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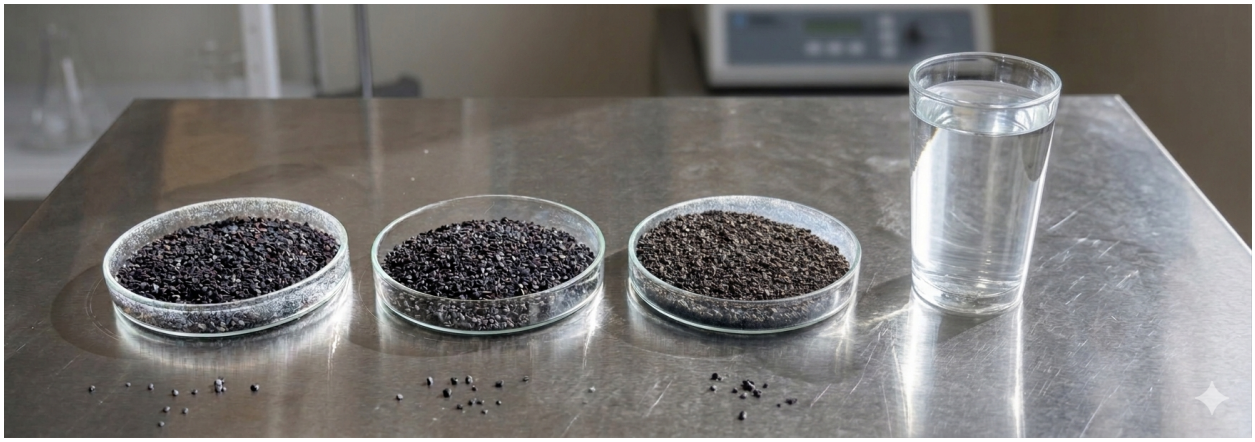

PFAS Removal in Drinking Water Treatment

Evaluating Granular Activated Carbon Performance Across Filter Conditions and Compound Chain Lengths

Master's thesis in Infrastructure and Environmental Engineering

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CHALMERS UNIVERSITY OF TECHNOLOGY
Gothenburg, Sweden 2026
www.chalmers.se

Master's thesis 2026

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Master's Thesis 2026
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Cover: Picture of fresh, regenerated and aged granular activated carbon that was used in the laboratory experiments in this work.

Typeset in LaTeX
Printed by Chalmers Reproservice
Gothenburg, Sweden 2026

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Abstract

Safe drinking water requires the effective removal of persistent chemical contaminants, of which per- and polyfluoroalkyl substances (PFAS) represent a particularly concerning group due to their environmental persistence, resistance to degradation, and documented toxicity at low concentrations. This study investigated the occurrence of PFAS in drinking water systems, evaluated the limitations of conventional and advanced treatment methods, and examined how the condition of granular activated carbon (GAC) filter media affects PFAS removal efficiency.

Batch adsorption experiments were conducted using fresh (F-GAC), thermally regenerated (R-GAC), and aged (A-GAC) filter media across a range of PFAS compounds varying in carbon chain length, including the ultra-short-chain TFA (C2) through to the long-chain PFOA (C8). Removal efficiency, adsorption kinetics, and isotherm behaviour were analysed and compared across filter conditions.

The results demonstrated that GAC filter condition has a substantial impact on removal performance. Fresh GAC achieved >99% removal for all analysed PFAS compounds, with kinetics well described by the pseudo-first-order model, consistent with rapid adsorption onto abundant accessible surface sites. Regenerated GAC retained considerable removal capacity, though with greater dependence on chain length and kinetics better described by the pseudo-second-order model, reflecting increased surface heterogeneity and more complex adsorption interactions following thermal regeneration. The aged GAC filter showed severely limited capacity, achieving meaningful removal only for PFOA, while short-chain and ultra-short-chain compounds exhibited desorption behaviour, with effluent concentrations exceeding influent concentrations. Across all conditions, adsorption affinity and capacity increased with carbon chain length, consistent with the role of hydrophobic interactions as the dominant adsorption mechanism.

These findings highlight the critical importance of filter condition in drinking water treatment and suggest that aged GAC may pose an active risk of PFAS release. Timely regeneration or replacement of GAC filter media is essential for maintaining effective PFAS removal, particularly for shorter-chain compounds.

Keywords: PFAS, granular activated carbon (GAC), adsorption, drinking water treatment, thermal regeneration, adsorption kinetics, chain length, removal efficiency, water treatment

Acknowledgements

This master's thesis was conducted at the Department of Architecture and Civil Engineering at Chalmers University of Technology, within the Master's Programme in Infrastructure and Environmental Engineering. The work was carried out between September 2025 and June 2026 and encompasses 60 credits.

I would like to express my gratitude to my supervisor and examiner Thomas Pettersson for his guidance and support throughout this work. I would also like to extend my sincere thanks to my co-supervisor Amir Saeid Mohammadi for his enthusiasm and dedication to this project, and for his greatly appreciated assistance with the laboratory work, experimental design, and interpretation of results. I am also grateful to Olof Bergstedt at Kretslopp och Vatten for generously providing the filter media used in the experiments. Finally, I would like to thank all colleagues at the Division of Water Environment and Technology for their support and encouragement throughout the course of this work.

Fatimah Al-Nuwab, Gothenburg, June 2026

List of Acronyms

Below is the list of acronyms that have been used throughout this thesis listed in alphabetical order:

AC	Activated Carbon
AE	Anion Exchange
BSE	Backscattered Electron
CEC	Chemical of Emerging Concern
DAF	Dissolved Air Floatation
DOM	Dissolved Organic Matter
DWTP	Drinking Water Treatment Plant
EBCT	Empty Bed Contact Time
FF	Foam Fractionation
GAC	Granular Activated Carbon
GMF	Granular Media Filtration
HI	Hazard Index
LC-MS	Liquid Chromatography–Mass Spectrometry
MCL	Maximum Contaminant Level
MF	Micro Filtration
MRM	Multiple Reaction Monitoring
NF	Nano Filtration
PAC	Powdred Activated Carbon
PFAS	Per- and Polyfluoralkyl Substance
PFCA	Perfluoroalkyl Carboxylic Acids
PFO	Pseudo-First-Order
PFSA	Perfluoroalkyl Sulfonic Acids
PPCP	Pharmaceuticals and Personal Care Product
PSO	Pseudo-Second -Order
RE	Removal Efficiency
RO	Reverse Osmosis
RPF	Relative Potency Factor
SDG	Sustainable Development Goal
SE	Secondary Electron
SEM	Scanning Electron Microscopy
UF	Ultra Filtration
UV	Ultraviolet

Parameters

Below is a list of the parameters that have been used throughout this thesis.

Parameters

C_t	Concentration at time t
q_t	Adsorption capacity on adsorbent at time t
k_1	Adsorption mass transfer coefficient for PFO
k_2	Adsorption mass transfer coefficient for PSO
K_L	Langmuir adsorption coefficient
K_F	Freundlich adsorption coefficient
n	Sorption intensity in Freundlich model
m/z	Mass-to-charge ratio

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1

Introduction

Clean drinking water is an essential factor for all humans, and the UN sustainable development goals (SDGs) make it clear that it is a priority. The clean water goals and specifically the target 6.1 states that by 2030 we should "achieve universal and equitable access to safe and affordable drinking water for all ". This means that drinking water should be provided to all free of microbial and chemical contamination (UN-Water, 2021). Long term exposure to chemicals can lead to serious health issues, therefore it is important to ensure that the provided drinking water does not contain harmful chemicals. In order to achieve drinking water free of chemicals it is important to manage and monitor it, which should be done by following regulations and also control the removal of chemicals in drinking water treatment plants (DTWP). (World Health Organization, 2022). However, conventional drinking water treatment plants often do not remove micropollutants effectively, making it essential to understand the mechanisms governing their removal and to evaluate treatment processes capable of reducing their concentrations.

Among the chemical contaminants of growing concern in drinking water systems are per- and polyfluoroalkyl substances (PFAS), a large group of synthetic compounds characterised by their exceptional chemical stability and persistence in the environment. Despite increasing regulatory attention, PFAS have been detected in water sources worldwide, in many cases at concentrations exceeding health-based guideline values. Understanding the occurrence of PFAS in drinking water and evaluating effective treatment strategies for their removal is therefore of considerable importance.

1.1 Background

In recent years, increasing attention has been directed towards chemical hazards in raw water sources which can steam from anthropological sources called chemicals of emerging concern (CECs). When these CECs reach water bodies used for drinking water production, they can potentially compromise the water quality and pose risks for human exposure. Some of the different types of CECs found in the waters are pesticides, pharmaceuticals and personal care products (PPCPs), flame retardants and per- and polyfluoroalkyl substances (PFASs) (Tröger et al., 2021). Many of these substances have guideline or health based values derived from their toxicity, representing concentrations that are not expected to pose a significant health risk

over a lifetime of drinking water consumption. Exceeding these values over prolonged or lifelong exposure may increase the likelihood of adverse health effects (World Health Organization, 2022).

One class of CEC that are highly persistent are PFAS, which are synthetically made and have been used extensively since the 1950s. They have a wide application in both industry and consumer products for instance in fire-fighting foam, nonstick coating, electronics and waterproof fabrics due to their exceptionally strong carbon-fluoride bonds making them stable, hydrophobic and resistant to both chemical and biological degradation (Gaines, 2023). This stability also means they accumulate in the environment and the main pathway for PFAS into the human body is through food and drinking water (European Parliament and Council of the European Union, 2020). Due to their persistence and toxicity they are being phased out and limit values in drinking water is now set (Sveriges geologiska undersökning, n.d.). Under the Stockholm convection some of the most widely used PFAS, such as the traditional long chained perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), commonly referred to as legacy PFAS are now being heavily restricted (United Nations Environment Programme, Persistent Organic Pollutants Review Committee, 2023).

To mitigate risks with these legacy PFAS, newer alternatives have emerged consisting of shorter carbon chains and are supposed to have similar properties but less toxic and persistent. There are especially four new alternatives that have been most widely used namely, hexafluoropropylene oxide-dimer acid (HFPO-DA, commercial name: GenX), dodecafluoro-3H-4,8-dioxanonanoate (ADONA), 6:2 chlorinated polyfluoroalkyl ether sulfonate (6:2 Cl-PFAES) and 6:2 fluorotelomer sulfonamide alkylbetaine (6:2 FTAB). HFPO-DA and ADONA are used mostly to replace PFOA while the other two mentioned replace PFOS (Cheng et al., 2025). Even though these PFAS substitutes are intended to be less toxic, emerging research suggests that their toxicity and degradation rates may have been underestimated (Hamid et al., 2024; Shi et al., 2024).

Several PFAS removal processes have been evaluated in DWTPs, and activated carbon (AC) is one method that has been proven to work for PFAS removal (Kang et al., 2025). PFAS tend to adsorb onto the hydrophobic surface of AC, because of their hydrophobic nature, making AC effective for their removal in DWTP. The longer carbon chain the better removal, meaning that PFOS and PFOA with chain length of 8 carbon molecules are removed better than others. However, the newer generation that are becoming increasingly popular have very different chemical structure, have shorter chain lenght often less than 6 carbon molecules and are not very well studied.

Even though the newer generations are supposed to be less toxic they have showed signs of toxicity and based on the fact that they are becoming more and more common it is important to analyse and see the removal efficiency of them.

1.2 Aim

This project aims to investigate the performance of activated carbon for the adsorption of PFAS from drinking water. It will analyse existing literature and studies on PFAS removal in drinking water treatment plants (DWTPs), with a particular focus on the performance of different types of activated carbon. The study will compare granular activated carbon (GAC) filters with different properties, examining their adsorption capacities on legacy and replacement PFAS, affinities, and removal kinetics. Additionally, it will explore competitive adsorption between different PFAS compounds to better understand treatment challenges and performance limitations. To achieve this aim, the report seeks to answer the following research questions:

1. What types of PFAS compounds are present in raw and drinking water and pose health risks?

2. What are the efficiency and mechanisms of PFAS removal in drinking water treatment plants?

3. How effective are different types of GAC filters specifically, fresh, regenerated and aged GAC in removing PFAS, and how do they compare in terms of adsorption capacity, affinity and removal kinetics?

4. What are the differences in adsorption behaviour and removal efficiency between legacy and replacement PFAS compounds, and how are these affected by competitive adsorption processes?

2

Literature review

In the following section a litterature review will be presented that will discuss the different properties of per- and polyfluoroalkyl substances (PFAS), the occurence of PFAS in raw and finished drinking water, it will also present the regulations regarding PFAS in drinking water and lastly it will present earlier work on the removal of PFAS in DWTP.

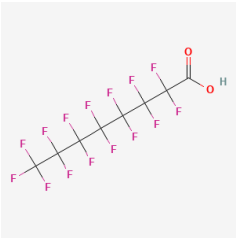
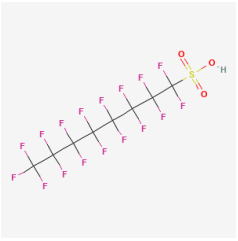
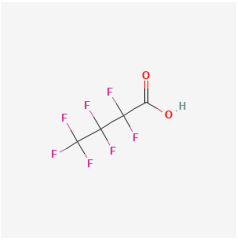
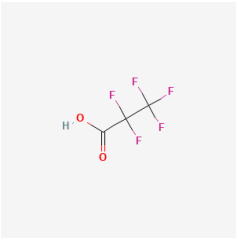
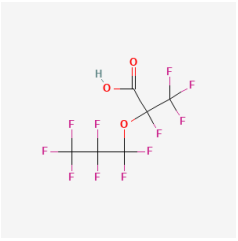
2.1 Physicochemical Properties and Classification of PFAS

PFAS is a class of commercial chemicals widely used due to their unique physicochemical properties (Gaines, 2023). They typically occur as anionic, water-soluble, and non-volatile compounds, characteristics that contribute to their persistence in the environment (Ateia et al., 2019; Buck et al., 2011). Structurally, PFAS consist of a hydrophobic, fluorinated carbon chain (tail) and a hydrophilic functional head group. The nature of the functional group largely determines the chemical behaviour and classification of PFAS.

The two most common PFAS classes are perfluoroalkyl sulfonic acids (PFSAs), which contain a sulfonate functional group (e.g., perfluorooctane sulfonate, PFOS), and perfluoroalkyl carboxylic acids (PFCAs), which contain a carboxylate functional group (e.g., perfluorooctanoic acid, PFOA), as shown in Table 2.1. The carbon–fluorine bond is among the strongest in organic chemistry, and its strength increases with the degree of fluorination. This exceptional bond stability explains the high thermal and chemical resistance of PFAS and their pronounced resistance to environmental degradation (Rahman et al., 2014).

As a result of these properties, legacy PFAS are more prone to persistence and bioaccumulation. In contrast, short-chain PFAS and PFAS alternatives generally exhibit reduced persistence, bioaccumulation potential, and toxicity. However, short-chain PFAS (C4-C6) are more mobile and have lower molecular weights, which increases their transport in aquatic environments and makes them more challenging to remove from water treatment systems (Hamid et al., 2024).

Table 2.1: Representative PFAS compounds and their chemical structures. All chemical data were obtained from PubChem (n.d.).

Chemical	Chain length	Hydrophobicity Log K_{ow}	Molecular weight [g/mol]	Chemical structure
Perfluorooctanoic acid (PFOA)	8	4.9	414.07	
Perfluorooctanesulfonic-acid (PFOS)	8	5	500.13	
Perfluorobutanoic acid (PFBA)	4	2.2	214.04	
Perfluoropropionic acid (PFPrA)	3	1.6	164.03	
Trifluoroacetic acid (TFA)	2	0.9	114.02	
Hexafluoropropylene oxide-dimer acid (HFPO-DA)	6	3.6	330.05	

2.2 Occurrence of PFAS in Raw Water

PFAS compunds are released into nature from human activity mainly from point sources such as industry and waste water treatment plants. They are commonly found in raw water sources used for drinking water production and have been detected at varying levels all across the world (Alameddine et al., 2025; Appleman et al., 2014; Belkouteb et al., 2020; Nguyen et al., 2017).

Legacy PFAS have been detected in drinking water sources globally, typically in concentrations in the range between nanogram per liter (ng/L) and microgram per liter (µg/L), depending on the land use and the level of contamination in the area surrounding the water source. (Gebbink et al., 2017; Rahman et al., 2014). Several monitoring studies have confirmed that the occurrence of PFAS varies significantly by region and land use, where urban and industrial areas generally exhibit higher concentrations due to wastewater discharge and industrial emissions. For example, Nguyen et al. (2017) found that surface waters in urban areas were dominated by legacy PFAS.

High concentrations of PFAS in raw water are of particular concern because conventional drinking water treatment processes are often insufficient to remove them. For instance, Kang et al. (2025) reported that 77.8% of treated water samples taken from 15 full-scale DWTPs along the Nakdong River in South Korea exceeded the U.S. EPA drinking water limit of 4 ng/L for PFOA, demonstrating the challenge of effectively removing PFAS in treatment plants. In a study by Boone et al. (2019) it was found that in 20 out of 25 studied DWTPs across the United States PFAS did not have a significant removal efficiency, meaning that PFAS occurred in finished drinking water in similar concentrations as in influent water. Chronic exposure to PFAS through drinking water even at low concentrations is associated with health risks, leading to increasingly stricter regulatory limits (World Health Organization, 2022).

Increasing attention has recently been given to ultrashort (C2-C3), short (C4-C6) and PFAS alternatives, which are used as replacements for longer-chain compounds. Although these substances have lower bioaccumulation potential, they have been detected at high concentrations in aquatic environments and drinking water sources. Gebbink et al. (2017) reported elevated levels of HFPO-DA in river water in the Netherlands due to industrial discharge, it was also detected in finished drinking water at concentrations up to 11 ng/L.

PFAS precursors are also of intrest as they can break down into ultrashort PFAS such as trifluoroacetic acid (TFA) (Ateia et al., 2019). TFA is one of the most commonly detected ultrashort PFAS with reported concentrations of over $1\mu g/L$, and recent monitoring in the Netherlands showed that ultrashort PFAS were more dominant than any other PFAS group in both raw and treated drinking water (Sadia et al., 2023). In Sweden, TFA has been detected in Lake Mälaren at approximately 650 ng/L, indicating widespread environmental occurrence even in regions without known point sources (Pesticide Action Network Europe, 2024).

2.3 Regulations

Regulations governing PFAS in drinking water have undergone several substantial revisions over the past decades, reflecting the rapidly evolving understanding of their toxicity and persistence. This section reviews current and proposed PFAS regulations in drinking water in the European Union (EU), Sweden, and the United States.

Under the EU Drinking Water Directive 2020/2184, two alternative regulatory approaches for PFAS have been established. The first sets a parametric value of 500 ng/L for total PFAS, while the second sets a stricter limit of 100 ng/L for the sum of 20 specifically identified PFAS. These selected PFASs are often refered to as PFAS20 and are compounds considered of particular concern for human health. EU Member States may choose whether to implement the limit for total PFAS or the limit for PFAS20 (European Parliament and Council of the European Union, 2020).

In Sweden, the EU limit for the sum of 20 PFAS has been adopted, with the addition of one further compound, namely 6:2 fluorotelomer sulfonate (6:2 FTS), resulting in a regulatory limit of 100 ng/L for PFAS21. In addition, Sweden has implemented a substantially lower limit for PFAS4, defined as the sum of perfluorooctanoic acid (PFOA), perfluorooctane sulfonate (PFOS), perfluorohexane sulfonate (PFHxS), and perfluorononanoic acid (PFNA), with a threshold value of 4 ng/L (Livsmedelsverket, 2022). This limit is based on the European Food Safety Authority's (EFSA) 2020 assessment of a tolerable intake for PFAS4 (Shrenk et al., 2020), an evaluation that was not incorporated into the EU Drinking Water Directive. The regulatory values for PFAS4 and PFAS21, 4 ng/L and 100 ng/L respectively, also serve as guideline values for drinking water from small-scale and private water supplies, including individual wells, in accordance with guidance from the Swedish National Food Agency (Livsmedelsverket, 2022).

Table 2.2: Compounds included in PFAS4 and PFAS21 (Livsmedelsverket, 2022).

Compound	Abbreviation	PFAS4	PFAS21
Perfluorohexane sulfonic acid	PFHxS	X	X
Perfluorooctanoic acid	PFOA	X	X
Perfluorooctane sulfonic acid	PFOS	X	X
Perfluorononanoic acid	PFNA	X	X
Perfluorobutane sulfonic acid	PFBS		X
Perfluoropentane sulfonic acid	PFPeS		X
Perfluorohexanoic acid	PFHxA		X
Perfluoroheptanoic acid	PFHpA		X
Perfluorodecanoic acid	PFDA		X
Perfluoroundecanoic acid	PFUnDA		X
Perfluorododecanoic acid	PFDoDA		X
Perfluorotridecanoic acid	PFTrDA		X
Perfluorotetradecanoic acid	PFTeDA		X
Perfluorobutanoic acid	PFBA		X
Perfluoropentanoic acid	PFPeA		X
Perfluoroheptane sulfonic acid	PFHpS		X
Perfluorodecane sulfonic acid	PFDS		X
Perfluorododecane sulfonic acid	PFDoDS		X
6:2 Fluorotelomer sulfonic acid	6:2 FTS		X
Perfluorooctane sulfonamide	PFOSA		X
N-methyl perfluorooctane-sulfonamidoacetic acid	MeFOSAA		X
N-ethyl perfluorooctane-sulfonamidoacetic acid	EtFOSAA		X

Both Sweden and the EU are currently considering a further regulatory proposal based on a toxicity-equivalent approach. The reasoning for this is that the toxicity of different PFAS compounds differs substantially. This proposal introduces the concept of PFOA equivalents and assigns relative potency factors (RPFs) to individual PFAS based on their toxicological profiles. Under this framework, the sum of 24 PFAS compounds, adjusted by their respective RPFs, would be limited to 4.4 ng PFOA equivalents/L. Most of the legacy PFAS compunds are more toxic, and can for example have RPF of 2 for PFOS and 10 for PFNA, while many of the emerging short-chain PFAS and PFAS alternatives have lower RPFs. This means that the emerging compounds can occur at higher concentrations before reaching the same toxicological threshold (European Parliament and Council of the European Union, 2020; Sveriges geologiska undersökning, 2023). However, as legacy PFAS are phased out, increasing production and use of short-chain PFAS and PFAS alternatives can lead to higher environmental concentrations, raising future concerns regarding long-term exposure (Hamid et al., 2024). Although this proposal is not yet implemented, this approach represents the most recent regulatory development and may be adopted in the near future.

In the United States, the Environmental Protection Agency (EPA) finalised new drinking water regulations for PFAS in 2024. These regulations establish maximum contaminant levels (MCLs) of 4 ng/L for both PFOS and PFOA. Additionally, individual MCLs of 10 ng/L have been set for PFNA, PFHxS and HFPO-DA. The regulation also introduces a Hazard Index (HI) MCL to address the combined health effects of mixtures containing two or more of four toxicologically similar PFAS: PFNA, PFHxS, HFPO-DA, and perfluorobutane sulfonate (PFBS). The HI is calculated by summing the ratios of each measured PFAS concentration to its respective Health-Based Water Concentration (HBWC), set at 10 ng/L for PFNA, PFHxS, and HFPO-DA, and 2,000 ng/L for PFBS. An HI greater than 1 indicates an exceedance of the health-protective level and triggers regulatory action, while an HI of 1 or below is considered safe. Full enforcement of the new MCLs will take effect in 2029 (U.S. Environmental Protection Agency, 2024).

Prior to the 2024 rule, the U.S. EPA had established a health advisory level of 70 ng/L for PFOS, PFOA, and their combined concentration (U.S. Environmental Protection Agency (EPA), 2021). The transition from this advisory level to the current MCLs represents a more than ten-fold reduction in permissible concentrations, illustrating the increasing recognition of PFAS toxicity at very low exposure levels.

Older studies on PFAS removal in DWTPs often reached conclusions that no longer align with today's regulatory and risk-assessment frameworks. For example, in the study earlier mentioned in section 2.2 Boone et al. (2019) identified PFAS in finished drinking water but concluded that the detected levels were not problematic based on the standards in place at the time, which have since then changed drastically. When evaluated against current standards, most waters from the 25 analysed DWTPs would exceed today's regulatory limit, underscoring the importance of reanalysing PFAS removal performance under stricter contemporary guidelines. Concentrations that were previously considered acceptable would today be interpreted as indicators of high PFAS contamination.

2.4 DWTP performance in PFAS removal

Various treatment processes have been tested for their ability to remove PFAS from drinking water. Advanced membrane technologies such as nanofiltration (NF) and reverse osmosis (RO) have shown high removal efficiencies, often exceeding 95% (Ma et al., 2024). RO is the treatment process with highest removal with >99% removal even for short-chain PFAS. However this is also the most costly method and that is why NF or other methods could be better when taking cost into account (Appleman et al., 2014).

Conventional treatment processes commonly used in DWTPs including coagulation, flocculation and physical separation processes such as sedimentation, microfilteration (MF), ultrafilteration (UF) or dissolved air floatation (DAF) can remove PFAS to a certain extent (Appleman et al., 2014). The coagulation and flocculation rely on electrostatic and hydrophobic interactions between the PFAS molecules and

either the formed precipitate or the organic matter in the water. However, their efficiency is strongly influenced by water composition. The presence of low molecular weight organic matter can inhibit PFAS removal, whereas high molecular weight organic compounds, such as humic acid, can enhance it. See figure 2.1 for coagulation mechanisms and the impact of background water matrix for removal of PFAS. As a result, conventional coagulation-flocculation processes are unreliable for consistent PFAS removal, and achieving higher removal efficiencies is challenging (Wang et al., 2025).

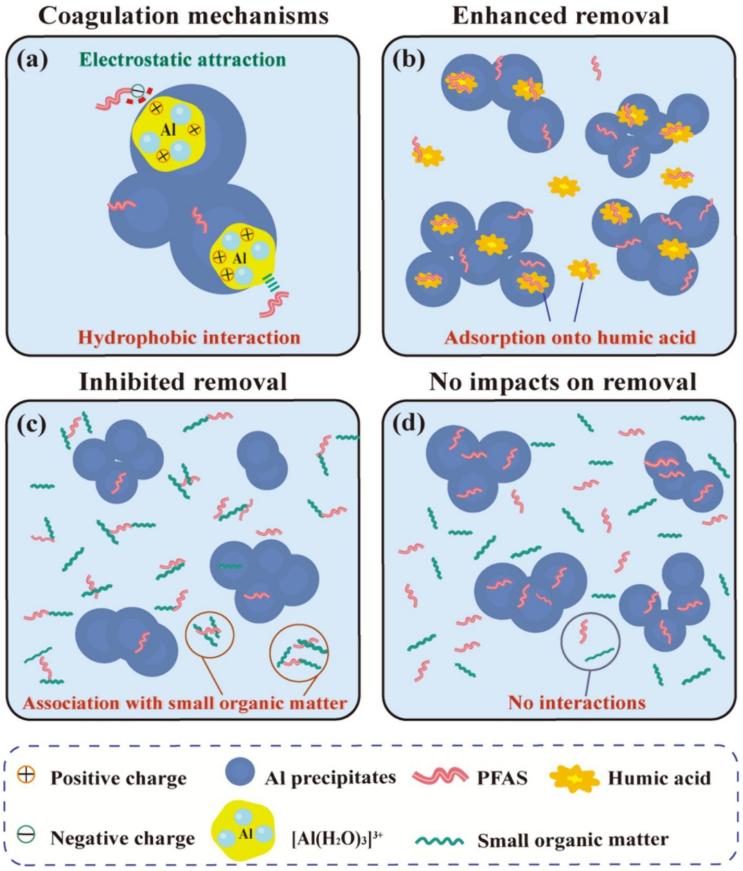


Figure 2.1: Coagulation mechanisms for removal of PFAS (a); impacts of background matrices on removal of PFAS during coagulation (b-d). Used with permission of the author (Wang et al., 2025).

Following conventional treatment, sedimentation combined with MF or UF has shown no substantial effect on PFAS removal. DAF, however, has demonstrated some degree of success (Appleman et al., 2014). Similarly, foam fractionation (FF) exploits the surfactant properties of PFAS by using air bubbles to drive contaminants toward the surface. Unlike DAF, FF employs considerably larger air bubbles, which allows PFAS molecules to embed within the bubble while keeping their polar headgroup oriented toward the surrounding water. FF has been applied primarily in industrial and wastewater contexts, where it has shown promising results for long-chain PFAS removal, particularly at elevated concentrations. Its efficacy for short-chain PFAS is notably poorer, and since the method has seen limited imple-

mentation at drinking water treatment plants (DWTPs), its performance in that setting remains difficult to assess (Smith et al., 2023).

Other types of treatment processes that are oxidation-based, such as chlorination and ozonation, are generally ineffective for PFAS removal, due to the strong carbon–fluorine bonds that resist breakdown (Kang et al., 2025). In addition to this aeration and ultraviolet (UV) disinfection also have similar limitation in PFAS removal. In some cases they have lead to an increase in PFAS or a breakdown of PFAS precursors into other types of PFAS (Appleman et al., 2014).

One method that have been studied for removal of PFAS is adsorption and the two main processes used to achieve adsorption is anion exhange (AE) and activated carbon (AC). They operate in different ways but have both managed to achieve good removal efficiency for PFAS. AE is a liquid-liquid separation process that works by exchanging negatively charged anions such as PFAS with anions such as hydroxide or chloride on a positively charged resin (Riegel et al., 2023). AC work by adsorption onto the surface through different mechanisms. Overall removal efficiency can be high, although it varies depending on water composition and the characteristics of the activated carbon used (Belkouteb et al., 2020; Kang et al., 2025).

Multiple studies have reported that AE in some cases preforms better than AC when it comes to removal capacity and loading, which is gram adsorbed PFAS per gram filter media (McCleaf et al., 2017; Riegel et al., 2023). However, the cost of AE is high and the method is not implemented in the same extent as AC (Riegel et al., 2023). Therefore activated carbon methods are well suited to be analysed for PFAS removal in DWTP.

2.5 Activated carbon in removal of PFAS

Activated Carbon (AC) filters are typically produced via carbonization and subsequent activation of carbon rich precursor materials, which may originate from botanical sources, coalified plant matter, or agricultural by-products. The use of botanical sources such as coconut shells and waste derived precursors has gained increasing interest as a sustainable and cost effective alternative to conventional activated carbons (Oubagaranadin & Murthy, 2011). Recent studies on PFAS removal indicate that the performance of AC in drinking water treatment can be high irrespective of their precursor origin. For example, Holliday (2020) demonstrated that removal efficiencies exceeding 95% can be achieved using both coconut shell–based granular activated carbon (GAC) and bituminous coal–based GAC. These findings suggest that parameters related to the physicochemical properties of the ACs and the characteristics of the PFAS compounds play a more critical role in removal performance than the origin of the activated carbon itself (Cantoni et al., 2021).

The adsorption mechanisms that act when AC is used are electrostatic interactions, hydrophobic interactions, surface complexation and hydrogen bonding. Out of these mechanisms the predominant one is hydrophobic interactions. Some characteristics

of the AC that can affect the performance is the porosity, the particle size, the origin and the surface charge (Tan et al., 2023). In a study by Cantoni et al. (2021) the surface charge was identified as the main component of the AC that affected the performance of it.

Other factors influencing removal efficiency of AC are related to the characteristics of the compounds being targeted. When PFAS are the target compounds various studies have identified the chain length of the PFAS as the most important factor, where longer PFAS compunds with chain-length > 6 are removed significantly better (Alameddine et al., 2025; Belkouteb et al., 2020; Cantoni et al., 2021; Eschauzier et al., 2012; Kang et al., 2025; McCleaf et al., 2017; Tan et al., 2023). The functional group of the PFAS compound also influences removal efficiency. Perfluoroalkane sulfonic acids (PFSAs), which carry a sulphonate functional group, are generally removed more effectivley than perfluoroalkyl carboxylic acids (PFCAs), which carry a carboxylate group. This difference arises from the degree of fluorination, where in PFSAs all carbon atoms are fully fluorinated, whereas in PFCAs the carbon of the carboxylate group remains unfluorinated, reducing the overall hydrophobicity of the molecule. As illustrated in Table 2.1, PFOS and PFOA represent a comparable pair of compounds with identical chain length but differing functional groups, and the sulphonate-bearing PFOS is generally removed to a greater extent than PFOA (Belkouteb et al., 2020; McCleaf et al., 2017; Ochoa-Herrera & Sierra-Alvarez, 2008; Tan et al., 2023). PFAS comes in both linear and branched isomers and when comparing the removal of these two isomers it is found that the linear PFAS are better removed in DWTP using AC (Belkouteb et al., 2020; McCleaf et al., 2017). When taking these factors into account longer PFAS chains and a higher degree of fluorination increase the hydrophobicity. This strengthens hydrophobic interactions and, as a result, leads to higher removal efficiencies.

Activated carbon can be incorporated into DWTPs in several configurations, the most common of which are powdered activated carbon (PAC) or granular activated carbon (GAC). While both forms operate through the same adsorption mechanisms and are governed by the same compound- and material-related factors described above, they differ in how they are applied within the treatment train. PAC is typically dosed directly into the water prior to settling and granular media filtration (GMF), whereas GAC is implemented as a fixed-bed filter positioned after the GMF process (Alameddine et al., 2025). Beyond the factors previously discussed, each configuration is subject to additional process-specific parameters. For PAC, the applied dose is a key determinant of performance, with higher doses yielding greater removal efficiencies (Ahrens et al., 2025). For GAC, the empty bed contact time (EBCT) which is the the theoretical residence time of water within the carbon bed assuming an empty reactor volume, plays a similar role, as longer contact times allow for more complete adsorption and thus improved removal (Belkouteb et al., 2020).

PAC is a single-use material that is disposed of after application, whereas GAC can be regenerated once its adsorption capacity is exhausted and then reused multiple times, thereby reducing both operational costs and environmental impacts (Alamed-

dine et al., 2025). Regeneration is most commonly carried out through thermal treatment up to 900 ° C, which has been shown to achieve greater than 99.9% PFAS destruction during the GAC reactivation process (Mayerberger et al., 2025). However, when exhausted, or if GAC is not properly regenerated it may begin to leach previously adsorbed contaminants back into the water, posing risks to treated water quality (Appleman et al., 2014).

Regeneration have been shown to not impair the removal of long-chain PFAS and in some cases it even enhances performance due to the increased mesoporosity of reactivated carbon compared with the more microporous fresh carbon. Greater mesoporosity reduces pore blocking and strengthens hydrophobic interactions with long-chain PFAS. In contrast, short-chain PFAS removal depends mainly on microporous surface area, but competition with dissolved organic matter (DOM) often limits their adsorption, making them more difficult to remove (Cantoni et al., 2021).

Alameddine et al. (2025) demonstrated that when PAC and GAC are evaluated under comparable conditions and with matrix effects taken into account, the two adsorbents exhibit largely similar PFAS removal efficiencies. PAC may be somewhat easier and more cost-effective to implement in situations where immediate treatment is required. However, GAC offers slightly higher performance and generates less residual waste, as it can be reactivated and reused, thereby eliminating the PFAS captured during treatment (Alameddine et al., 2025).

2.6 Summary and Discussion of Literature

This literature review highlights that PFAS contamination in drinking water remains a significant and evolving challenge for drinking water treatment plants, particularly in light of increasingly stringent regulatory limits. The literature demonstrates that both legacy and emerging PFAS alternatives are widespread in raw water sources, often at concentrations that exceed current or proposed guideline values. While long-chain PFAS have been the primary regulatory focus, short-chain PFAS and PFAS alternatives are becoming more prevalent. This due to substitution, their higher mobility and resistance to conventional treatment processes which raise concerns for long-term water safety. These findings underscore that historical assessments of treatment performance are no longer sufficient under modern regulatory and toxicological frameworks.

Activated carbon was identified as one of the most practical and widely applied methods for PFAS removal in DWTPs. The reviewed studies show that PFAS adsorption onto activated carbon is governed primarily by hydrophobic interactions and is strongly influenced by PFAS chain length, functional group, and molecular structure, as well as by carbon properties such as surface charge and porosity. While PAC and GAC generally show comparable removal efficiencies when evaluated under similar conditions, GAC offers advantages in long-term operation due to its reusability and lower waste generation. However, both PAC and GAC perform better for long-chain PFAS than for short-chain and PFAS alternatives, which are

more susceptible to competition from dissolved organic matter and are less strongly adsorbed.

Overall, this review indicates that although activated carbon remains an effective and essential treatment option for PFAS, it is not a complete solution, particularly for short-chain PFAS and PFAS alternatives. Competitive adsorption effects and the limited removal of short-chain PFAS pose ongoing challenges for compliance with future regulatory limits. These findings emphasize the need for careful selection and management of activated carbon systems, continuous performance monitoring, and further research into treatment strategies that can address the increasing dominance of replacement PFAS compounds in drinking water sources.

3

Methods

This section describes the laboratory methods used in this study. The experiments were conducted to address the third and fourth research questions in chapter 1.2, which focuses on the performance of GAC in removing PFAS, as well as the competitive adsorption behaviour among different PFAS compounds.

3.1 Chemicals and Reagents

The PFAS compounds selected for the experiments were chosen based on the literature review, which indicated that short-chain PFAS are generally removed less efficiently than long-chain compounds. The literature also indicated that legacy PFAS compounds are more toxic and therefore of greater concern for removal. Therefore it was interesting to analyse a wide range of compounds to get a good comparison of their behaviour. Accordingly, the following compounds were investigated in the laboration: perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), perfluorobutanoic acid (PFBA), perfluoropropionic acid (PFPrA), and trifluoroacetic acid (TFA). The molecular properties of the selected compounds can be found in table 2.1.

These compounds represent a broad range of chain lengths, including long-chain, short-chain, and ultra-short-chain PFAS. This selection also allows for comparison between different functional groups, enabling assessment of their influence on adsorption behaviour. Stock solutions were prepared at a concentration of 1000 mg/L for each PFAS in methanol, it was neutralized to achieve a pH of 7 and was subsequently used in the experiments.

3.2 Activated Carbon Preparation

Three types of GAC were obtained from one drinking water treatment plant in Gothenburg, Sweden: (i) an aged GAC that had been in operation for approximately five years (A-GAC), (ii) a thermally reactivated GAC (R-GAC) that was regenerated at 800 °C and (iii) a fresh, unused GAC (F-GAC). All GAC samples were of the Filtrasorb TL830 type, which is an agglomerated coal-based granular activated carbon.

The A-GAC and R-GAC samples were dried prior to use to ensure accurate mass measurements and to avoid accounting for residual water content. The F-GAC was initially rinsed with ultra pure water to remove fine particulate material generated during storage and handling. Because the dry GAC particles can abrade against each other, a fraction of the material may become pulverized, forming a thin layer of carbon fines. Rinsing the GAC with ultra pure water removed these fines, after which the material was dried to obtain clean GAC particles free from powdered residues. Subsequently, all GAC samples were soaked in ultra pure water for 24 h before the experiments to achieve full water saturation and to better represent normal operational conditions.

3.3 Batch Adsorption Experiments

Batch adsorption experiments were conducted in 50 mL polypropylene centrifuge tubes. Each tube was filled with 25 mL of ultrapure water, to which 0.25 g of GAC was added, resulting in an adsorbent concentration of 10 g/L. After that, the PFAS stock solution was added.

3.3.1 Adsorption Kinetics

After the soaking period, 2.5 µL of PFAS stock solution was added to each tube, resulting in an initial PFAS concentration of 100 µg/L. Although this concentration exceeds levels typically observed in environmental waters, it was selected to enhance detectable differences in adsorption behaviour among PFAS compounds and GAC types. All experiments were performed in duplicate to ensure reproducibility. For each GAC type, procedural blanks containing GAC but no PFAS were prepared, along with control samples containing PFAS but no activated carbon.

Following PFAS addition, the tubes were placed on an orbital shaker and agitated at 30 rpm at a room temperature of approximately 20 °C. Samples (10 mL) were collected at seven time points: 0 min, 5 min, 15 min, 45 min, 120 min, 240 min, and 24 h. Early sampling points were selected to capture rapid adsorption kinetics, as most adsorption is expected to occur within the first 15 min based on previous studies (McCleaf et al., 2017). Later time points were included to assess equilibrium conditions and determine the maximum adsorption capacity of the activated carbon.

A sacrificial sampling approach was employed, whereby the entire batch reactor was sacrificed at each sampling time point. Consequently, one tube was prepared for each time point, resulting in seven tubes per GAC type. This approach, previously applied in PFAS batch adsorption studies using GAC (Pranić et al., 2025), avoids changes in solute concentration and solid-to-liquid ratios that may occur during repeated sampling from the same reactor. Compared with conventional batch tests which require larger reactor volumes to maintain negligible sampling effects (<5% of the total volume), this method substantially reduced both material and water consumption, requiring a total volume of only 175 mL (7 × 25 mL) for each GAC.

The removal efficiency (RE [%]) for each PFAS and GAC was calculated according to equation 3.1.

$$RE = \frac{C_0 - C_t}{C_0} \times 100 \tag{3.1}$$

where C_0 and C_t $[\mu g/L]$ are the PFAS concentrations at initial time and time t [min].

To analyse the kinetics the nonlinear pseudo-first-order (PFO) (Lagergren, 1898) and pseudo-second-order (PSO) (Ho & McKay, 1999) models were used based on equations 3.2 and 3.3. These models are used to describe adsorption processes and assess the rate at which contaminants are removed from aqueous systems. The PFO model assumes that adsorption rate is proportional to the number of unoccupied adsorption sites and is often associated with diffusion-controlled or physical adsorption processes. In contrast, the PSO model assumes adsorption is dependent on the square of available adsorption sites and is frequently used to describe systems where stronger adsorbate–adsorbent interactions may occur. Comparing the fit of PFO and PSO models to experimental data enables assessment of adsorption behavior and provides insight into the mechanisms governing PFAS uptake onto GAC.

$$PFO : q_t = q_e(1 - e^{-k_1 t}) \tag{3.2}$$

$$PSO : q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \tag{3.3}$$

where q_t and q_e $[\mu g/g]$ are the adsorption capacity on adsorbent at time t [min] and at equilibrium, k_1 $[h^{-1}]$ and k_2 $[g \cdot \mu g^{-1} \cdot h^{-1}]$ are the coefficients for adsorption mass transfer for each of the models PFO and PSO.

3.3.2 Adsorption Isotherms

Adsorption isotherm experiments were conducted to evaluate the equilibrium adsorption capacity of the three GAC materials. PFAS stock solution was added to the tubes to obtain initial PFAS concentrations of 0.1, 1, 10, 100, and 500 µg/L. The tubes were agitated on an orbital shaker at 30 rpm for 24 h at room temperature (̃20 °C). These experiments were conducted for all three GAC types and were performed in duplicate. The adsorbent loading where analysed through the Langmuir (Langmuir, 1918) and Freundlich (Freundlich, 1906) isothermal models using the following the equations 3.4 and 3.5.

$$Langmuir : q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \tag{3.4}$$

$$Freundlich : q_e = K_F C_e^{1/n} \tag{3.5}$$

where q_m $[\mu g/g]$ is the maximum adsorption capacity, C_e $[\mu g/L]$ is the concentration at equilibrium, K_L $[L/\mu g]$ and K_F $[(\mu g/g)(\mu g/L)^{-1/n}]$ are the Langmuir and Freundlich adsorption coefficients which represents constants related to adsorption affinity, lastly 1/n [-] shows the sorption intensity and is the Freundlich exponent.

3.4 Analytical Methods

The following section describes the analytical techniques employed to characterise the PFAS concentrations in water samples and the physical properties of the GAC materials. Compound quantification was performed by LC-MS, while SEM was used to examine the surface morphology and porosity of the GAC samples

3.4.1 LC-MS Analysis and Quantification

All water samples were filtered through hydrophilic polyethersulfone (PES) membrane filters (0.2 μm pore size) prior to analysis to prevent particulate contamination from the GAC or other suspended matter from interfering with the measurements. Blanks were processed in the same way.

The concentration of selected PFAS compounds in the adsorption experiments was determined by Liquid Chromatography–Mass Spectrometry (LC-MS), using a Xevo TQ-S micro Triple Quadrupole Mass Spectrometer (Waters Corporation). LC-MS operates by first separating compounds chromatographically based on their physicochemical properties as they interact with a stationary phase column, before ionising the eluting analytes and detecting them according to their mass-to-charge ratio (m/z). The triple quadrupole configuration enables highly selective quantification by isolating precursor ions, inducing fragmentation, and monitoring specific product ions. The analysis was performed using Multiple Reaction Monitoring (MRM), a targeted mass spectrometry acquisition mode in which predefined precursor-to-product ion transitions are monitored for each analyte. This approach minimises matrix interference and allows for accurate and sensitive detection of PFAS compounds at trace concentrations.

For quantification, seven calibration standards were prepared by spiking ultra pure water with the PFAS stock solution to yield the following concentrations: 250, 125, 50, 25, 12.5, 5.0, and 2.5 μg/L. Each standard was analysed by LC-MS, producing chromatograms in which each of the five target PFAS compounds was represented by a single characteristic peak (Appendix A). Compound specific calibration curves were subsequently constructed by plotting peak intensity against known concentration, and sample concentrations were then interpolated from these curves based on their respective measured intensities (Appendix B).

3.4.2 GAC Surface Characterisation by SEM

The surface morphology and porosity of the three GAC samples were examined using Scanning Electron Microscopy (SEM). A Quanta 200 ESEM FEG (FEI), equipped with a Schottky field emission gun for high spatial resolution, was operated in high vacuum mode at an accelerating voltage of 10 kV. Imaging was performed in both secondary electron (SE) mode, to assess surface topography, and backscattered electron (BSE) mode, to provide compositional contrast for identifying surface depositions and biofilm on the fresh, aged, and regenerated GAC samples.

4

Results and discussion

This chapter presents the results of the PFAS removal experiments conducted on fresh, regenerated, and aged GAC filters, followed by analysis of the adsorption kinetics, isotherm behaviour, and surface morphology of the filter media. The results are discussed in relation to established adsorption theory and the implications for full-scale drinking water treatment. Each section addresses a distinct aspect of GAC performance, progressing from removal efficiency to the underlying physicochemical mechanisms.

4.1 GAC Surface Structure and Porosity

Scanning electron microscopy (SEM) imaging revealed clear morphological differences between the fresh (F-GAC), regenerated (R-GAC), and aged (A-GAC) filter media.

The F-GAC surface appeared relatively smooth with a uniform pore structure, as visible in the magnified region of the granule shown in Figure 4.1. Pore openings were small, consistent with an intact, unmodified carbon matrix.

The R-GAC exhibited a noticeably rougher surface texture with several protruding fragments visible in the backscattered electron (BSE) image in Figure 4.2. These are attributed to mineral residues or ash deposits remaining after thermal regeneration at approximately 800°C, at which temperature any residual organic material or biofilm would have been fully combusted. The secondary electron (SE) image further reveals a substantial increase in pore size relative to F-GAC, with pores reaching up to 100 μm in diameter. The magnified image in Figure 4.2 also shows secondary internal pores within the walls of larger pores, suggesting that the thermal regeneration process may expand or restructure deeper pore networks within the carbon matrix.

The A-GAC showed markedly different surface characteristics, with a visible biofilm layer apparent in the BSE image in Figure 4.3. Even at higher magnification, the external porosity of the granule appeared largely obscured by this biological coating. As the samples were dried prior to SEM analysis, the biofilm had partially contracted and cracked, which is visible in the images. At the highest magnification, the sur-

face topography reflects a composite of the underlying carbon matrix and overlying biological material, likely comprising biofilm and possibly algal matter, which accounts for the irregular and heterogeneous texture observed. This surface fouling is consistent with the reduced adsorption performance of the A-GAC reported in literature in preceding sections, as biofilm accumulation and pore occlusion would limit PFAS access to internal adsorption sites.

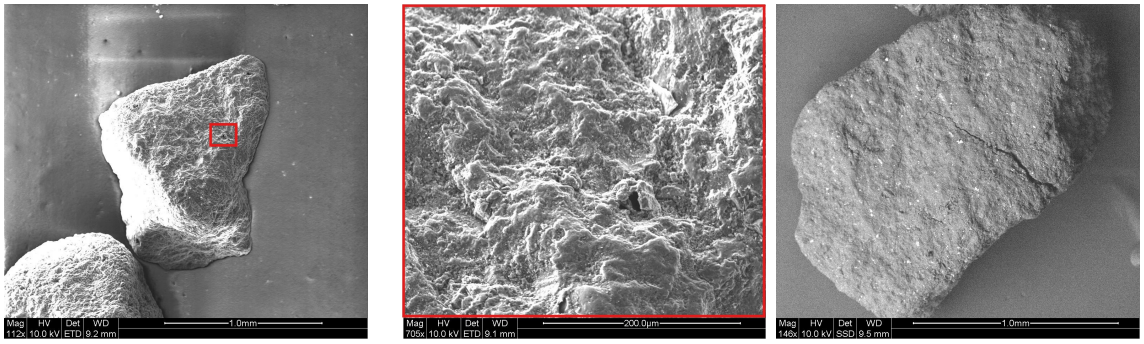


Figure 4.1: SEM image SE mode of F-GAC full particle (left), zoomed into a pore (middle) and BSE mode (right).

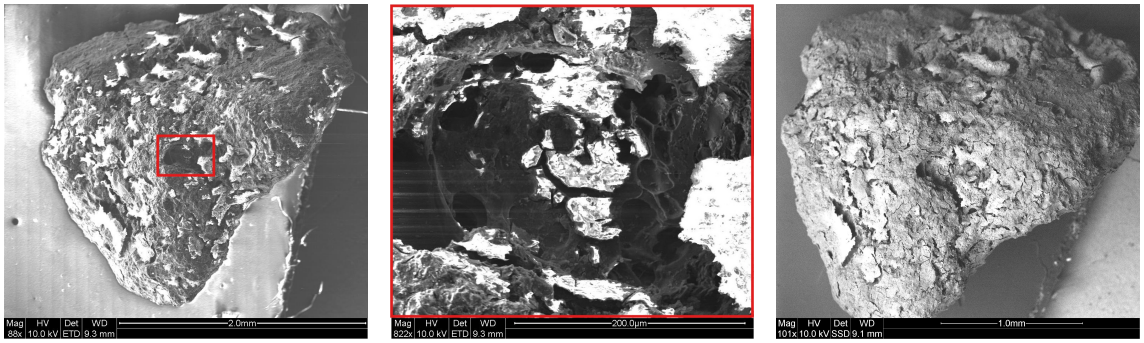


Figure 4.2: SE image of R-GAC full particle (left), zoomed into a pore (middle) and BSE image (right).

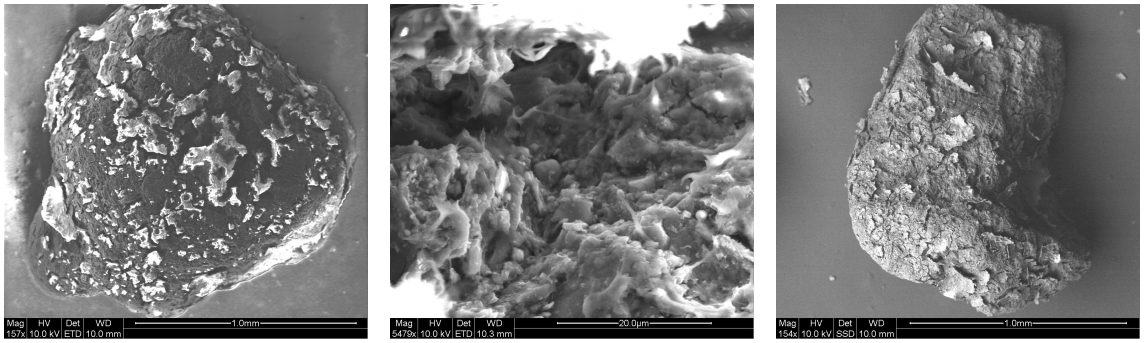


Figure 4.3: SE image of A-GAC full particle (left) and zoomed in (middle) and BSE image (right).

4.2 PFAS Removal Efficiency in GAC filters

The PFAS concentration data obtained from the kinetic batch experiments are presented in Figures 4.4, 4.5, and 4.6. The performance of the three GAC filters varied substantially, with removal efficiency strongly influenced by both filter age and PFAS chain length.

One compound that was not detected in any samples at detectable limits is PFOS (see all concentrations in appendix A). In no sample from any GAC filter, at any sampling point was PFOS detected. This indicates strange behaviour of it since PFOS was detected with correct concentrations in the standard samples where no filter media was added. In the literature PFOS is shown to have better adsorption than other PFASs, but in this case even the samples with A-GAC at the initial time (t = 0 min), no PFOS was detected. So this could indicate that PFOS is adsorbing directly or that it is transforming. However because of this PFOS have not been included in the analysis and modelling parts of the report.

The A-GAC filter demonstrated highly limited adsorption capacity, achieving meaningful removal only for the long-chain compound PFOA, with a removal efficiency (RE) of 73.8%. Notably, short-chain and ultra-short-chain compounds appeared in higher concentration in the samples than the what was added initially (see Figure 4.4). This could be because PFOS, and PFOA with higher affinity adsorbs to A-GAC and takes up sites from short chain PFAS that might have been there already which causes them to desorb from the GAC and leach into the water. When looking at the result from the blank samples with A-GAC and no PFAS non of the compounds were leaching out of the GAC, meaning that it is not the filter that is leaching but rather the fact that when more PFAS is added it might be triggering the short chained PFAS to release. This is of significant operational concern, as it suggests the aged filter may act as a secondary contamination source for shorter-chain PFAS.

The F-GAC filter, by contrast, demonstrated high removal efficiency across all analysed PFAS compounds regardless of chain length, achieving >90% removal within 15 minutes and >99% removal at 45 minutes. See figure 4.6. This confirms that adsorption capacity is high in new filter media.

The R-GAC filter showed intermediate and chain-length-dependent performance (see Figure 4.5). Removal efficiency increased with both contact time and carbon chain length across all compounds. At equilibrium, PFOA (C8) reached >99% RE, while the shorter-chain compounds showed progressively lower removal: PFBA (C4) achieved 73%, PFPrA (C3) achieved 54%, and the ultra-short-chain TFA (C2) achieved 46%. These results suggest that thermal regeneration partially restores adsorption capacity, but the recovery is insufficient for reliable removal of short- and ultra-short-chain PFAS.

Although R-GAC achieved complete removal of PFOA at equilibrium, this was reached considerably slower than for F-GAC, indicating that thermal regeneration affects adsorption kinetics even for long-chain PFAS. This contrasts with findings

by Cantoni et al. (2021), who reported that regeneration does not impair long-chain PFAS removal. However, the two studies differ in water matrix composition: the present study used ultra pure water, while Cantoni et al. (2021) used tap water. The organic matter present in tap water may have influenced adsorption behaviour, which could partly account for the observed discrepancy. Overall, these results suggest that while thermal regeneration partially restores adsorption capacity, the recovery is insufficient for reliable removal of short- and ultra-short-chain PFAS.

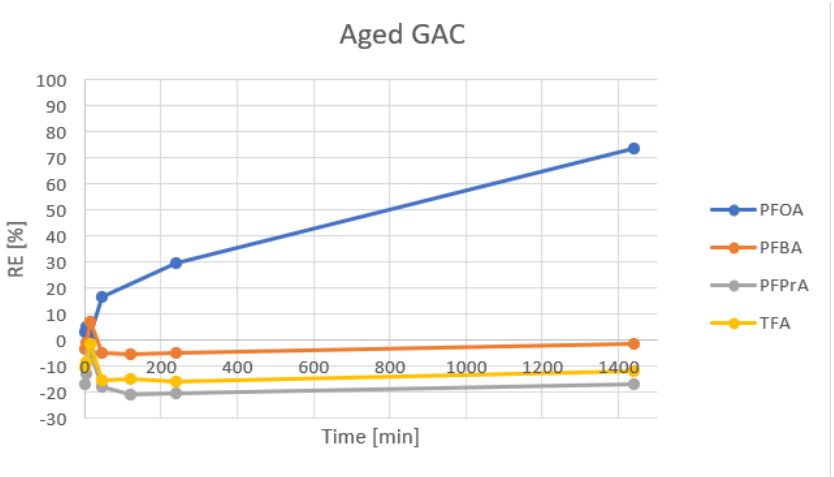


Figure 4.4: Removal efficiency (RE) for aged GAC

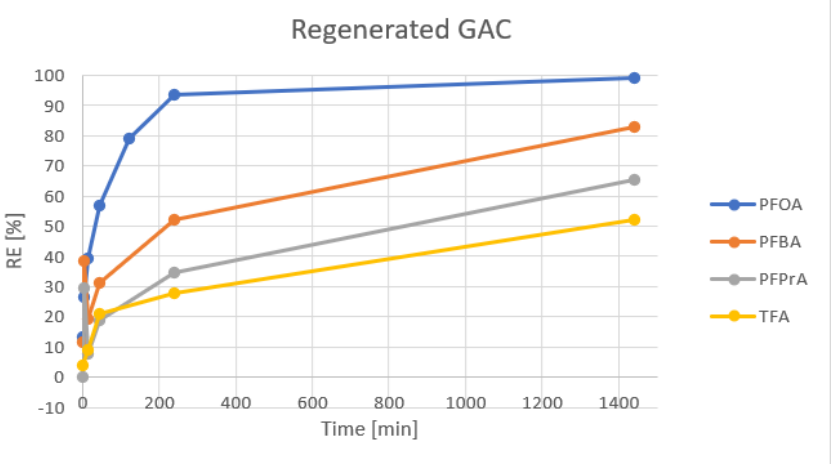


Figure 4.5: Removal efficiency (RE) for regenerated GAC

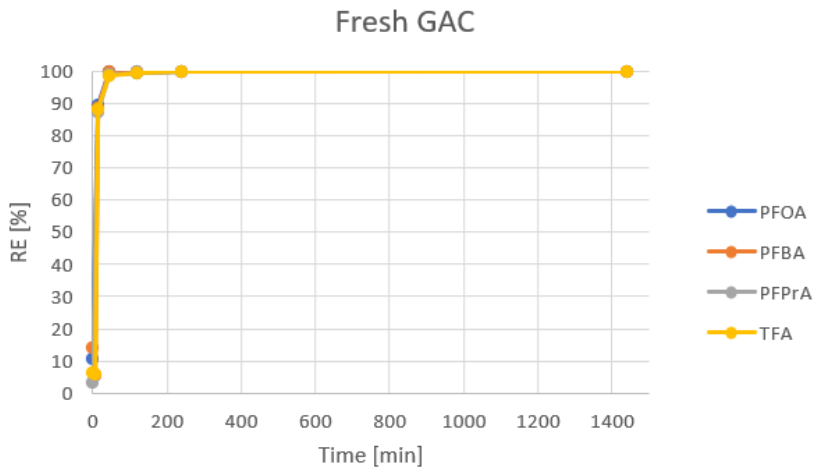


Figure 4.6: Removal efficiency (RE) for fresh GAC

4.3 Adsorption Kinetics of PFAS on GAC Filters

The adsorption kinetics varied considerably across the three filter conditions, consistent with the removal efficiency patterns described in the previous section.

For the A-GAC filter, only PFOA exhibited kinetic behaviour that could be modelled, with both the pseudo-first-order (PFO) and pseudo-second-order (PSO) models yielding comparable fits ($R^2 = 0.95$ for both). The remaining compounds showed no meaningful correlation with either model, likely reflecting the near-absence of net adsorption.

For the F-GAC filter, all PFAS compounds were better described by the pseudo-first-order (PFO) model than the pseudo-second-order (PSO) model. This indicates that adsorption kinetics were dominated by the rapid occupation of abundant, accessible surface sites, with external mass transfer playing an important role. This behaviour is consistent with an intact pore structure and unmodified surface chemistry.

The R-GAC filter exhibited the opposite trend, with the PSO model providing a better fit than PFO across all compounds. This pattern held for both long- and short-chain PFAS, though the correlation was weaker for shorter-chain compounds. PSO-dominated kinetics suggest that adsorption is increasingly governed by surface interaction kinetics and site heterogeneity, implying that rate-limiting chemisorption-like mechanisms become more prominent following regeneration.

Two mechanisms likely account for this shift. First, thermal regeneration can induce pore blockage or restructuring through partial collapse of micropores, deposition of residual organics or ash, and reduction of accessible surface area. Second, regeneration may alter surface chemistry by modifying oxygen-containing functional groups, surface charge, and hydrophobicity. Both changes are particularly consequential for PFAS adsorption, which depends on a combination of electrostatic interactions, hy-

drophobic effects, and fluorocarbon affinity for carbonaceous surfaces. As a result, many energetically favourable adsorption sites may no longer be accessible following regeneration, which would disproportionately affect the shorter-chain compounds that rely more heavily on these interactions.

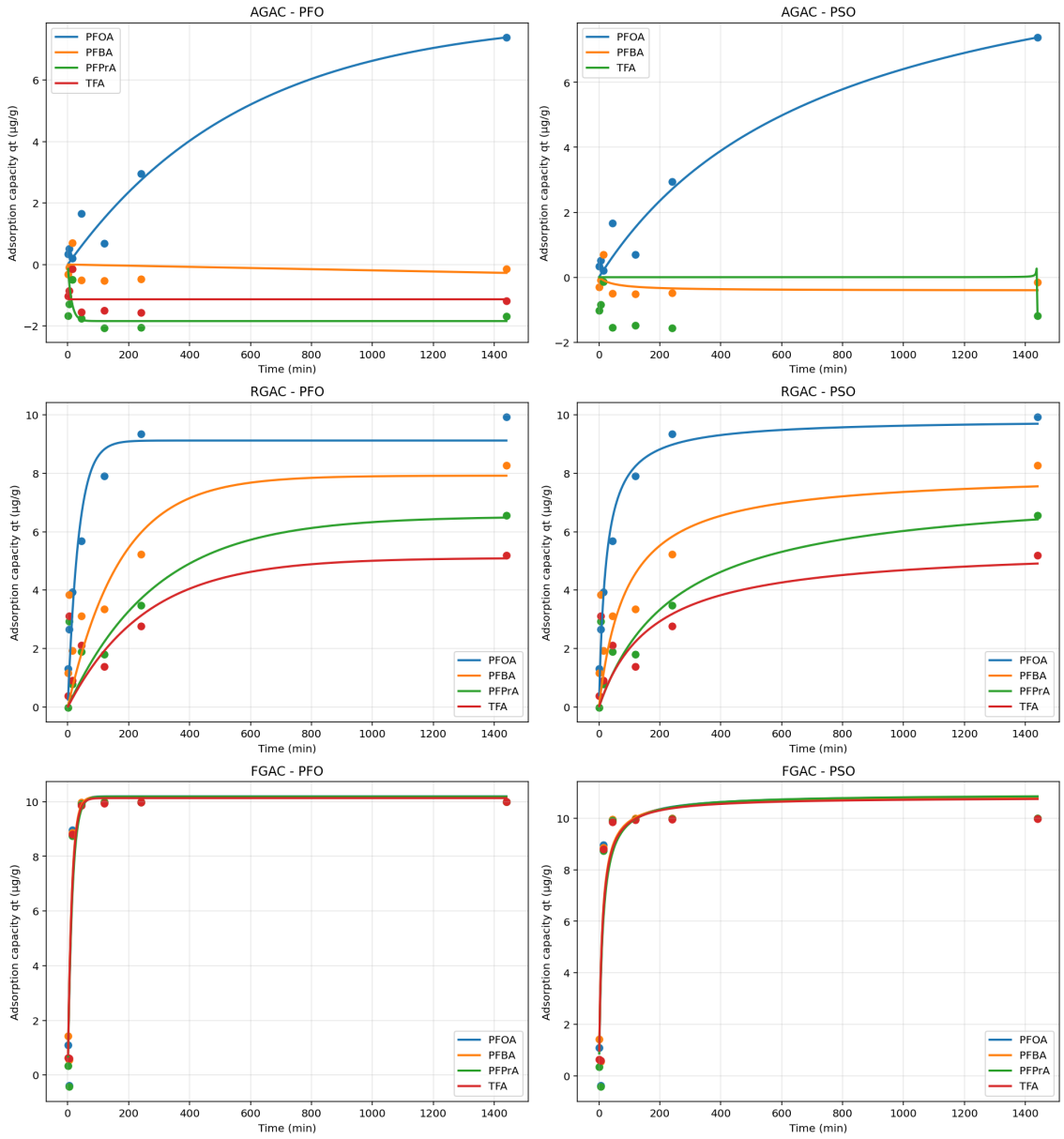


Figure 4.7: The PFO (graphs on left side) and PSO (graphs on right side) model for adsorption kinetics of PFAS onto aged, regenerated and fresh GAC filters.

4.4 Adsorption Isotherms

Table 4.1: Langmuir and Freundlich isotherm parameters for PFAS adsorption on different GAC types.

GAC Type	PFAS	$q_{\max}$	K_L	R_L^2	K_F	n	R_F^2
A-GAC	TFA	19.31	0.0035	0.975	0.0794	1.206	0.971
A-GAC	PFPrA	7.74	0.0043	0.905	0.0481	1.310	0.897
A-GAC	PFBA	9.20	0.0051	0.949	0.0745	1.367	0.944
A-GAC	PFOA	26.34	0.0133	0.991	0.8432	1.764	0.978
R-GAC	TFA	231.40	0.0007	0.975	0.1340	0.998	0.974
R-GAC	PFPrA	79.20	0.0036	0.958	0.4989	1.256	0.948
R-GAC	PFBA	78.32	0.0097	0.970	1.3656	1.384	0.955
R-GAC	PFOA	115.39	0.2510	0.859	22.0336	1.369	0.819

The fresh GAC filter is not included in the isotherm analysis, as it achieved >99% removal at equilibrium for all PFAS compounds even at the elevated test concentration of 500 μg/L. Under these conditions, meaningful isotherm modelling was not possible as the system did not produce the range of equilibrium concentrations required to parameterise either model.

When fitting the isotherm data for A-GAC and R-GAC to the Freundlich and Langmuir models, the Langmuir model provided a slightly better fit for both the aged and regenerated GAC. This suggests that both monolayer adsorption and surface heterogeneity contribute to PFAS uptake. This also indicates that the isotherm behaviour of PFAS on A-GAC and R-GAC is well described by equilibrium frameworks.

A notable exception was PFOA on R-GAC, where both models had lower correlation. This may indicate that PFOA adsorption on regenerated media does not follow one single equilibrium mechanism, potentially due to the coexistence of multiple adsorption pathways, saturation effects, nonlinear behaviour at the tested concentrations or insufficient equilibration conditions within the experimental concentration range.

4.4.1 Langmuir Parameters

The Langmuir model assumes monolayer adsorption onto uniform adsorption sites. The maximum adsorption capacity (q_{max}) reflects the theoretical upper limit of PFAS uptake per unit mass of GAC. For the aged GAC, the capacity followed the order PFOA > TFA > PFBA > PFPrA, consistent with the expectation that the longest-chain compound adsorbs most strongly due to its greater hydrophobicity, while shorter-chain compounds exhibit progressively weaker adsorption. For the regenerated GAC, q_{max} values were approximately one order of magnitude higher than for the aged GAC, indicating that thermal regeneration substantially restored or increased the number of accessible adsorption sites.

The Langmuir affinity constant (K_L) describes the strength of the adsorbate–surface

interaction. The affinity followed the order PFOA > PFBA > PFPrA > TFA across both filter conditions, confirming that chain length is a primary determinant of binding strength, where longer-chain PFAS bind more strongly to GAC surfaces, as shown in Figure 4.8. This observation may further explain the behaviour observed for the A-GAC filter. As the filter approaches its adsorption capacity and contains previously adsorbed PFAS, the introduction of PFAS compounds may induce competitive adsorption effects. Specifically, long-chain PFAS, which generally exhibit a higher affinity for activated carbon, may displace previously adsorbed short-chain PFAS from available adsorption sites. The desorption of these weaker-bound compounds could subsequently increase their concentrations in the effluent, consistent with the elevated levels of short-chain PFAS observed in the results (see Figure 4.4).

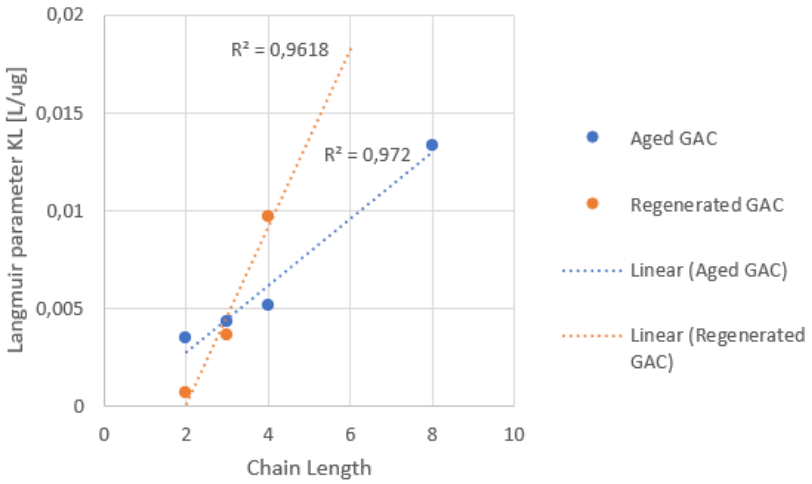


Figure 4.8: Langmuir affinity constant K_L as a function of PFAS carbon chain length.

4.4.2 Freundlich Parameters

The Freundlich model assumes a heterogeneous surface with multilayer adsorption. The Freundlich affinity parameter (K_F) followed the same chain-length-dependent trend as K_L, with PFOA exhibiting the highest value, further supporting the role of hydrophobicity and chain length in governing adsorption strength (see Figure 4.9). The heterogeneity parameter n provides a measure of adsorption favourability, with $n > 1$ indicating favourable adsorption conditions. PFOA consistently yielded the highest n value, confirming that adsorption becomes increasingly favourable with increasing chain length.

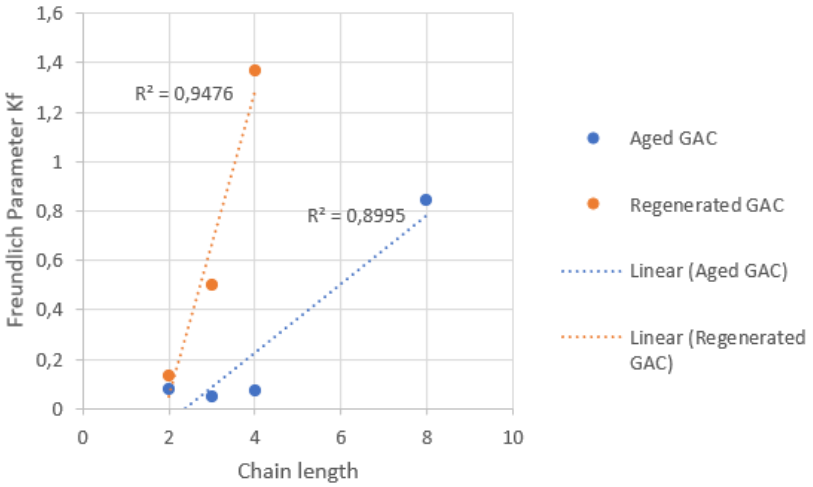


Figure 4.9: Freundlich affinity constant K_F as a function of PFAS carbon chain length.

5

Conclusion and Further Studies

This chapter presents the main conclusions drawn from the literature and experimental results and discusses potential directions for further research.

5.1 Conclusion

PFAS compounds represent a significant and widespread contamination challenge, with concentrations exceeding regulatory health limits detected in both raw and treated drinking water sources. Conventional treatment methods and advanced oxidation processes have proven insufficient for reliable PFAS removal. Adsorption onto activated carbon has demonstrated strong removal performance in the literature, with hydrophobic interactions identified as the dominant uptake mechanism. As a consequence, longer-chain PFAS compounds, which possess greater hydrophobicity, are consistently better removed than their shorter-chain counterparts.

The experimental results of this study are in agreement with this established understanding. Chain length was one of the primary determinants of removal efficiency and adsorption affinity across all GAC conditions. Long-chain PFAS, particularly PFOA, exhibited the highest adsorption capacity and binding affinity regardless of filter condition, as reflected in both the Langmuir q_{max} and K_L parameters. Notably, even the aged GAC filter, despite its severely limited overall capacity, retained the ability to remove PFOA, underscoring the strong affinity between long-chain PFAS and carbonaceous surfaces. PFOS, although included in the experimental setup, was not detected in any of the laboratory samples and was therefore excluded from the kinetic and isotherm analyses. These findings confirm that carbon chain length is a critical factor governing PFAS adsorption onto GAC, and that this relationship holds across varying degrees of filter aging and regeneration.

Short-chain and ultra-short-chain PFAS proved considerably more difficult to remove. The aged GAC filter achieved no measurable removal of these compounds, and in several cases their effluent concentrations exceeded influent concentrations, suggesting desorption of previously accumulated PFAS from the filter media. This behaviour may indicate competitive adsorption, whereby longer-chain PFAS displace shorter-chain compounds from available surface sites due to their higher affinity. Collectively, these observations highlight that PFAS removal by GAC is governed

not only by filter condition, but also by the physicochemical properties of individual compounds and the presence of competing micropollutants in the water matrix.

The fresh GAC filter achieved rapid and near complete removal of all analysed PFAS compounds, with adsorption kinetics well described by the pseudo-first-order model. This is consistent with the occupation of abundant, accessible surface sites and the dominance of external mass transfer, characteristic of an intact and unmodified carbon matrix. The regenerated GAC filter also achieved meaningful removal across all compounds, though performance was reduced relative to fresh GAC and showed stronger chain-length dependence. The pseudo-second-order model provided a better fit for the regenerated filter, reflecting a shift toward surface interaction limited kinetics and greater site heterogeneity, consistent with the structural and chemical changes to the carbon matrix induced by thermal regeneration, as confirmed by SEM imaging.

Overall, this study demonstrates that GAC filtration is an effective method for PFAS removal in drinking water treatment, but that performance is strongly dependent on filter condition, PFAS chain length and the presence of competing organic micro pollutants. Fresh GAC provides reliable broad-spectrum removal, regenerated GAC retains substantial capacity but with reduced efficiency for shorter-chain compounds, and aged GAC presents an operational risk, particularly with respect to the potential release of short-chain PFAS into treated water. These findings highlight the importance of filter monitoring and timely regeneration or replacement in full-scale drinking water treatment systems.

5.2 Further studies

Several aspects of PFAS adsorption onto GAC require further investigation to build on the findings of this study.

The behaviour of PFOS during GAC adsorption requires closer examination. Due to its sulphonate functional group and distinct molecular structure compared to the carboxylate PFAS that showed results in this study, it remains unclear whether PFOS is removed through the same adsorption mechanisms, and whether it competes with other PFAS compounds for available adsorption sites. Of particular interest would be exposing aged GAC exclusively to long-chain PFAS such as PFOS, in order to assess whether previously accumulated short-chain PFAS are displaced and released into the effluent. Such experiments would provide valuable insight into competitive adsorption dynamics and the potential risks associated with aged filter media in systems where PFAS composition varies over time.

The relationship between filter age and adsorption performance could be investigated more systematically. This study demonstrated that a heavily aged GAC filter had severely limited capacity, but the threshold at which filter performance becomes unacceptable remains undefined. Future work should examine GAC filters of varying operational ages to determine at what point breakthrough occurs and performance

deteriorates to an unacceptable level, which would provide practical guidance for filter replacement intervals in full-scale treatment systems.

Isotherm experiments for fresh GAC were inconclusive in this study due to the exceptionally high removal efficiency of the media, which prevented the establishment of meaningful equilibrium concentrations across the tested range. Future isotherm studies on F-GAC should employ substantially higher adsorbate-to-adsorbent ratios to generate the concentration gradient necessary for model fitting.

Finally, the effect of water matrix composition on PFAS adsorption performance represents an important area for future research. Natural water matrices contain competing organic matter, ions, and other micro pollutants that may influence adsorption kinetics and capacity. Investigating these matrix effects would improve the transferability of the results to real-world drinking water treatment conditions.

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A

PFAS concentration

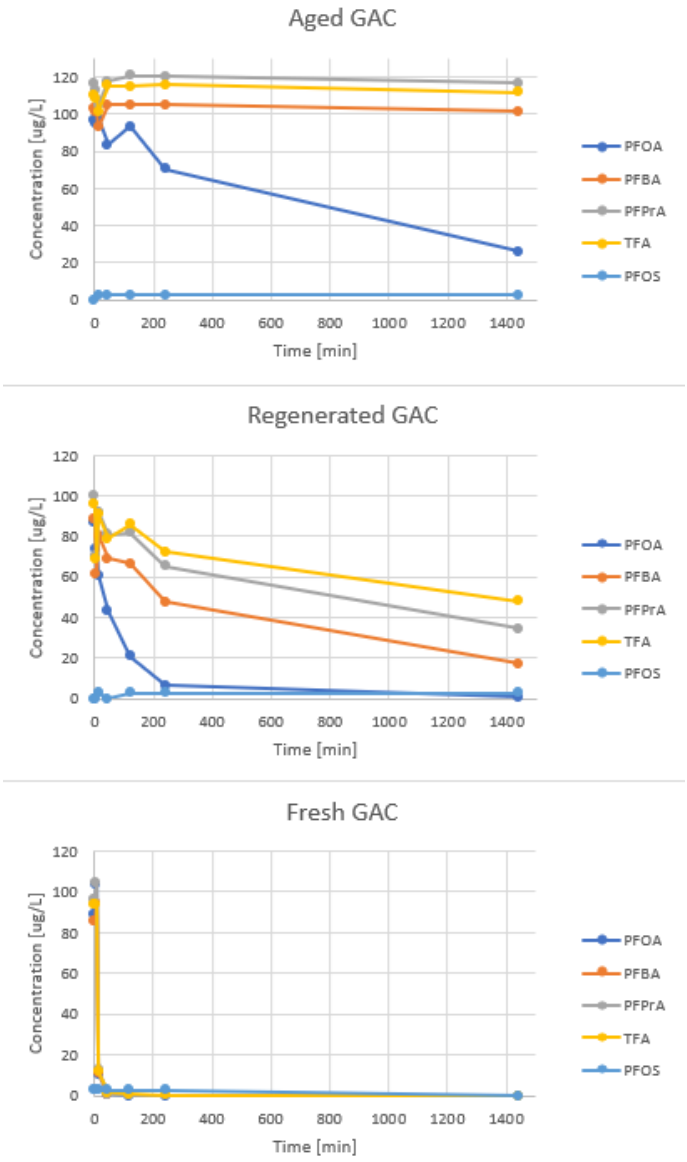

Figure A.1: PFAC concentration in A-GAC, R-GAC and F-GAC over time.

Table A.1: PFAS concentrations measured during the kinetic batch experiment for A-GAC.

Time (min)	**TFA (μg/L)**	**PFPrA (μg/L)**	**PFBA (μg/L)**	**PFOA (μg/L)**	**PFOS (μg/L)**
0	110.3	116.6	103.1	96.6	0
5	108.5	112.8	100.9	94.8	0
15	101.4	104.9	93.0	98.0	0
45	115.5	117.6	105.0	83.4	0
120	114.9	120.6	105.2	93.1	0
240	115.6	120.4	104.8	70.5	0
1440	111.9	116.8	101.5	26.2	0

Table A.2: PFAS concentrations measured during the kinetic batch experiment for R-GAC.

Time (min)	**TFA (μg/L)**	**PFPrA (μg/L)**	**PFBA (μg/L)**	**PFOA (μg/L)**	**PFOS (μg/L)**
0	100.7	104.5	94.4	94.9	0
5	100.3	103.1	91.9	85.3	0
15	91.2	94.4	82.0	73.1	0
45	79.6	80.3	68.5	45.1	0
120	80.1	77.7	63.0	15.8	0
240	71.1	66.8	50.9	8.5	0
1440	53.9	45.8	26.9	1.4	0

Table A.3: PFAS concentrations measured during the kinetic batch experiment for F-GAC.

Time (min)	**TFA (μg/L)**	**PFPrA (μg/L)**	**PFBA (μg/L)**	**PFOA (μg/L)**	**PFOS (μg/L)**
0	93.7	96.6	85.8	89.1	0
5	94.0	104.3	94.5	103.8	0
15	12.0	12.6	11.4	10.4	0
45	1.5	0.9	0.3	0.7	0
120	0.6	0.3	0	0.1	0
240	0.3	0.2	0	0	0
1440	0.1	0	0	0	0

B

Chromatograms for standards

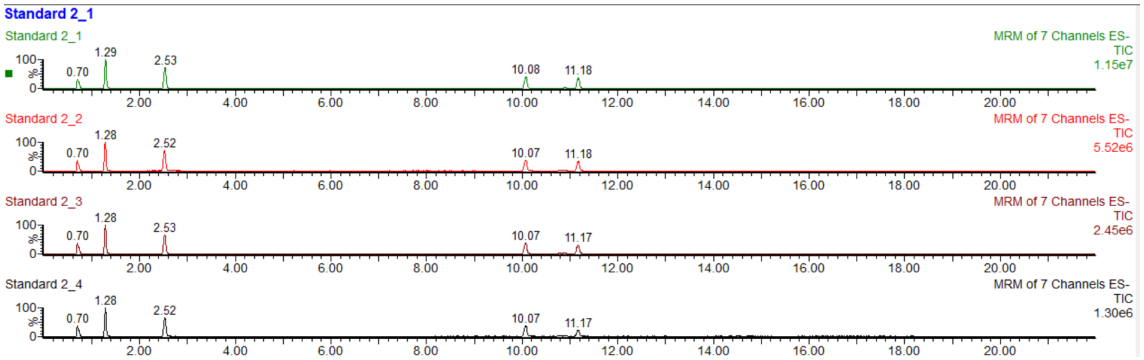

Figure B.1: Chromatography curves for standards 1-4

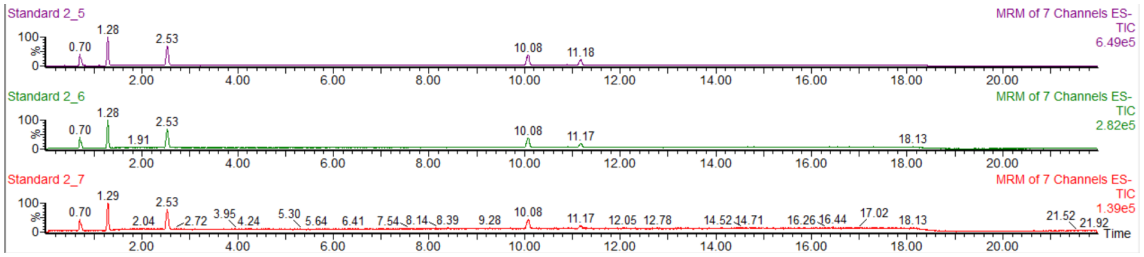

Figure B.2: Chromatography curves for standards 5-7

C

Calibration curves

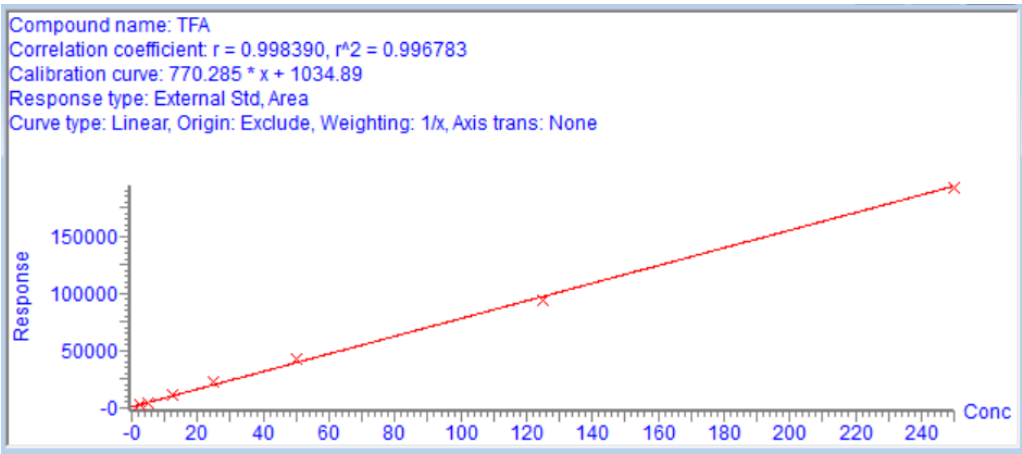

Figure C.1: TFA calibration curve

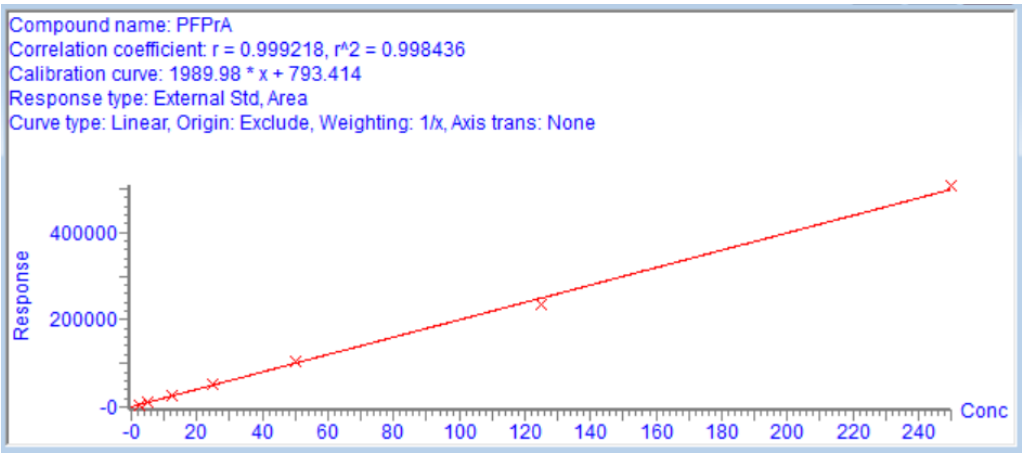

Figure C.2: PFPrA calibration curve

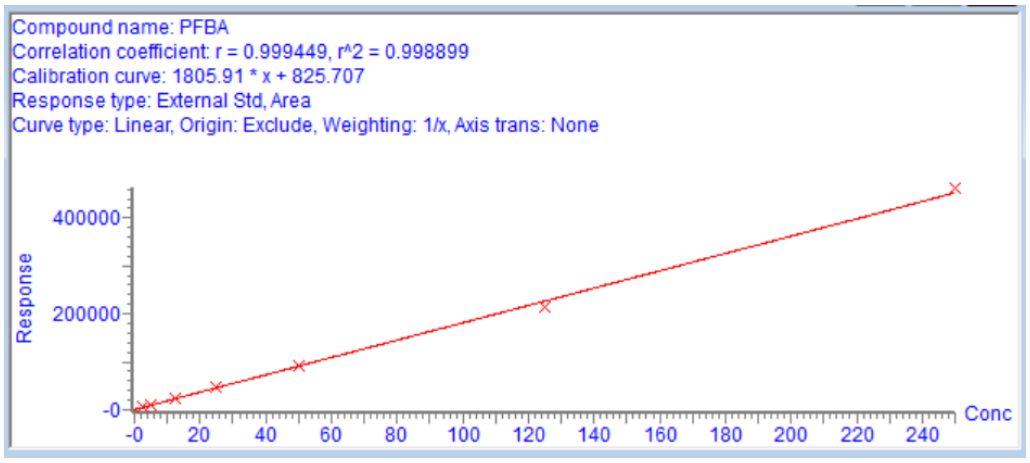

Figure C.3: PFBA calibration curve

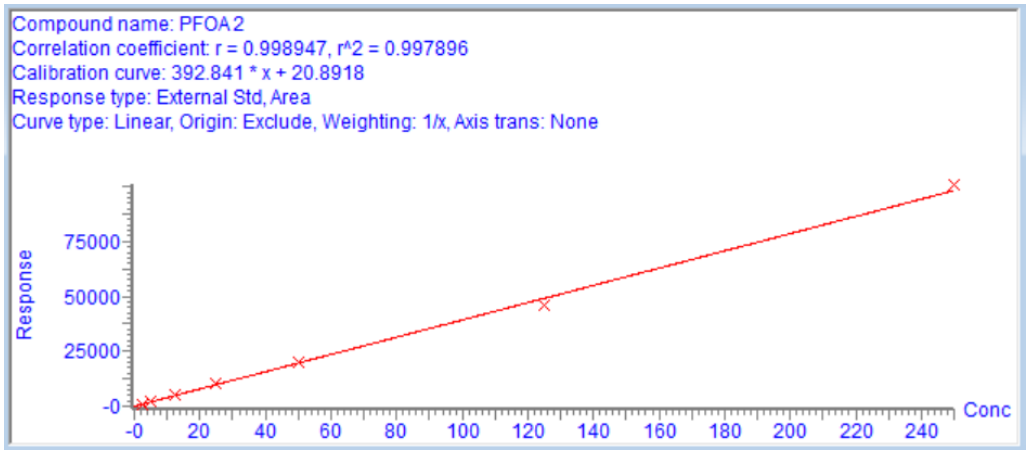

Figure C.4: PFOA calibration curve

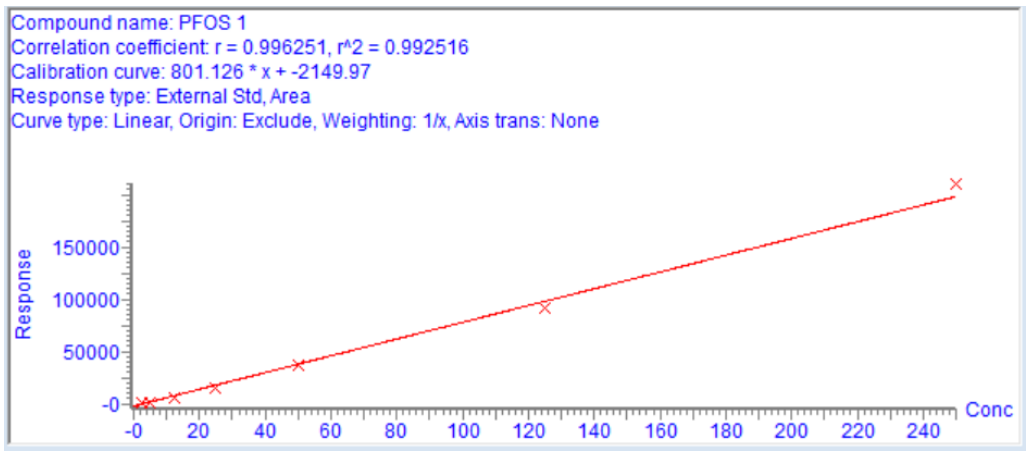

Figure C.5: PFOS calibration curve

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