behavior observed at highly contaminated sites therefore does not necessarily describe PFAS transport at environmentally relevant concentrations, which appear to be more complex. Moreover, in the field studies (papers **II** and **III**), the more complex field conditions further show that not only the Schmutzdecke and the unsaturated zone, but also varying water levels as well as different point sources play an important role in retention and subsequent release of PFAS in the subsurface.

One such PFAS release process can be collapsing air-water interfaces during wetting and drying cycles (Higgins and Luthy, 2006; Kolade et al., 2025), which can be caused by changing groundwater levels or during intermittent MAR operation. During drainage, the total air-water interfacial area expands, trapping PFAS in the unsaturated zone. Re-wetting then rapidly destroys these air-water interfaces and was shown to release pulses of the previously sorbed long-chained PFAS (paper **III**). Especially at the wells between the infiltration site and the wellfield investigated in paper **III**, significantly increased PFAS concentrations ($p < 0.001$) could be observed around each of the three times where the infiltration is turned on again after longer shut-off periods between summer 2023 and 2024. While previous literature has observed PFAS remobilization through wetting and drying cycles (Cáñez et al., 2021; Hubert et al., 2025; Kolade et al., 2025; Maizel et al., 2021), causing this effect by intermittent MAR operation and already at low ambient PFAS concentrations has not been observed before. This further emphasizes that sorbed PFAS in the subsurface can, even at low input concentrations, pose a risk to underlying groundwater systems for years to decades (Høisæter et al., 2019; McFarlan and Lemke, 2024; Röhler et al., 2021; Weber et al., 2017; Xiao et al., 2015).

Natural attenuation in MAR systems therefore appears to be temporary rather than permanent, particularly for long-chained PFAS retained in the infiltration layer and unsaturated zone (papers **II**-**IV**), but also at high PFAS concentrations leading to competition for sorption sites and subsequent release (paper **IV**). Intermittent MAR operation further amplifies PFAS release pulses, resulting in that continuous MAR operation is favorable (paper **III**). Furthermore, while previous understanding of PFAS transport has been dominated

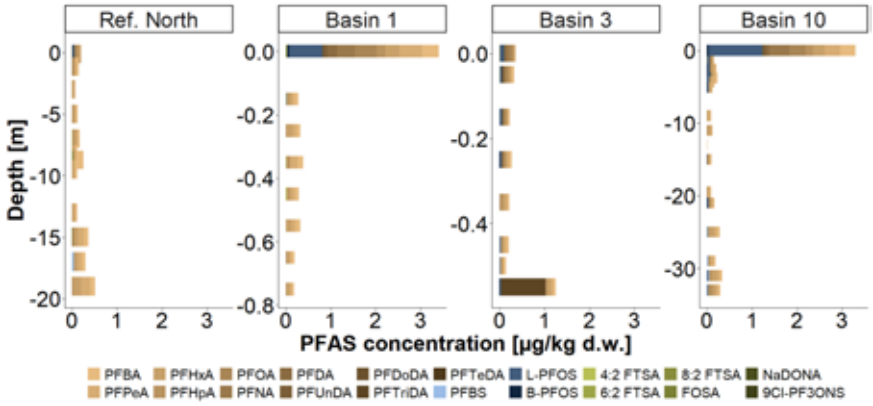

Figure 6.4: PFAS concentrations [μg kg^{-1} dry weight] in depth-discrete subsamples of the infiltration basin sand at site B. PFCAs are displayed in brown, PFSAs in blue, and precursors or other PFAS in green, with darker colors showing longer-chained PFAS. The figure was reprinted from paper **II** (Mumberg et al., 2026), CC BY 4.0 license.

by laboratory-derived sorption behavior often assessed at high PFAS concentrations (Bertelkamp et al., 2012; Høisæter et al., 2019; Weber et al., 2017; Xiao et al., 2026), this thesis shows that, at environmentally relevant concentrations, PFAS concentration levels, field-scale hydrology and MAR operation become equally important controls on PFAS fate and transport. Future experimental studies should therefore focus on investigating PFAS transport under environmentally relevant concentrations, since extrapolation from highly contaminated laboratory studies may misrepresent field behavior at low ambient concentration sites.

6.2.3 Sources and factors affecting PFAS transport under field conditions

Paper **III** demonstrates that local PFAS point sources additionally affect PFAS concentrations in the artificially recharged groundwater depending on

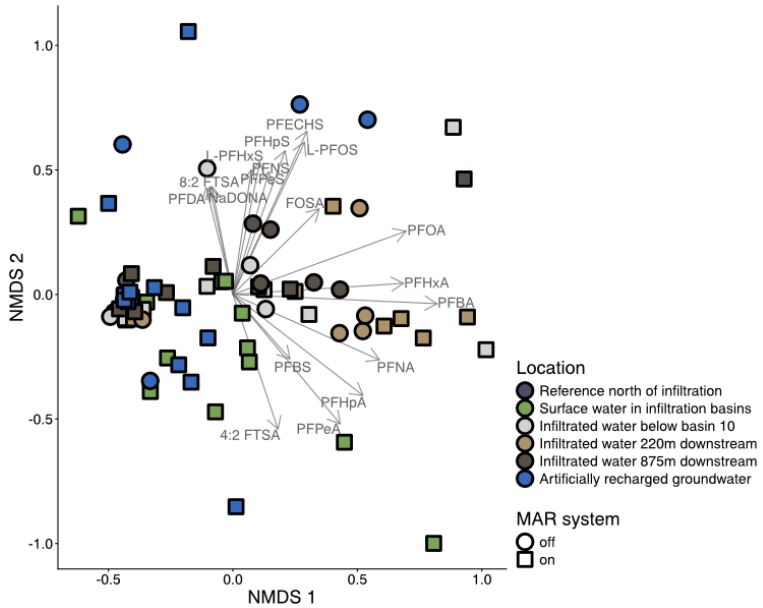

Figure 6.5: Non-metric multidimensional scaling (NMDS; Bray-Curtis, stress = 0.152, k = 2) converting samples from the five sampling locations over the two-year monitoring period at site B into a distance matrix. Brown colors show the two sampling locations downstream of the infiltration site, while the blue color indicates the effect at the wellfield. Circles and squares mark if the MAR system was in operation or not. The figure was simplified and reprinted from paper **III** (in review) with permission from the American Chemical Society, copyright 2026.

MAR operation at site B. The capped, back-filled gravel pit, identified in paper **III**, leaches primarily PFCAs such as PFBA, PFHxA, and PFOA into the groundwater that then mixes with the artificially infiltrated groundwater as shown at the sampling locations 220 m and 875 m downstream of the infiltration site, respectively (Figure 6.5). A second point source contributing primarily the AFFF-typical PFSAs (PFBS, PFHxS, and PFOS) at site B, was identified in paper **III** to be a PFAS plume originating from a military airbase

site that has been used for firefighting training (Figure 4.2). During continuous MAR operation at site B, the increased relative groundwater gradient directs the AFFF plume away from the wellfield (Uebel, 2025). Moreover, the PFSA concentrations in the natural groundwater are diluted by the infiltrated surface water. However, when the MAR system is not in operation for about one to two months, the lower groundwater level gradient allows the highly PFAS-contaminated AFFF plume to reach the wellfield (Sörengård et al., 2022a; Uebel, 2025), resulting in increased concentrations of especially the regulated, legacy PFSAs (Figure 6.5). Consequently, reliable prediction of PFAS transport behavior requires detailed characterization of groundwater flow, local PFAS sources, MAR operation, as well as site-specific assessment.

6.3 Aim 3: Optimizing MAR systems for effective PFAS removal - operational strategies and pre-treatment options

The literature review in paper **I** showed that MAR can act as a shortcut for PFAS into groundwater systems and that redesign or operational adjustment of MAR systems is needed to be able to retain PFAS before they enter groundwater systems while retaining the cost advantages of MAR. The results from papers **II**–**V** then allow this general recommendation to be developed into site-specific operational and pre-treatment strategies.

For site A, raw surface water PFAS concentrations were generally low and did not exceed the new Swedish $\sum_4$PFAS threshold. In the artificially recharged groundwater retrieved at the wellfield of site A, the $\sum_4$PFAS threshold was only exceeded in fall (paper **II**) and possibly related to the release of previously sorbed PFAS from the aquifer material due to surfactants produced by algae blooms in the infiltrated surface waters that compete for the same sorption sites as PFAS. Based on these findings, a possible operational strategy could therefore be to monitor the algae blooms in the surface water and either reduce the infiltration volume during these weeks, use alternative surface water sources, if possible, or couple respective (pre-)treatment during the algae bloom times (papers **II**, **IV** and **V**).

At site B, PFAS concentrations were in general slightly higher, but MAR rather acted as dilution method for PFAS leaching into the groundwater from

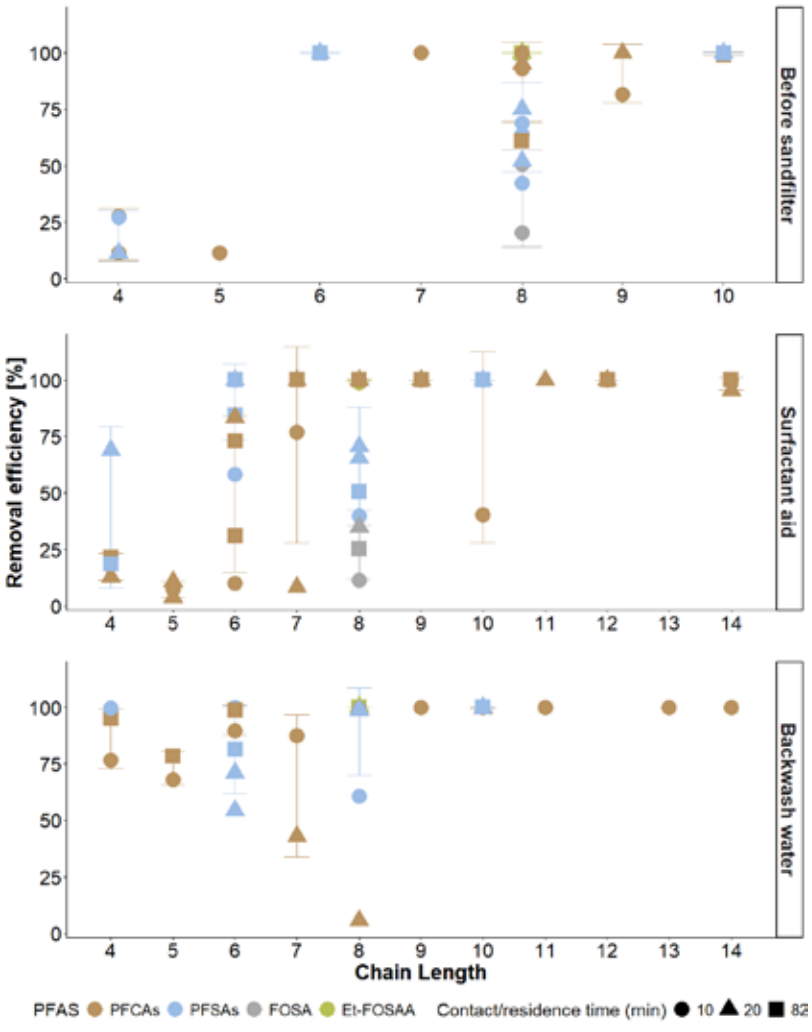

Figure 6.6: PFAS removal efficiencies [%] against alkyl chain length for each tested water type (top to bottom) and contact time (symbols). PFCAs are displayed in brown, PFSAs in light blue, FOSA in grey, and Et-FOSAA in green. The figure was reprinted from paper **V** (manuscript in preparation).

a back-filled gravel pit (paper **III**). Moreover, paper **III** also showed that the increased hydraulic gradients generated by the artificial infiltration redirected an AFFF plume from a military firefighting training site away from the wellfield and therefore reduced PFAS concentrations in the artificially recharged groundwater when the MAR system was operated. Finally, intermittent MAR operation and the corresponding changes in groundwater levels led to the release of previously sorbed PFAS from the collapsing air-water interfaces, making intermittent shutdowns an operational risk for water producers. Consequently, at site B, continuous MAR operation aids to keep the PFAS concentrations at the wellfield low and is therefore recommended.

Moreover, MAR coupled to simple pre-treatment, such as a sorbent layer on top of the infiltration sand (paper **IV**) or continuous FF (paper **V**), as possible multi-barrier treatment train was shown to be effective even at low ambient PFAS concentrations in surface waters used for MAR (Figure 6.6). Especially the cost-efficient FF coupled to MAR could further reduce the amount of PFAS in the groundwater to eventually reduce treatment costs due to lower PFAS loads at the drinking water treatment plant (Franke et al., 2021; McCleaf et al., 2023; We et al., 2024). Previous studies have mainly focused on FF for highly contaminated waters, such as landfill leachate or AFFF-impacted waters, where removal efficiencies are generally high due to the elevated PFAS concentrations (McCleaf et al., 2021; Smith et al., 2022, 2023c). The results of paper **V** demonstrate that FF can also achieve high removal efficiencies for ambient PFAS concentrations in the low ng L^{-1} range, and especially for the four regulated legacy PFAS (Livsmedelverket, 2022; Miljøministeriet, 2024; United States Environmental Protection Agency, 2024). In detail, the FF pilot study in paper **V** shows that FF performance depends on both contact time, with 20 minutes contact time yielding the highest removal efficiencies, and on the characteristics of the treated water (Figure 6.6). Backwash water containing elevated particle concentrations achieved particularly high enrichment factors, indicating that suspended particles may provide additional sorption sites that facilitate PFAS transfer into the foam (McCleaf et al., 2017; We et al., 2024). However, the increased variability and difficulties in closing the PFAS mass balance also indicate that particle-associated transport processes require further investigation before full-scale implementation. The addition of the biodegradable surfactant Tween® 80 increases PFAS removal efficiencies to 75-100% across the tested contact times of 10, 20, and 82 minutes and

PFAS chain lengths (Figure 6.6), although enrichment factors are lower than without surfactant addition.

As demonstrated in paper **V**, FF can act as promising complementary pre-treatment step that can extend the applicability of FF to drinking water production systems relying on surface water and MAR, particularly where operational adjustment of MAR alone cannot reliably prevent PFAS input. Moreover, reduced PFAS inputs into the MAR system due to FF pre-treatment, will eventually reduce the amount of PFAS accumulating in the clogging layer and unsaturated zone. This will then, in a long-term perspective, decrease the risk for remobilization peaks, as well as allow MAR systems to rather act as a dilution technique to reduce PFAS concentrations in groundwater systems, than a shortcut for PFAS into groundwater systems. At sites where MAR primarily acts as a hydraulic barrier or dilution system (papers **II** and **III**), such as Site B, pre-treatment may thus further enhance the beneficial effects of MAR by reducing the PFAS mass entering the aquifer while maintaining the hydraulic advantages of artificial recharge. However, full-scale implementation would require evaluation of concentrate management, energy demand, process stability, and the removal of shorter-chained PFAS (Franke et al., 2021; Smith et al., 2023c; We et al., 2024).

6.4 Meeting current and upcoming drinking water guidelines

The recommendations from paper **I** that MAR systems need to be redesigned for CEC removal are supported by the field and experimental findings of papers **II**–**IV**, which identify operation, hydrogeology, contaminant concentraion levels, and site-specific sources as factors impacting the fate of PFAS in MAR systems. The new or upcoming stricter PFAS drinking water guidelines result in that low ambient PFAS concentrations previously regarded as environmentally insignificant background concentrations are now operationally relevant for drinking water production. This is particularly important for MAR systems, where low ambient PFAS concentrations can be continuously introduced into groundwater systems over long periods of operation. The challenge is therefore no longer whether PFAS occur in source surface waters, but how drinking water production systems can be adapted to manage their ubiquitous presence at environmentally relevant concentrations.

The results of this thesis show that MAR alone should not be regarded as a reliable PFAS treatment technology. While the infiltration process can efficiently reduce DOC and other conventionally removed compounds (paper **I** and **II**), PFAS behaved fundamentally different, remaining highly mobile under field conditions and occasionally increasing in the recovered groundwater due to competition for sorption sites, intermittent MAR operation and groundwater level changes, or local point sources (papers **II**-**IV**). Consequently, no single operational strategy can optimize PFAS removal across all MAR systems. At some sites, targeted monitoring and temporary operational adjustment may help reduce infiltration during periods of increased risk. At other sites, continuous MAR operation can aid to dilute PFAS groundwater concentrations or redirect PFAS plumes in the groundwater away from the wellfield, as well as reduce PFAS remobilization associated with groundwater-level fluctuations. This emphasizes that effective PFAS management should combine site-specific strategic operation of MAR systems with detailed monitoring and, where necessary, complementary (pre-)treatment technologies (papers **I**-**V**).

More generally, the findings demonstrate that future compliance with increasingly stringent drinking water guidelines will require a shift from passive groundwater recharge towards actively managed MAR systems. Consequently, MAR should be regarded as one component of a broader treatment train rather than as a stand-alone PFAS treatment technology.

6.5 Limitations and future research

As demonstrated in this thesis, extensive and high-resolution PFAS monitoring at the field scale is urgently needed to equip MAR systems as a part of a PFAS treatment train (papers **I**-**III**). Future PFAS drinking water guidelines, that are likely to be even lower than the currently implemented ones (Figure 2.3) and possibly include a broader range of PFAS, will pose analytical challenges of detecting more target PFAS at lower concentrations. However, technological advances could also decrease MDLs and thereby shorten analysis time and decrease contamination problems during extraction by allowing for direct injection without pre-concentration, even in the low ng L^{-1} range. This will eventually also reduce costs and thereby decrease constraints in temporal resolution. Future work should therefore increasingly focus on environmentally relevant PFAS concentrations rather than highly contaminated sites.

Furthermore, this thesis shows that predicting PFAS transport requires resolving groundwater hydrology (paper **III**). Future studies should therefore combine long-term PFAS monitoring with detailed hydrological characterization and a better understanding of the unsaturated zone, including groundwater flow paths, mixing processes, and temporal variations in groundwater levels, to improve field-scale understanding of PFAS transport. Moreover, MAR systems differ substantially in hydrogeology and operation. Investigating additional MAR concepts such as for example bank filtration systems would determine whether the mechanisms identified in this thesis are transferable beyond the basin infiltration systems investigated here.

Future research on integrating MAR into PFAS treatment trains, should both look at optimizing the Schmutzdecke operationally and with additional sorbent materials, as well as at scaling up the sorbent tests or the sorption to the nylon mesh in paper **IV** and the FF pilot in paper **V** to full-scale.

An interesting future question in regard to the Schmutzdecke could be if the clogging layer can be utilized as first PFAS removal step by optimizing the renewal cycles to avoid the release of PFAS from the Schmutzdecke. This is a challenging question, since the clogging layer has to be built up and contain sufficient amounts of organic matter to effectively sorb the long-chained PFAS, but should not be saturated with PFAS to avoid breakthrough. Furthermore, formation of the clogging layer depends on the organic matter content of the respective infiltrated surface water. During the saturated column experiments in paper **IV**, sorption to the top layer was observed already after the few weeks of operation with, but with no significant difference in PFAS removal between new sand and aged material. However, extremely high PFAS concentrations were used during the spiked tests of the column experiments in paper **IV**. Moreover, due to the low removal efficiencies of the clogging layer, this would rather be an optimization step in an existing system than a sophisticated PFAS treatment technique. Another question would be how to handle the scraped off clogging layer or added sorbent material if, depending on the PFAS concentrations, it classifies as contaminated waste. However, the measured $\sum_{29}$PFAS concentrations in the Schmutzdecke in paper **II**, were below current EU regulation for persistent organic pollutants in waste (European Parliament and of the Council of the European Union, 2019; Naturvårdsverket, 2025), but could exceed the limits when adding a sorbent layer (paper **IV**).

When scaling up the FF pilot, an interesting approach could be to utilize the foam that is naturally generated at the inlet to the rapid sand filters and to test if the biodegradable surfactant Tween® 80 can directly be added to the water in the sand filter basins. Further studies would then need to determine whether the foam building up on the rough concrete walls of the basins can be removed automatically and at which intervals. Finally, disposal and treatment of the foam concentrate by electrochemical oxidation or other integrated treatment trains (Smith et al., 2023a,b) would need to be investigated.

Ultimately, future research should focus on integrating hydrogeological assessment, optimized MAR operation, and complementary PFAS (pre-)treatment technologies into robust, site-specific treatment trains capable of meeting increasingly stringent PFAS drinking water guidelines.

CHAPTER 7

Conclusion

This thesis investigated the research question *"Does MAR act as a source or sink for ambient PFAS concentrations into groundwater systems and drinking water supplies?"* by assessing PFAS prior to, during, and after MAR in detail.

In paper **I**, two main processes that improve the CEC removal efficiency of MAR were identified. Those were (1) increased amounts of electron deficient organic matter and the formation of a clogging layer at the bottom of the infiltration basins, and (2) increased subsurface residence times due to lower infiltration rates. However, the literature review also showed that studies on the fate and transport of CECs and especially PFAS in MAR systems remain limited, even though MAR globally is a widely-used water treatment strategy and CECs are ubiquitously present in surface waters. These knowledge gaps formed the basis for the subsequent field and experimental investigations presented in papers **II**-**IV**.

Overall, seasonal PFAS peaks at the two investigated sites could not be predicted using the measured environmental proxies, and their sources and transport were controlled by site-specific combinations of diffuse inputs, local groundwater contamination, and operational or seasonal release processes. Moreover, there was no significant correlation between the amount and type of organic matter in the infiltrated surface water and the corresponding PFAS concentrations, but the Schmutzdecke sorbed especially PFSAs and long-chained PFAS, while shorter, more mobile PFCAs passed through the clogging layer (papers **II**-**IV**). However, fluctuations in temperatures across the freezing point and infiltration of algae blooms that possibly compete for the same sorption sites as PFAS and thereby cause unintentional desorption and release pulses of PFAS to the underlying groundwater were discussed in paper **II**. Additionally, intermittent MAR operation causes increased PFAS concentrations in the artificially recharged groundwater due to two effects, as shown in paper **III**: (1) the changes in groundwater levels could lead to the release of previously sorbed PFAS from the collapsing air-water interfaces, and (2) the changed groundwater level gradients allowed an AFFF plume at site B to reach the wellfield for the artificially recharged groundwater when the infiltration was turned off, while the increased groundwater levels under MAR operation directed the plume away from the wellfield. Consequently, PFAS transport could not be explained by compound characteristics alone but required detailed knowledge of local hydrology as well as MAR operation.

Together, Papers **II**–**IV** showed that natural attenuation of PFAS during MAR is limited and reversible. These results further emphasize that the question whether MAR acts as a source or sink for PFAS remains highly site-specific with local groundwater levels and hydrology, infiltration rates and operation, PFAS point sources in the vicinity, PFAS concentration levels, the composition of the Schmutzdecke, as well as seasonal effects in the surface water impacting the removal efficiency of PFAS. MAR can simultaneously act as a partial sink for PFAS in the clogging layer and can dilute the underlying groundwater if surface water PFAS concentrations are lower than the groundwater concentrations, but can also act as a PFAS source if surface water PFAS concentrations are higher, show seasonal peaks or if previously sorbed PFAS are remobilized years to decades later from the infiltration material. Overall, MAR can therefore not be classified consistently as either a source or a sink for PFAS.

With increasingly stricter drinking water guidelines limiting certain PFAS to the low ng L^{-1} range, small changes in reported PFAS concentrations or their potential treatment can have a large impact. Moreover, these stricter PFAS drinking water guidelines can eventually lead to that MAR may increasingly be regarded as a potential contaminant pathway rather than a simple, cost-effective solution for water treatment. This eventually creates a trade-off between water-supply security and the protection of drinking-water quality, a challenge that may become even more pronounced in the future with climate change and increasing environmental pollution of CECs such as PFAS. To equip MAR systems for the future, high-resolution monitoring as well as investigation of MAR as part of a drinking water treatment train are urgently needed. Apart from recommendations on strategic operation and optimization of MAR systems, paper **V** assessed the applicability of FF as one possible, cost-efficient method for low ambient PFAS concentrations in the MAR raw water. Since during FF, PFAS are highly concentrated into a comparatively small foam fraction, the following destructive treatment can be applied to substantially smaller volumes and higher PFAS concentrations, thereby reducing the overall energy demand and treatment costs compared with direct destruction of the full water volume. Nevertheless, FF should, similarly to MAR, not be regarded as a stand-alone treatment technology, since the generated foam still requires PFAS destruction, but as one possible step in a drinking water treatment train.

Finally, the fate of ambient PFAS concentrations during MAR is governed by the interaction between PFAS physio-chemical properties and concentrations, hydrogeology, as well as MAR operation, meaning that future drinking water protection requires actively managed, site-specific treatment trains rather than reliance on natural attenuation alone.

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CHAPTER 8

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