Evaluation of two-stage GAC filtration for PFAS reduction in drinking water

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Evaluation of two-stage GAC filtration for PFAS reduction in drinking water

Utvärdering av tvåstegs GAC-filtrering för PFAS reducering i dricksvatten

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Abstract

PFAS, or per- and polyfluoroalkyl substances, are a group of chemicals found in a wide range of industrial and consumer products. Due to their persistent nature and widespread use, concerns have been raised about their impact on human health and the environment. Humans are exposed to PFAS through the air and dust (both indoor and outdoor) as well as consumption (water and diet). As a result, various jurisdictions around the world are taking steps to regulate these substances. The new regulation limits the sum of PFAS 4 to 4 ng/L in Sweden with a transitional period that will end in 2026. This thesis, performed in collaboration with Uppsala Water and Waste Ltd, aimed to evaluate the removal efficiency of PFAS with two-stage granular active carbon (GAC) filtration and determine the number of bed volumes before individual PFAS breakthroughs and how the flow rate and empty bed contact time (EBCT) affects the time until GAC regeneration, as well as the selective removal of long versus short PFAS. The hypothesis of this study was that a two-stage GAC filtration would enhance removal efficiency and extend the time until GAC regeneration compared to a single-stage GAC filtration. The assumption is that a two-stage process would double the number of bed volumes before PFAS 4 breakthrough. A reduction in flow rate would lead to higher removal efficiency by extending the empty bed contact time (EBCT), ultimately prolonging the time until breakthrough for both stages due to increased adsorption. This thesis is based on a pilot experiment that consisted of eleven plastic columns divided into two stages. Nine of the columns were filled with three different granular activated carbon (GAC) – new Aquasorb® 6300 and both new and regenerated (“Old”) Filtrasorb® 400-E. The experiment started in June 2022 and continued until January 2023. Approximately 40 000 bed volumes of water were treated during this time. The analysis included a total of 34 PFASs. The results showed that a two-stage process was more effective in removing PFAS and extending the time until GAC regeneration compared to a single-stage process. The study also found that long-chained PFASs have a higher level of adsorption than short-chained ones, which is consistent with previous research. The lower flow rate gave a lower removal efficiency than the higher one, contradicting the hypothesis, but this is probably due to the amount of water already treated by the columns before the flow rate was reduced. Thus, the effects of the flow rates could not be directly compared. Based on previous research, a lower flow rate should provide better removal efficiency. The study concluded that a two-staged GAC filtration can approximately treat three and a half times as many bed volumes as a single stage before the limit of PFAS 4, 4 ng/L were reached. These results are unique for Bäcklösa drinking water treatment plant during this time period, and PFAS concentration and removal efficiency could vary elsewhere. The evaluation of removal techniques in drinking water production contributes to protecting human health from harmful water pollution.

Keywords: Per- and polyfluoroalkyl substances (PFAS), Granular activated carbon (GAC), drinking water, removal efficiency, DWTP

Sammanfattning

Per- och polyfluoralkylsubstanser (PFAS) är en grupp kemikalier som vanligtvis återfinns i en rad olika industri- och konsumentprodukter. På grund av deras härdighet mot nedbrytning och utbredda användning har oro väckts om deras påverkan på människors och naturs hälsa. Människor exponeras huvudsakligen för PFAS genom luft och damm (både inomhus och utomhus) samt genom konsumtion (vatten och kost). Som ett resultat av detta vidtar olika myndigheter och beslutsfattare runt om i världen åtgärder för att reglera PFAS ämnen. I Sverige har en ny reglering fastställt gränsen för PFAS 4 till 4 ng/L, med en övergångsperiod som avslutas 2026. Detta examensarbete, som utfördes i samarbete med Uppsala Vatten och Avfall AB, syftade till att utvärdera avskiljningseffektiviten av PFAS med en tvåstegs filtreringsprocess med granulärt aktivt kol (GAC). Arbetet syftade även till att bestämma antalet bäddvolymer vatten tills gränsvärden för individuella PFAS:er överskrids, effekterna av flödeshastighet på tiden till regenerering av GAC och den selektiva borttagningen av långa och korta PFAS. Hypotesen i det här arbetet var att en tvåstegs GAC-filtrering skulle förbättra avskiljningseffektiviten och förlänga tiden till GAC-regenerering jämfört med en enstegsprocess. I hypotesen antogs att en tvåstegsprocess skulle kunna fördubbla antalet bäddvolymer före överskridande av nya gränsvärdet för PFAS 4. Pilotexperimentet bestod av elva plastkolonner uppdelade i två steg, där nio av kolonner fylldes med tre olika granulärt aktivt kol (GAC) – ny Aquasorb® 6300 och både nytt och regenererat ("gammalt") Filtrasorb® 400-E. Experimentet startade i juni 2022 och pågick till januari 2023. Under denna tid behandlades cirka 40 000 bäddvolymer av vatten, och analysen omfattade totalt 34 PFAS:er. Resultaten från arbetet visade att en tvåstegsprocess mer effektivt tar bort PFAS ur dricksvatten samt förlänger tiden till GAC-regenerering jämfört med en enstegsprocess. Dessutom visade resultaten att långkedjade PFAS:er har en högre adsorption än kortkedjade, vilket stämmer överens med tidigare forskning. Den lägre flödeshastigheten gav en sämre avskiljningseffektivitet än den högre, i motsats till vad hypotesen förutsade, men detta berodde förmodligen på att kolonnerna redan hade använts för att behandla en stor volym vatten innan flödeshastigheten i experimentet sänktes. Därmed kunde inte effekten av olika flöden jämföras direkt. Baserat på tidigare forskning bör lägre flödeshastighet ge bättre avskiljningseffektivitet. Slutsatsen av arbetet var att en tvåstegs GAC-filtreringsprocess kan behandla ungefär tre och en halv gånger så många bäddvolymer vatten som en enstegsprocess innan gränsvärdet för PFAS 4 på 4 ng/L överskrids. Det är värt att notera att dessa resultat är unika för Bäcklösa grundvatten under denna tidsperiod, och PFAS-koncentrationen och avskiljningseffektiviten kan variera för andra grundvattentäkter (och tidsperioder). Utvärderingen av borttagningsmetoder i dricksvattenproduktionen bidrar till att skydda människors hälsa från skadliga vattenföroreningar.

Nyckelord: Per- och polyfluoralkylsubstanser (PFAS), Granulärt aktivt kol (GAC), dricksvatten, avskiljningseffektivitet, vattenverk

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Abbreviations

4:2 FTSA	Fluorotelomer sulfonic acid
6:2 FTSA	Fluorotelomer sulfonate
8:2 FTSA	Fluorotelomer sulfonate
AE	Anion exchange
AIX	Anion exchange
BV	Bed volume
DOC	Dissolved organic carbon
DWTP	Drinking water treatment plant
EBCT	Empty bed contact time
ECHA	The European Chemicals Agency
EFSA	The European Food Safety Authority
EtFOSA	N-ethyl perfluorooctane sulfonamide
EtFOSAA	N-ethylperfluorooctanesulfonamido acid
EtFOSE	N-ethyl perfluorooctane sulfonamidoethanol
EPA	United States Environmental Protection Agency
EU	The European Union
FOSA	Perfluorooctanesulfonamide 1
FOSAA	Perfluorooctane sulfonamidoacetic acid
GAC	Granular activated carbon
HPFHpA	7H-perfluoroheptanoic acid
IX	Ion exchange
LOR	Limit of reporting
MeFOSA	N-methyl perfluorooctane sulfonamide
MeFOSAA	N-methylperfluorooctanesulfonamido acid
MeFOSE	N-methyl perfluorooctane sulfonamidoethanol
MU	Measurement uncertainty
NF	Nanofiltration
PFAS	Per- and polyfluoroalkyl substances
PFOA	Perfluorooctanoic acid
PFNA	Perfluorononanoic acid
PFHxS	Perfluorohexanesulfonic acid
PFOS	Perfluorooctanesulfonic acid
PFBA	Perfluorobutanoic acid
PFPeA	Perfluoropentanoic acid
PFHxA	Perfluorohexanoic acid
PFHpA	Perfluoroheptanoic acid
PFDA	Perfluorodecanoic acid
PFBS	Perfluorobutanesulfonic acid

PFUnDA	Perfluoroundecanoic acid
PFDoDA	Perfluorododecanoic acid
PFTrDA	Perfluorotridecanoic acid
PFPeS	Perfluoropentanesulfonic acid
PFHpS	Perfluoroheptanesulfonic acid
PFNS	Perfluorononanesulfonic acid
PFDS	Perfluorodecanesulfonic acid
PFUnDS	Perflouroundecane sulfonate
PFDoDS	Perfluorodecanesulfonic acid
PF37DMOA	Perfluoro-37-dimethyloctanoic acid
PFTeDA	Perfluorotetradecanoic acid
SLV	Swedish Food Agency (Livsmedelsverket)
RE	Removal efficiency
RO	Reverse Osmosis

Introduction

In Sweden, the water produced by municipalities undergoes the strictest control, meeting the quality standards set by the EU directive 2020/2184 and the Swedish Food Agency to guarantee its purity and absence of harmful microorganisms or chemicals (Svenskt Vatten AB 2021; LIVSFS 2022:12). Only cold tap water is considered food and thus regulated. The Swedish Water organization (2017, 2021) emphasizes that hot tap water should never be consumed as it may contain higher levels of bacteria and trace elements of substances such as metals like copper. The exposure to PFAS in humans mainly comes from the air and dust (both indoor and outdoor) and consumption (water and diet) (Shoeib et al. 2004; Vestergren & Cousins 2009; US Environmental Protection Agency 2016a; b; Hu et al. 2019). Several studies indicated widespread PFAS exposure in humans, wildlife and water sources (Olsen et al. 2007; Vestergren & Cousins 2009; US Environmental Protection Agency 2016b; a; Fiedler & Sadia 2021). For example, Fiedler and Sadia (2021) found the presence of PFASs in human milk samples from various regions around the world as part of the global monitoring plan under the Stockholm Convention on Persistent Organic Pollutants from 2016-2019. The widely spread, and therefore the likelihood of exposure, with the growing evidence linking exposure to PFASs with negative health effects among humans, lead to PFOS being phased out in 2001. Health consequences include a potentially increased risk for kidney cancer (Li et al. 2022), thyroid issues (Andersson et al. 2019; Li et al. 2021) and negatively affecting reproduction health (Blake & Fenton 2020; Ding et al. 2020; Hammarstrand et al. 2021).

All PFAS substances are produced synthetically and do not occur naturally in the environment and are notoriously resistant to degradation, either as individual compounds or as degradation products, and as a result, they persist in the environment (Kemikalieinspektionen 2023). With the strong polar covalent bond between the carbons and fluorides, the PFASs have high thermal and chemical stability (Rahman et al. 2014). The high stability is making them difficult to destroy and together with their ability to repel both oil and water, they are used as a surfactant in multiple industrial and consumer products. For example, fire extinguishers, non-stick cooking pans, textiles, leather, paper packaging, cleaning agents, paints, ski wax, and cosmetics (Rahman et al. 2014; Franke et al. 2021; Kemikalieinspektionen 2023). The provision of safe drinking water and reduction of PFAS in production and circulation can be linked to several sustainable development goals set by the United Nations for 2030. These include Clean Water and Sanitation (Goal 6), Good Health and Well-being (Goal 3), Responsible Consumption and Production (Goal 12), Life Below Water (Goal 14), and Life on Land (Goal 15).

The most common technique utilized to remove PFASs from drinking water is filtration with granular activated carbon (GAC) materials. The adsorption capacity of GAC for PFASs is however impacted by the chain length and functional group of the compounds. Short-chain PFCAs and PFSAs are known to break through quickly, making GAC filtration challenging for long-term PFAS removal efficacy (Franke et al. 2021). This project aimed to evaluate the removal efficiency of PFASs when using two-stage filtration with GAC. As well as addressing how many bed volumes of water a two-stage filtration system can treat compared to a single-stage system.

1.1 Hypotheses

It is hypothesized that implementing a two-stage GAC filtration will enhance removal efficiency and extend the time until GAC regeneration, compared to a single-stage GAC filtration. The hypothesis is that two-stage GAC adsorption provides more efficient use of GAC to remove PFASs, especially for PFAS 4. The assumption is that a two-stage process will approximately double the number of bed volumes before PFAS 4 breakthrough. Additionally, it is believed that a reduction in flow rate will lead to higher removal efficiency by extending the empty bed contact time (EBCT), ultimately prolonging the time until breakthrough for both stages due to increased adsorption.

Literature review

2.1 PFAS classes

Polyfluoroalkyl and perfluoroalkyl substances (PFASs) are a broad and intricate group of more than 4,700 different substances with diverse properties and extensive application in society (Franke et al. 2021). What unites all PFAS substances is their marked resistance to degradation, and some may have damaging effects on human health and the environment. All PFAS substances are produced synthetically and do not occur naturally in the environment and are notoriously resistant to degradation, either as individual compounds or as degradation products, and as a result, they persist in the environment (Kemikalieinspektionen 2023). PFASs can be subdivided based on functional groups. The class perfluoroalkyl substances include, for example, perfluoroalkane sulfonates (PFSAs), perfluoroalkyl carboxylates (PFCAs) and perfluoroalkyl sulfonamidoacetic acids (FASAAs) (Buck et al. 2011; Ahrens & Bundschuh 2014). The most common PFASs are PFOSs with C8 fluorocarbon, as illustrated in Figure 1, and PFOAs with C7 fluorocarbon (Ahrens & Bundschuh 2014). Short-chain PFASs are predominantly hydrophilic, and as a result, they are more mobile in water systems. Long-chain PFASs tend to bind to particles since they have a higher hydrophobicity, thus leading to a significant bioaccumulation potential compared to short-chained PFASs (Ahrens & Bundschuh 2014). According to the terminology proposed by Buck et al. (2011), short-chained perfluoroalkyl sulfonic acids (PFSAs) have a carbon chain length of six or fewer carbons, while short-chained perfluoroalkyl carboxylic acids (PFCAs) have eight or fewer carbons in their chain.

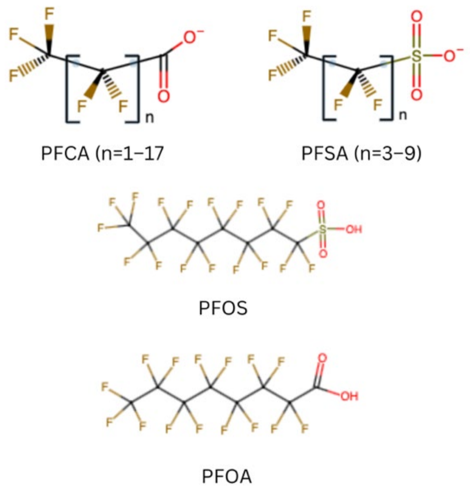

PFCA (n=1–17)
PFSA (n=3–9)
PFOS
PFOA

Figure 1. Chemical structures of PFCA, PFSA, PFOS and PFOA. The illustrations were made by the author using Marvin JS by ChemAxon with inspiration from Ahrens & Bundschuh (2014) and Östlund (2015).

PFOA is classified as a persistent, bioaccumulative, and toxic (PBT) substance, meaning that it is proven to be difficult to break down, accumulate in living organisms, and have toxic effects (Kemikalieinspektionen 2023). PFOAs seem to stay at a similar concentration level in species from different trophic levels, while PFOSs have been shown to accumulate through the food chain (Ahrens & Bundschuh 2014). The levels of PFOS have declined in biota since the phasing out in 2001 while the long-chained PFCAs demonstrated either stagnant or increasing trends depending on chemical properties, specie and location (ibid.). Due to this and the degradation of PFAS precursors to PFCA and PFSA, Ahrens & Bundschuh (2014) argue that the PFASs can be considered persistent in the environment. The accumulation potential of especially the long-chained PFASs degradation product PFCA results in organisms downstream being more exposed (Ahrens & Bundschuh 2014).

Because PFASs are both hydrophobic and oleophobic, they do not accumulate in adipose tissue like other bioaccumulative substances. Instead, they bind to proteins and accumulate in other organs such as the liver and blood (Kemikalieinspektionen 2023). In particular, PFASs easily bind to serum albumin and fatty acid-binding proteins, causing the PFASs to accumulate in tissue (Ahrens & Bundschuh 2014). The bioaccumulation potential of PFASs varies depending on their chemical and structural properties, as well as the species (ibid.). Due to the resistance of PFAS substances to degradation and their mobility in soil, combined with the fact that many of these substances are water-soluble, drinking water sources are at risk of long-lasting contamination. The spread and exposure of PFAS substances occur throughout the entire life cycle of the substance, starting from its manufacturing and ending with its disposal (Ahrens & Bundschuh 2014; Kemikalieinspektionen 2023).

2.2 Consequences of PFAS in drinking water

Several PFAS substances, including PFOS, PFOA, and PFNA, have been classified as suspected carcinogens and reproductive hazards for humans and wildlife. For the vast majority of PFAS compounds, however, their effects on health are still largely unknown. Nevertheless, there are compelling reasons to consider all PFAS as deleterious to health (Kemikalieinspektionen 2023). Most studies on the health effects of PFAS are experimental studies on animals. These studies have shown that PFAS exposure can cause numerous effects in mammals, such as liver and blood lipid disorders, thyroid hormone abnormalities, immune system dysfunction, and reproductive issues. Certain PFAS substances have also been observed to cause tumours and mammary gland development (ibid.). Although further studies are required to determine whether these findings are relevant to humans, several studies have already been conducted on groups of people living in areas with high concentrations of PFAS in their groundwater.

Studies on cancer incidence or mortality at occupational or environmental exposure to PFOS and PFOA included in a risk assessment from EFSA (2020) provided insufficient support for human carcinogenicity. According to EFSA, this aligns with the recent International Agency for Research on Cancer (IARC) report on PFOA (2016), which found limited evidence for carcinogenicity. Studies on this subject between IARC's and EFSA's assessment have not changed EFSA's conclusion for PFOS and PFOA, and pointed out the limited information on other PFASs effects (EFSA 2020). A later study by Li et al. (2022) examined the association between exposure to PFASs in drinking water and cancer incidence in Ronneby, Sweden which had high levels of PFAS exposure through their drinking water. They found that high exposure to PFASs in drinking water was associated with a moderately increased risk of kidney cancer in this population. Li et al. highlighted the need for further research on the health effects of PFAS exposure and the importance of reducing it.

The study by Andersson et al. (2019) investigated the association between exposure to perfluorinated compounds (PFCs) in drinking water and thyroid disease in a cohort of individuals from Ronneby, Sweden. The study found that high exposure to PFCs in drinking water was associated with an increased risk of thyroid disease. But no association between PFAS in drinking water and hyperthyroidism could be concluded. Likewise, could not Li et al. (2021) conclude an association between PFAS in drinking water to thyroid hormones in any age group in Ronneby, Sweden. But the study's strongest evidence of association where in young boys who had been exposed to PFAS around their birth. Li et al. (2021) suggested that further research should be conducted on the impact of in utero and infant exposure on thyroid development. This since thyroid hormones play a crucial role in the growth and development of fetuses and infants (ibid.).

Studies done in the US however found that exposure to PFASs may alter thyroid function (Lewis et al. 2015; Crawford et al. 2017). Lewis et al. identified increased levels of the thyroid hormones FT3, TT3, and FT4 among adult females. But during adolescence, they found an increase in thyroid-stimulating hormone (TSH) levels in males and a decrease in TSH levels in females (ibid.).

The study of Crawford et al. (2017) found PFC levels to be related to thyroid hormone levels in women of reproductive age. A positive correlation could be made between PFOA and T3 and PFNA with both T3 and fT4. They did not find a correlation between PFCs and TSH like Lewis et al. (2015). The studies suggest that exposure to PFASs may disrupt thyroid hormone homeostasis.

Apart from studying the PFC linked to levels of thyroid hormone, the study of Crawford et al. was mainly studying associations between PFCs and ovarian reserve, which is an indicator of a woman's remaining fertility potential. But there was not a significant association between the two in their report. On the other hand, Ding et al. (2020) found that PFAS exposure can lead to changes in the structure and function of the ovary, including disruption of the menstrual cycle, decreased ovarian reserve, and reduced fertility. The reason for this would be the capability of PFAS to penetrate the blood-follicle barrier. Ding et al. suggest that PFAS exposure may interfere with hormonal signalling and oxidative stress in the ovary, leading to these negative effects.

Hammarstrand et al. (2021) explored the association between exposure to perfluoroalkyl substances (PFASs) in drinking water and the risk of polycystic ovarian syndrome (PCOS), uterine leiomyoma (fibroids), and endometriosis in a Swedish cohort. The study found that exposure to PFASs in drinking water was associated with an increased risk of PCOS and perhaps fibroids, but not for endometriosis. Furthermore, Blake and Fenton (2020) reviewed the impact of PFAS exposure on health outcomes. The study found that exposure to PFAS during early life, particularly in utero through the placenta, can lead to latent health effects, including endocrine disruption, decreased fertility, and altered immune function. The study suggests that the placenta may act as a target tissue for PFAS and play a role in peri- and postnatal effects and suggest further studies on the potential role of the placenta in mediating these effects. All mentioned studies highlight the need for further research on the impact of PFC and PFAS exposure on reproductive health and underscore the importance of reducing PFAS exposure in the environment to protect human health. In addition, other studies have shown that PFAS substances can also affect the immune system, birth weight, cholesterol levels in the blood and liver enzymes (SLV 2023).

2.3 Current limits and legislation in the EU and Sweden

Legislation at both global, EU and national levels regulates individual PFAS substances. Several PFASs, such as PFOA, perfluorinated carboxylic acids (C9-14 PFCAs), and PFHxS, are listed on the candidate list of substances of very high concern (SVHC) under the REACH regulation due to their threat to human health and wildlife (Kemikalieinspektionen 2023; ECHA n.d.). The use of some of those substances – PFOS, PFOA and PFHxS, along with their salts and precursors – are limited by EU’s POPs (Persistent Organic Pollutants) regulation via The Stockholm Convention. The European Commission has decided to restrict perfluorinated carboxylic acids (C9-14 PFCAs), their salts and precursors starting February 2023 (ECHA n.d.).

A grouping approach is reflected in the EU Directive 2020/2184 imposing a limit of 0.5 µg/l (500 ng/L) for “the totality of per- and polyfluoroalkyl substances”. But this limit will only apply once the technical guidelines for monitoring this are established by the EU Commission, which has a deadline of January 12, 2024. The maximum limit for “Sum of PFAS” (the same as PFAS 20) in the Member States of the EU is 0.10 µg/L (100 ng/L) (EU 2020/2184). The Swedish regulation has a limit of 100 ng/L for sum PFAS 21 (PFAS 20 and 6:2 FTSA) (LIVSFS 2022:12). New drinking water regulations have been adopted by the Swedish Food Agency in response to the EU Directive 2020/2184 on the quality of drinking water. The directive sets minimum requirements for Member States to implement in their national laws to protect human health from the harmful effects of drinking water pollution and improve access to drinking water (SLV 2022). The new regulation limits the sum of PFAS 4 to 4 ng/L in Sweden and the transitional period will end on January 1, 2026 (LIVSFS 2022:12; SLV 2022).

2.4 Treatment technology in drinking water

The effectiveness of treatment for PFAS in drinking water can vary based on the specific type of PFAS present in the incoming water, and therefore different PFAS may require different treatments to achieve the best removal.

Nanofiltration (NF) and reverse osmosis (RO) are both membrane filtration technologies but differ in their mechanism of action and the type of contaminants they remove (US Environmental Protection Agency 2018). Reverse osmosis uses high pressure and a semipermeable membrane to remove dissolved salts, total dissolved solids, and inorganic contaminants from water. RO will remove a wider range of compounds compared to nanofiltration (ibid.). NF is a lower-pressure filtration process than RO and uses a semi-permeable membrane to separate charged particles based on their size and charge. NF is typically used to remove dissolved organic compounds, ions, and smaller-sized inorganic contaminants while leaving minerals RO would have removed (US Environmental Protection Agency 2018; Franke et al. 2019, 2021). The advantages of membranes are that they remove both long and short-chained PFASs effectively, along with other contaminates as well (Crone et al. 2019; Franke et al. 2019, 2021). The disadvantages of this technology are the concentrated waste that needs further treatment to dispose of the contaminates making membrane filtration more expensive than other treatment options (Crone et al. 2019). Furthermore, Franke et al. (2021) emphasise that regulations and guidelines for drinking water and discharge into the environment largely affect the economic cost of membrane treatments.

Ion exchange (IX) treatment uses a resin material to exchange ions in the water with ions that are bound to the resin, effectively removing PFAS and other impurities (US Environmental Protection Agency 2018). Anion exchange (AE or AIX) is a specific type of ion exchange that is designed to remove anions from water, while ion exchange is a more general term that refers to the removal of both cations and anions from water (McCleaf et al. 2017; US Environmental Protection Agency 2018; Franke et al. 2019). The advantages of anion exchange are easy to target short chained PFASs and relatively simple operation (Crone et al. 2019). While one disadvantage is the treatment resin needs after usage (ibid.).

Granular active carbon (GAC) filtration uses activated carbon to adsorb PFAS, among other compounds, removing them from the water. GAC is the most researched and established technology for removing PFAS in drinking water treatment (US Environmental Protection Agency 2018; Franke et al. 2021). Activated carbon is a widely used treatment method in drinking water systems to remove natural organic compounds, taste and odour compounds, and synthetic organic chemicals. This is achieved through the process of adsorption, which involves the accumulation of a substance, like PFAS, at the boundary between a liquid and a solid. The high porosity of activated carbon gives it a large surface area, making it an effective adsorbent for contaminants. Activated carbon is produced from organic materials with high carbon content, such as wood, coal, and lignite, and is frequently used in the granular form known as granular activated carbon (GAC) (US Environmental Protection Agency 2018). GAC's adsorption capacity is determined by the compound's length and functional groups and removes long-chained PFAS effectively. The relatively rapid breakdown of some PFASs to short-chained once makes this technology a disadvantage for long-term PFAS removal with frequent regeneration needed to uphold effective PFAS removal (Hansen et al. 2010; Crone et al. 2019; Franke et al. 2021). When the surface is saturated, desorption occurs. This is when substances are released from the surface instead of being adsorbed by it. Therefore, regeneration of GAC is necessary to maintain the effective removal of PFAS (Zaggia et al. 2016).

McCleaf et al. (2017), evaluated the effectiveness of GAC and anion exchange (AE) columns in removing multiple PFASs from drinking water. The results of the study showed that both GAC and AE columns were effective in removing PFASs, with GAC demonstrating higher removal efficiency for long-chain PFASs and AE being more effective for short-chain PFASs. The study concluded that a combination of GAC and AE could be an effective solution for removing a broad range of PFASs from drinking water. Similarly, Franke et al. (2019) concluded that nanofiltration in combination with either GAC or anion exchange removed PFAS more effectively than GAC or anion exchange by themselves. Further, Belkouteb et al. (2020), investigated the performance of GAC in removing PFASs from drinking water at a full-scale treatment plant. The study found that GAC was effective in removing PFASs, with removal efficiency ranging from 60 % to 99 %. There appears to be limited research on the potential capacity of two-stage GAC filtration compared to single-stage filtration.

Materials and methods

3.1 Collaboration and study site

The project was conducted in collaboration with the municipal corporation Uppsala Water and Waste Ltd. This is part of a larger study to assess suitable GAC filtration systems for the removal of PFAS in drinking water treatment plants (DWTP) orchestrated by Philip McCleaf at Uppsala Water and Waste Ltd. The pilot experiment was conducted at Bäcklösa DWTP.

3.2 The experimental design

The evaluation of two-stage GAC-filtration in drinking water was based on a pilot experiment with ten plastic columns divided by stage and new versus reactivated GAC. As seen in Figure 1, the water was entering the experiment via sample point 12 after the full-scale sand filtration process in the DWTP Bäcklösa. Which is the same treatment sequence as the full-scale GAC-filtration. Duplicate columns were used as well as blank columns without GAC to determine if there was any PFAS removal due to the experiment's construction. Details on the setup of the experiment can be found in Appendix 1.

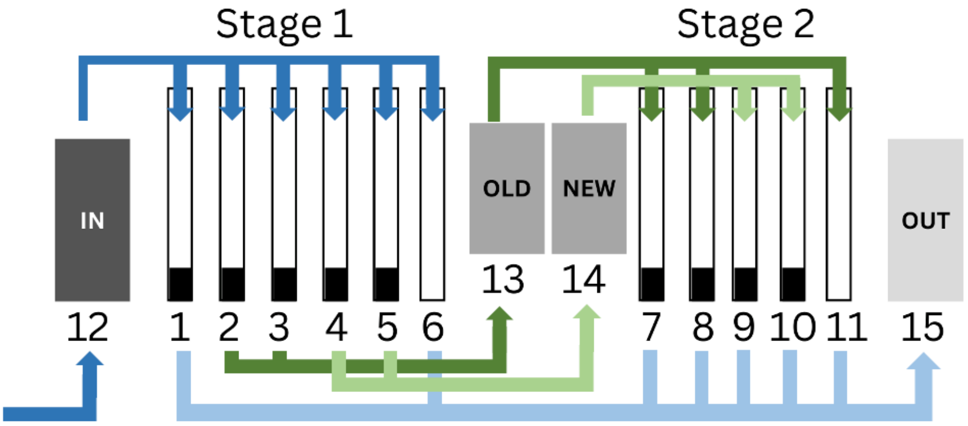


Figure 2. Schematic flow diagram of the pilot experiment and the sample points in Table 1. Incoming water came from the fullscale sand filtration in the DWTP Bäcklösa. The grey squares are referred

to as tanks (sample points number 12–15) and the white rectangles are referred to as columns (sample points 1–11). Sample points 6 and 11 were blanks without GAC. "OLD" and "NEW" are referring to GAC taken from the full-scale GAC filter and new GAC. Electric water pumps transport the water between tanks and columns.

Table 1 presents details about the sampling points, which GAC was used, and sample ID. Product specifications for the different GACs are given in Table 2.

Table 1. Sample point descriptions in the pilot experiment

Sampling point	Stage	Sampling point description	Sample ID	Average Bed volume (BV) (mL)
1	1	Aquasorb 6300 New	1 - 1AS6300	469
2	1	Filtrasorb 400 OLD from Bäcklösa GAC filter 1	2 - 1F400OldA	479
3	1	Filtrasorb 400 OLD from Bäcklösa GAC filter 1	3 - 1F400OldB	473
4	1	Filtrasorb 400 NEW	4 - 1F400NewA	455
5	1	Filtrasorb 400 NEW	5 - 1F400NewB	469
6		Blank	6 - 1 Blank	
7	2	Filtrasorb 400 OLD from Bäcklösa GAC filter 1	7 - 2 F400OldA	481
8	2	Filtrasorb 400 OLD from Bäcklösa GAC filter 1	8 - 2 F400OldB	487
9	2	Filtrasorb 400 NEW	9 - 2 F400NewA	449
10	2	Filtrasorb 400 NEW	10 - 2F400NewB	469
11	2	Blank	11 – 2 Blank	
12		Raw water tank from after Bäcklösa sandfilter 4-6	12 - Tank1Raw	
13		Water after columns 2,3	13 - Tank2AOld	
14		Water after columns 4,5	14 - Tank2BNew	
15		Water after columns 7,8,9,10	15 - Tank 3	

The columns were made by employees at Uppsala Vatten and Waste Ltd with 10 clear acrylic plastic pipes 212 cm x 53.5 mm (height x inner diameter) and stainless metal base with metal outlet valves and watertight rubber connecter between the base and column. At the bottom of the columns, a metal mesh filter was placed to support and keep the GAC in the column. The GAC used were new Aquasorb® 6300 and both new and regenerated ("Old") Filtrasorb® 400-E, as depicted in Table 1. The product specifications are listed in Table 2. The "Old" GAC was taken from about 30 cm deep in the GAC filter 1 at Bäcklösa with a metal sampler. The "Old" GAC was dried at 40 degrees Celcius in the lab oven at the DWTP before being weight and placed in the columns.

Table 2. Specification of GAC

	Aquasorb® 6300	Filtrasorb® 400-E	"Old" Filtrasorb® 400-E
Iodine adsorption min. (mg/g)	970	1000	807
Moisture content, as packed max. (%)	5	2	1
Apparent density min. (kg/m3)	420	520	554
Total ash content max. (%)	15	9	10
Particle size max.			
>12 mesh (0.70 mm)	5%	5%	1.9%
<40 mesh (0.425 mm)	4%	4%	0.7%
Effective Size (mm)	0.6	0.6–0.7	0.77
Methylene blue adsorption (ml/g)	250	300	NA
Surface area (BET) (m^2/g)	1000	1050	NA
Backwashed and drained density (kg/m^3)	380	450	NA
Mean Paticle Diameter (mm)	1.0	1.0	NA
Uniformity Coefficient	1.7	1.7	1.67

Information provided by the production companies Jacobi and Chemviron. The "old" Filtrasorb GAC was analysed at the regeneration facility Jacobi Carbons Service (Europe) GMBH in Premnitz, Germany. NA=not available.

Other materials used to create the experiment were the following.

- 4 plastic tanks 80 cm high and 450 mm in diameter were used as storage tanks for pumping intake tubing.
- Peristaltic pump with TPE tubing (Watson Marlow 520S)
- APE tube natural 3/8" O.D.*0.25" I.D (Parker Legris)
- PVC tube transaparent 8*12 mm 50 m (Lundgrens Beijer Tech)
- PVC tube unarmed 10*14 mm (Lundgrens Beijer Tech)
- PVC pipettes 15 cm, approx 11 cm after modifying
- HDPE containers (Emballator) 95.3*75*approx. 150 mm med metal nozzles with plastic packing to regulate overflow
- Funnels in PP-plastic 11 cm diameter (Menuett)
- A plastic gutter to collect overflow from the containers collecting and redirecting the water from Stage 1 to the sample points 13 and 14, as is shown in Appendix 1.
- Duct tape and PVC tape

During the first half of the time period, the experiment had a flow rate of around 99-98 mL/min, whilst the second half had an average flow rate of 50-49 mL/min. This was to study how the empty bed contact time (EBCT) would affect the removal efficiency of PFASs. As well as speed up the experiment to enable breakthrough of PFAS in the Stage 2 columns within a reasonable period of time.

3.3 Sampling

The pilot experiment started in July 2022 at Bäcklosa DWTP by Philip McCleaf, Uppsala Vatten and Waste Ltd. I was responsible for the sampling and monitoring in November and December 2022.

Water samples from all sampling points shown in Figure 1 were taken every week. Twice a week were pH, temperature, water height, GAC height and flow rate measured. Samples taken every two weeks were sent to ALS Scandinavia AB for PFAS measurements and for chemical analysis sampling occurred every third week. The samples that were not directly sent for analysis were stored in a refrigerator for later analysis if additional data points were required. The analysis method used at ALS was "OV-34aQ Perfluorinated compounds, PFAS (34) (Low LOR) in water".

Samples were collected in 250 mL PE-HD plastic bottles with screw caps and marked with adhesive labels. For sampling points 12–15, grab samples were taken directly from the tanks while samples were taken directly from the flow tube from each column for the other sampling points. Plastic gloves were used for the grab samples when needed. Temperature and pH were measured with Knick Portavo 908 Multi, water and GAC height were measured with a ruler by hand and the flow rate was assessed with a timer and volumetric cylinder.

3.4 Analytic methods

3.4.1 Analysed compounds

In total 34 PFASs were included in the analysis, which is described in Table 2.

Table 3. PFASs analysed. [1]Included in Swedish regulations LIVSFS 2022:12, [2]Included in EU directive 2020/2184.

Analysed compounds		CF length	PFAS 21[1]	PFAS 20[2]	PFAS 11[1]	PFAS 4[2]
PFCA						
Perfluorobutanoic acid	PFBA	C3	X	X	X	
Perfluoropentanoic acid	PFPeA	C4	X	X	X	
Perfluorohexanoic acid	PFHxA	C5	X	X	X	
Perfluoroheptanoic acid	PFHpA	C6	X	X	X	
Perfluorooctanoic acid	PFOA	C7	X	X	X	X
Perfluorononanoic acid	PFNA	C8	X	X	X	X
Perfluorodecanoic acid	PFDA	C9	X	X	X	
Perfluoroundecanoic acid	PFUnDA	C10	X	X		
Perfluorododecanoic acid	PFDoDA	C11	X	X		
Perfluorotridecanoic acid	PFTrDA	C12	X	X		
Perfluorotetradecanoic acid	PFTeDA	C13	X	X		
PFSA						
Perfluorobutanesulfonic acid	PFBS	C4	X	X	X	
Perfluoropentanesulfonic acid	PFPeS	C5	X	X		

Perfluorohexanesulfonic acid	PFHxS	C6	X	X	X	X
Perfluoroheptanesulfonic acid	PFHpS	C7	X	X		
Perfluorooctanesulfonic acid	PFOS	C8	X	X	X	X
Perfluorononanesulfonic acid	PFNS	C9	X	X		
Perfluorodecanesulfonic acid	PFDS	C10	X	X		
Perflouroundecane sulfonate	PFUnDS	C11	X	X		
Perfluorodecanesulfonic acid	PFDoDS	C12	X	X		
PFAS precursors						
Fluorotelomer sulfonic acid	4:2 FTSA	C4				
Fluorotelomer sulfonate	6:2 FTSA	C6	X			
Fluorotelomer sulfonate	8:2 FTSA	C8				
Perfluorooctanesulfonamide 1	FOSA	C8				
N-methyl perfluorooctane sulfonamide	MeFOSA	C8				
N-ethyl perfluorooctane sulfonamide	EtFOSA	C8				
N-methyl perfluorooctane sulfonamidoethanol	MeFOSE	C8				
N-ethyl perfluorooctane sulfonamidoethanol	EtFOSE	C8				
Perfluorooctane sulfonamidoacetic acid	FOSAA	C8				
N-methylperfluorooctanesulfonamido acid	MeFOSAA	C8				
N-ethylperfluorooctanesulfonamido acid	EtFOSAA	C8				
Others						
7H-perfluoroheptanoic acid	HPFHpA	C6				
Perfluoro-37-dimethyloctanoic acid	PF37DMOA					

Chemical characteristics were also measured; temperature, the conductivity of incoming water, pH, alkalinity, UV 254 nm, total organic carbon (TOC), 2,5-Dimethoxy-4-chloroamphetamine (DOC), ammonium (NH4), nitrite (NO2), fluoride (F), chloride (Cl), nitrate (NO3), sulfate (SO4), sodium (NA), magnesium (Mg), aluminium (Al), calcium (Ca), manganese (Mn), iron (Fe), copper (Cu), uranium (U) and potassium (K). Temperature, the conductivity of incoming water and pH was measured at DWTP Bäcklösa. The listed PFASs and the rest of the listed chemical characteristics were analysed at the commercial lab ALS Scandinavia AB in Danderyd. The water quality at sample points 12-15 are presented in Table 8, Appendix 2.

3.5 Calculations

In order to evaluate the removal efficiency, the Bed volumes (BV), the amount of water that has passed the GAC filter has to be calculated for each column according to the following equation.

$$BV_{treaded} = \frac{Q \cdot t}{V_{GAC}}$$

Equation 1. Treated bed volume

where V treated is the treated volume during operation time [m^3], V GAC is the volume of the GAC in the columns [m^3], Q [m^3s^{-1}] is the average flow rate during operation time and t [s] is operation time (Belkouteb et al. 2020). The volume of GAC was calculated using the volume formula for a cylinder with a diameter of 5.35 cm. The height used for the calculation was determined by averaging the two most recent measurements taken at that sampling date, resulting in a height that varied with cumulative bed volume and between columns due to natural variations of the granular sizes.

The following equation was used to assess the removal efficiency of different PFASs, where C_0 [ng L^{-1}] is the concentration of the substance in the incoming water and C [ng L^{-1}] is the concentration of the substance in the outgoing water (Belkouteb et al. 2020).

$$RE = \frac{C_0 - C}{C_0} \cdot 100$$

Equation 2. Removal efficiency

The removal efficiency was then plotted against cumulative bed volumes for different PFASs to determine the amount of water that could be treated before the breakthrough of each PFAS compound and the rate of decrease in removal efficiency for the total and each PFAS compound.

The following equation calculates the empty bed contact time to determine how the flow rate affects the regeneration time.

$$EBCT = \frac{V_{GAC}}{Q}$$

Equation 3. Empty bed contact time

where V GAC is the volume of GAC [m^3] and Q [m^3s^{-1}] is the average flow rate during operation time (Englund 2015).

Pearson’s correlation coefficient was used to determine if the analysed chemical characteristics were correlating with PFAS removal in this experiment. The coefficient varies between +1, positive correlation, and –1, negative correlation. 0 being no correlation between the variables. The correlation was statistically significant if the p-value was lower than 0.05. A p-value was calculated with three degrees of freedom and the t-statistic value was derived from the following formula.

$$t = \frac{r \cdot \sqrt{n-2}}{\sqrt{1-r^2}}$$

Equation 4. T-statistic

where r is the Pearson coefficient and n is the number of data points which was 5.

Results from pilot experiment

The results included in this thesis are from the start of the experiment in June 2022 until January 2023. This study is part of a larger project, and the pilot experiment will continue. Approximately 40 000 bed volumes were treated during this period, with variations between columns due to the irregular sizes of the granulated active carbon. These differences led to variations in GAC height between columns. This study aimed to evaluate the removal efficiency of PFAS with two-stage GAC filtration and determine the number of BV before individual PFAS breakthroughs and how the flow rate (EBCT) affects the regeneration time, as well as the selective removal of long versus short PFAS. A total of 34 PFASs were analysed and Table 4 provides an overview of the compounds categorised as PFAS 4, along with PFAS 11, 21 and total PFAS. The table summarises the average concentration of PFASs in ng/L after each column, with Stage 1 comprising Columns 1–6, where Column 6 is without GAC as a control, and Stage 2 comprising Columns 7–11, where Column 11 is without GAC as a control. According to the concentration values presented in Table 4, Stage 2 considerably reduces the average concentration of PFASs in all columns, for both new and old GAC, below the newly proposed limit of 4 ng/L for PFAS 4. Moreover, the concentration of PFNA was below the detection limit in the incoming water.

According to Table 4, the concentration of PFASs after the control columns was similar to the untreated incoming water for that stage, 12-Tank1Raw for 6-1Blank and 13-Tank2AOld for 11-2Blank. This shows that the construction of the experiment does not affect the removal of PFAS.

Table 4. Average concentration PFAS ng/L per column up to approximately from 0 to 40 000 bed volumes (BV) (n =14). ND=no detection (below detection concentration).

	PFOA	PFNA	PFHxS	PFOS	PFAS 4	PFAS 11	PFAS 21	Total PFAS
12-Tank1Raw	3.9	ND	48	19	71	85	92	134
1-1AS6300	1.4	ND	14	4.9	20	29	32	44
2-1F400OldA	1.4	ND	14	3.9	19	28	31	48
3-1F400OldB	1.4	ND	14	3.7	19	29	32	46
4-1F400NewA	1.1	ND	11	3.7	16	25	27	36
5-1F400NewB	1.1	ND	11	3.9	16	25	27	37
6-1Blank	**4.0**	**ND**	**50**	**20**	**74**	**89**	**95**	**133**
13-Tank2AOld	1.3	ND	12	3.0	17	26	28	42
14-Tank2BNew	1.2	ND	12	4.0	17	26	28	40
7-2F400OldA	0.40	ND	2.3	0.30	2.9	9.3	10	20
8-2F400OldB	0.40	ND	2.1	0.40	2.7	8.9	9.5	19

9-2F400NewA	0.30	ND	2.2	0.60	2.9	7.8	8.3	18
10-2F400NewB	0.30	ND	2.4	0.60	3.2	8.2	8.8	19
11-2Blank	**1.2**	**ND**	**13**	**4.0**	**18**	**27**	**29**	**39**
15-Tank3	0.20	ND	2	0.30	2	7.5	8.1	20

4.1 Removal efficiency of PFAS

Table 5 presents the average removal efficiencies of the whole time period of the two-stage filtration experiment along with the average removal efficiencies of the varying flow rates and empty bed contact times (EBCT). According to the table, PFASs PFBA (C3) and PFHxA (C5) were found to have the least efficient removal rates in regard to the whole experiment and time period. While the longer chain PFHpA (C6), PFOA (C7) and the four PFSA maintained removal efficiency above 80%. This indicates the selectivity of GAC for the longer chained PFASs and PFSA (McCleaf et al. 2017).

The first half of the experiment had a higher flow rate and an EBCT of 4.5 min whilst the latter half had a lower flow rate and an EBCT of 9 min. According to Table 5, longer EBCT appears to generate lower RE, especially for the short-chained PFCAs. It is important to note that the conditions for the two EBCT are not the same. Approximately 27 000 bed volumes of water had gone through the GAC filtration prior to EBCT 9 min and thus cannot directly be compared. In Table 5, the first half of the time period with bed volumes 0–27 000 in Stage 1 appears to have a higher RE % of PFCAs. While the second half of the time period with BV 28 000–40 000 in Stage 2 appears to have a higher RE % of PFSA. When separating the RE % by when the flow rate was changed, the RE % was higher between 0–27 000 BV in both stages. Since the EBCT increased in the latter phase of the experiment, it is possible for the GAC to have less adsorption efficiency after approximately 28 000 BV has gone through (Hansen et al. 2010; McCleaf et al. 2017; Belkouteb et al. 2020). The possibility of lowered removal efficiency after approximately 28 000 BV should be considered, as the EBCT increased during the latter phase of the experiment.

Table 5. Average removal efficiency (RE %) during different bed volumes of the experiment.

RE %		PFCA					PFSA			
n	BV	PFBA –C3	PFPeA –C4	PFHxA –C5	PFHpA –C6	PFOA –C7	PFBS –C4	PFHxS –C6	PFOS –C8	PFPeS –C5
56	0–40 000	44	43	66	81	91	84	95	98	93
28	0–27 000 EBCT 4.5 min	66	71	83	86	93	92	97	98	96
28	28 000–40 000 EBCT 9 min	19	11	47	76	89	75	93	98	90
28	Stage 1 0–27 000. EBCT 4.5 min	50	47	58	60	73	70	79	82	73
28	Stage 1 28 000–40 000	21	4	23	34	55	43	66	77	55

	EBCT 9 min									
28	Stage 2 0–27 000 EBCT 4.5 min	18	39	54	40	52	58	74	67	52
28	Stage 2 28 000–40 000 EBCT 9 min	-2	9	31	65	76	57	80	90	76

4.1.1 Removal efficiency of total PFAS

The removal efficiency (RE %) for total PFAS starts around 90% for Stage 1, as depicted in Figure 3. The RE follows a decreasing trend with cumulative bed volumes (BV) until the flow rate is decreased by half, around 27 000–29 000 BV, depending on the column. In contrast, Stage 2 exhibits a peak RE at a later point, around 16 000–20 000 BV. One possible explanation for the initially low removal efficiency in Stage 2 of the experiment could be associated with the very low concentration of PFASs exiting the first stage due to its high RE. Following the reduced flow rate and increased empty bed contact time (EBCT), both stages show an increased RE, with Stage 1 demonstrating a higher RE than Stage 2. The average RE after two-stage GAC filtration for Total PFAS was 86 %.

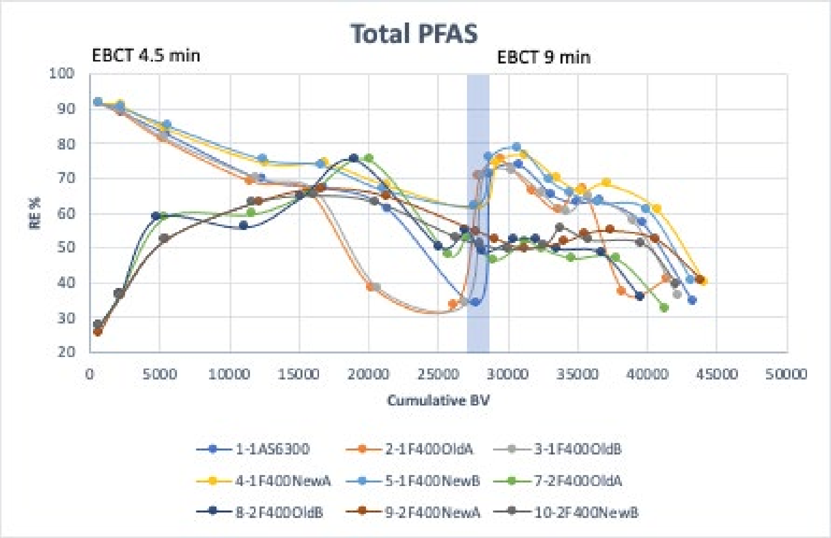


Figure 3. Removal efficiency total PFAS

Figures 4 and 5 illustrate the difference in RE between PFASs with carboxyl (PFCA) or sulphonate (PFSA) as a functional group. The removal efficiency, with a cumulative bed volume, appears to be higher overall for PFCA compared to PFSA, which is also shown in the previous Table 5. The average removal efficiency after Stage 2 was 29% higher for total PFSA in comparison to total PFCA. Upon analyzing the PFCA and PFSA with the highest concentrations, PFSA exhibits a 42% higher average removal efficiency compared to the PFCA. The higher removal efficiency of PFSA than PFCA has been confirmed by previous research (Appleman et al. 2014; McCleaf et al. 2017).

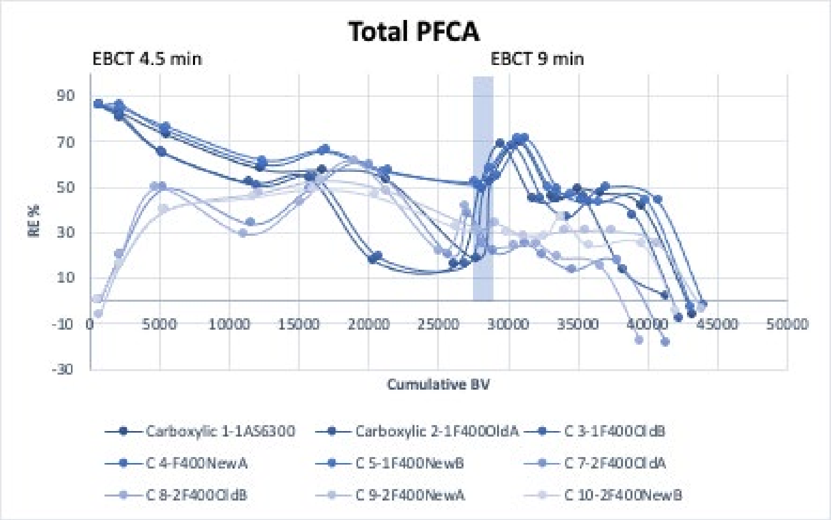


Figure 4. Removal efficiency perfluorinated carboxylic acids (PFCA)

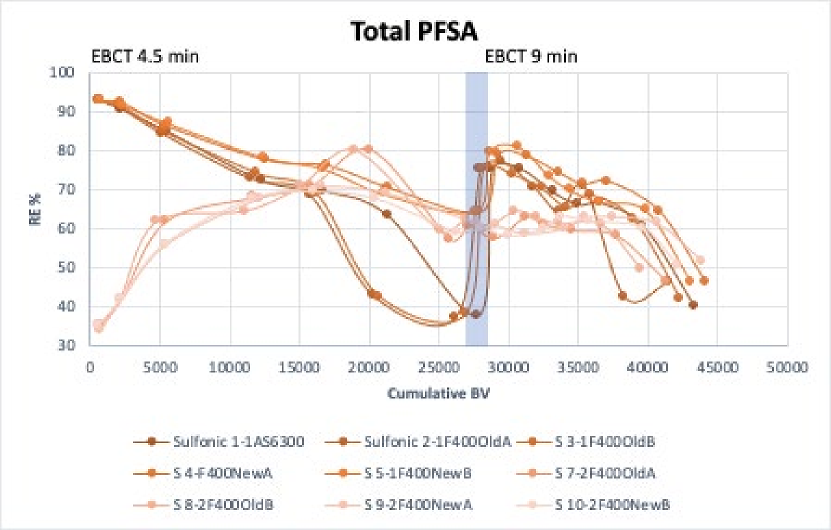


Figure 5. Removal efficiency perfluorinated sulfonic acids (PFSA)

4.1.2 Removal efficiency of PFAS 4

According to the data presented in Table 4, for the PFAS 4s the untreated water had an average concentration of PFHxS at 50 ng/L, followed by PFOS at 20 ng/L, PFOA at 4 ng/L, and PFNA below the detection limit. The overall removal efficiency of these compounds after Stage 1 was lower than that of total PFAS, as depicted in Figure 3 and 6. However, after Stage 2, the removal efficiency for PFAS 4 was higher than that for total PFAS, as shown in Figure 3 and 6. The average RE of PFAS 4 after two-stage GAC filtration was 90%, whilst the total PFAS had an average RE of 86%.

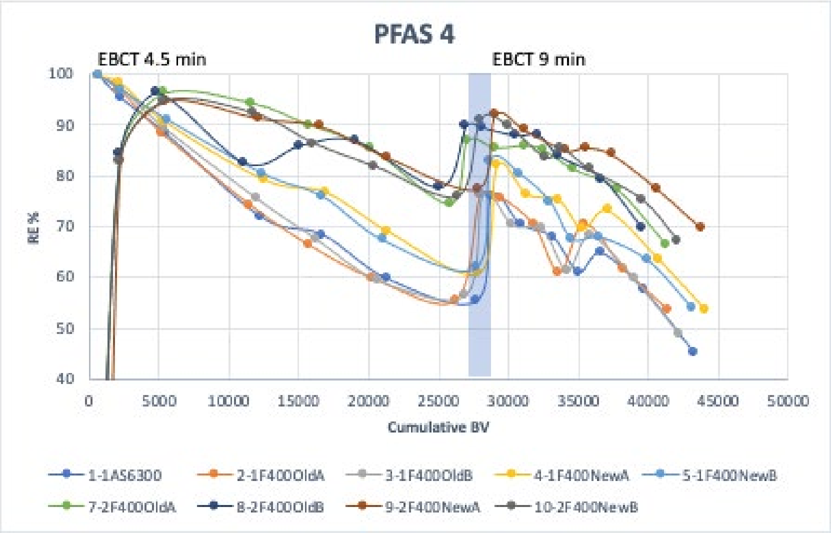


Figure 6. Removal efficiency PFAS 4

Table 6 illustrates the breakthrough point where the concentration of PFAS 4 exceeds the newly proposed limit of 4 ng/L. Based on the results presented in Table 6, it is clear that the cumulative bed volume until 4 ng/L reached is substantially lower using a single stage of GAC-filtration compared to a two-stage process. A second breakthrough point is visible after the flow rate was lowered. This only appears in Stage 2 where the second breakthrough of PFAS happened after 34 100 BV for the old GAC, respectively approximately 36 000 BV for the new GAC. The fact that the two-stage filtration had another breakthrough when the flow rate was lowered demonstrates the influence of the longer EBCT.

Table 6. PFAS 4 average concentration ng/L during cumulative BV exiting the column

STAGE 1				STAGE 2			
Average column 2 and 3^{Old}		Average column 4 and 5^{New}		Average column 7 and 8^{Old}		Average column 9 and 10^{New}	
BV	ng/L	BV	ng/L	BV	ng/L	BV	ng/L
669	0.3	691	0.30	670	0.30	680	0.50
2179	**2.4**	**2264**	**2.1**	2187	0.30	2218	0.30
5254	9.4	5513	7.8	5075	0.30	5396	0.30
11768	20	12491	16	11407	1.7	11940	1.1
15992	23	16811	16	15439	2.3	16320	2.4
20516	29	21223	23	**19564**	**3.6**	**20877**	**4.0**
26579	30	27797	26	25442	5.9	27086	6.4
EBCT change from 4.5 min to 9 min							
28069	17	28924	12	27014	2.1	28528	1.2
29886	17	31028	14	28576	2.0	30592	1.6
32096	19	33264	16	30853	2.5	33337	2.9
33895	23	34889	18	32285	2.3	**34646**	**3.0**
35611	25	36810	24	**34112**	**4.0**	36610	4.5
38624	29	40342	27	37251	5.4	40126	6.9
41838	28	43626	26	40438	6.5	42976	7.6

PFHxS

In the untreated water, PFHxS exhibited the highest concentration with an average of 50 ng/L, which was reduced to 11–14 ng/L after stage 1 and 2 ng/L after Stage 2 GAC filtration (Table 4). The removal efficiency for PFHxS, as depicted in Figure 7, resembles the pattern observed for PFAS 4 in Figure 6. The average RE of PFHxS was 95 % and PFAS 4 has an average of 96 %.

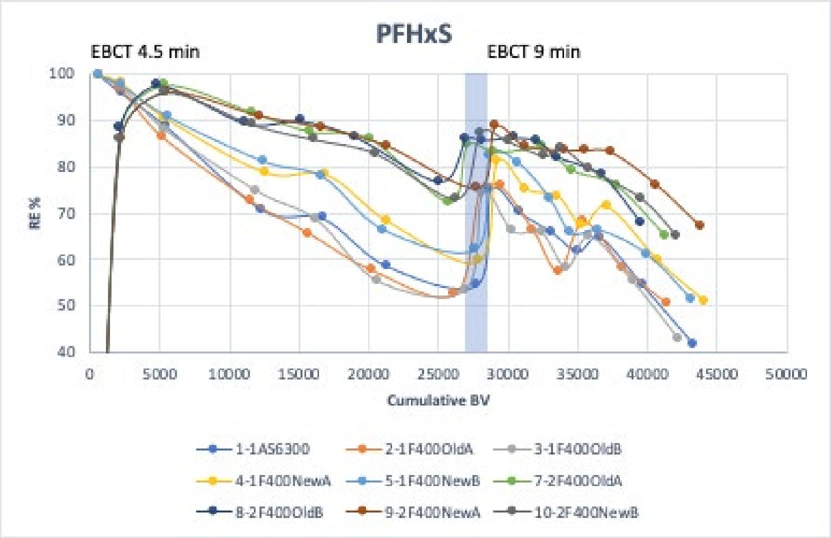


Figure 7. Removal efficiency PFHxS

PFOS

The removal efficiency of PFOS appears to be more stable than that of PFHxS, with a decrease between 7–20% depending on the column around 25 000–27 000 BV, as illustrated in Figure 8. Notably, as depicted in Figure 8, Column 8 in black diverges from its duplicate Column 7 at a bed volume of 11 129. A possible reason for divergence could be the higher flow rate, by 2 mL/min, resulting in a lower RE %. The water height was 5 cm lower on this sample date when approximately 11 100 BV had been treated, probably a result of the increased flow rate. The duplicate column did not have this change, explaining why only Column 8 was diverted.

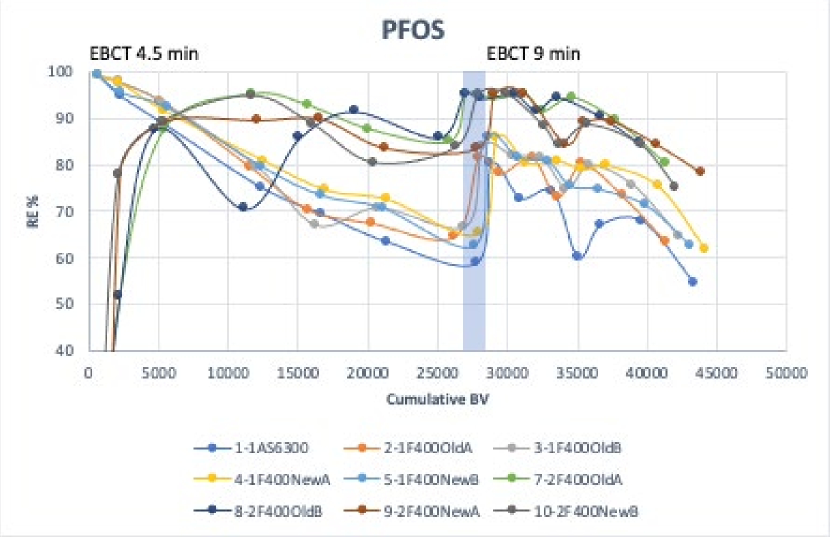


Figure 8. Removal efficiency PFOS

PFOA

Based on Table 4, the average concentration of PFOA in untreated water was 4 ng/L, which was reduced to 1 ng/L after Stage 1 and 0 ng/L after Stage 2. As depicted in Figure 8, the RE % of PFOA appears to be more scattered than that of PFHxS and PFAS 4. Notably, Column 8 in black diverges also PFOA from its duplicate Column 7 at two data points between bed volume 11 000–15 000, as shown in Figure 9.

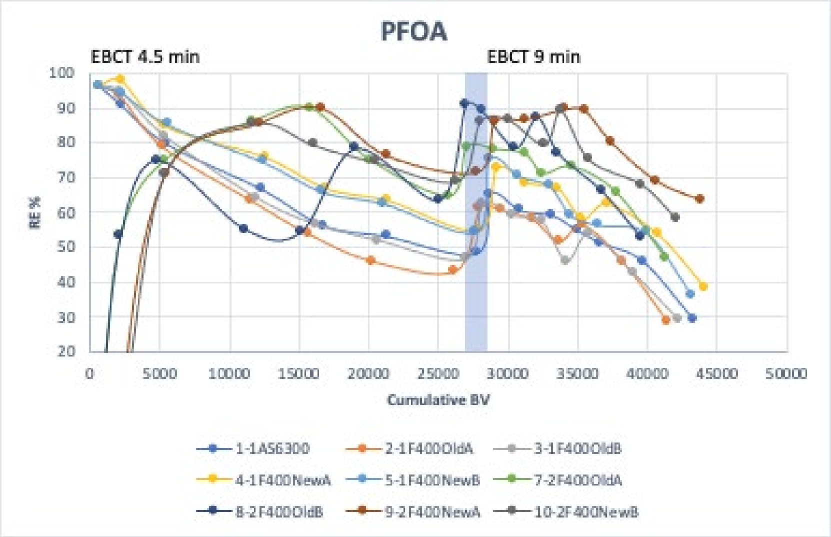


Figure 9. Removal efficiency PFOA

4.1.3 Removal efficiency of PFAS 11

The average removal efficiency of PFAS 11 after two-stage GAC filtration was 90 % compared to PFAS 4's 95 % and total PFAS with an average RE of 86 %. Figure 10 illustrates the RE during cumulative bed volumes and the graph resembles that of PFAS 4, except for Column 8 (in dark blue)), deviated along with its duplicate in green (Column 7). The graphs also differ after the flow rate changed with the RE % for Stage 2 peaking around 70 RE% for PFAS 11. In comparison to PFAS 4 around 90 RE % after the flow rate change for Stage 2.

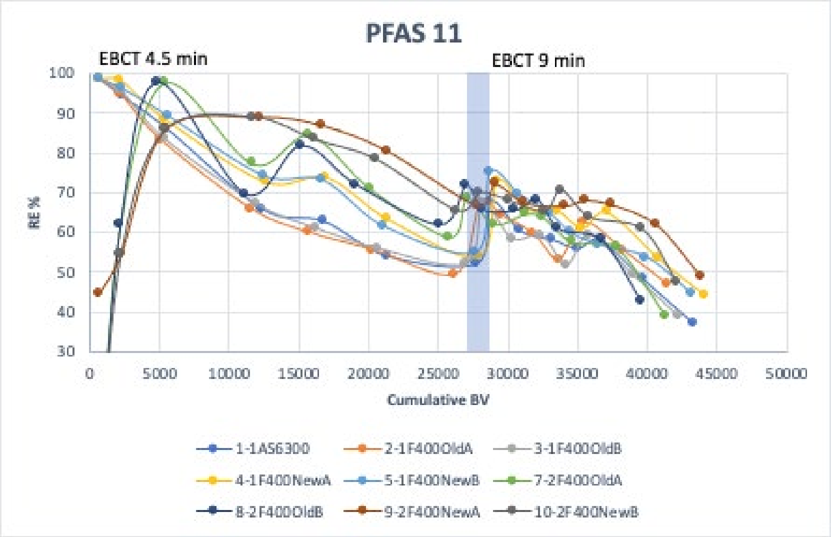


Figure 10. Removal efficiency PFAS 11

The PFAS 11 compounds with the highest concentration in untreated water were PFHxS and PFOS, as illustrated in Table 7, with concentrations of 48 ng/L and 19 ng/L, respectively. These compounds are also included in the group PFAS 4.

According to Table 7, the PFAS 11 compounds, which are not included in PFAS 4, with the highest concentration in untreated water were PFBS (6.0 ng/L), PFHxA (4.9 ng/L) and PFBA (3.8 ng/L). The removal efficiency of PFBS are shown in Figure 19 in Appendix 3.

Table 7. Average concentration PFAS 11 ng/L per column from 0 to 40000 BV (n=14).

	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFBS	PFHxS	PFOS	6:2 FTSA
12-Tank1Raw	3.8	1.9	4.9	1.1	3.9	ND	ND	6.0	48	19	ND
1-1AS6300	2.4	1.3	2.7	0.60	1.4	ND	ND	2.4	14	4.9	ND
2-1F400OldA	2.6	1.4	3.1	0.60	1.4	ND	ND	2.7	14	3.9	ND
3-1F400OldB	2.7	1.5	3.2	0.60	1.4	ND	ND	2.8	14	3.7	ND
4-1F400NewA	2.2	1.3	2.6	0.50	1.1	ND	ND	2.3	11	3.7	ND
5-1F400NewB	2.2	1.2	2.6	0.50	1.1	ND	ND	2.4	11	3.9	ND
6-1Blank	3.7	1.9	5.1	1.2	4.0	ND	ND	5.9	50	20	ND
13-Tank2BOld	2.5	1.4	3.0	0.60	1.3	ND	ND	2.7	12	3.0	ND
14-Tank2BNew	2.1	1.3	2.7	0.50	1.2	ND	ND	2.4	12	4.0	ND
7-2F400OldA	2.3	1.2	2.0	0.20	0.40	ND	ND	1.1	2.3	0.30	ND
8-2F400OldB	2.2	1.2	1.9	0.20	0.40	ND	ND	1.0	2.1	0.40	ND
9-2F400NewA	2.0	0.9	1.4	0.20	0.30	ND	ND	0.90	2.2	0.60	ND
10-2F400NewB	2.0	1.0	1.5	0.20	0.30	ND	ND	0.90	2.4	0.60	ND
11-2Blank	2.3	1.3	2.7	0.50	1.2	ND	ND	2.4	13	4.0	ND
15-Tank3	2.1	1.1	1.6	0.20	0.20	ND	ND	0.90	1.6	0.30	ND

4.1.4 Removal efficiency of PFAS 20 and 21

The removal efficiency (RE) of PFAS 20 and 21 are very similar to each other, average RE after two-stage filtration was 90 % for both groups. PFAS 20 and 21 have higher RE during Stage 2 compared to Total PFAS (Figure 3). Total PFAS had an average RE after two-stage filtration of 86% in comparision. PFAS 20 and 21 had similar RE compared to PFAS 11 but lower compared to PFAS 4 with an average RE after Stage 2 of 96%.

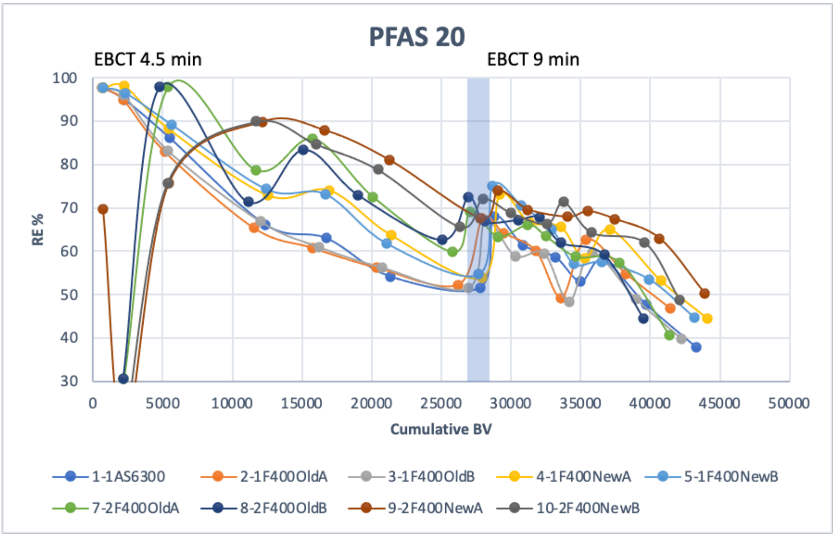


Figure 11. Removal efficiency PFAS 20

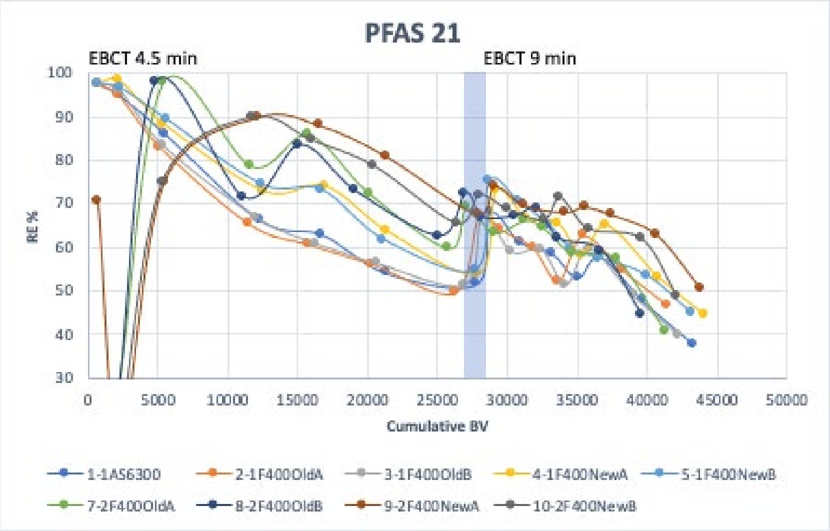


Figure 12. Removal efficiency PFAS 21

4.2 Differences between new and old GAC

In each stage, there were two columns with new GAC and two with "old" GAC taken from the fullscale GAC filter in the DWTP Bäcklösa. As was portrayed in Table 4, in treated water the average concentration of PFAS 4, 11, 20, 21 and Total PFAS per column differed between the columns with old GAC and new. They differed slightly more in Stage 1 than 2. For example, the average concentration of Total PFAS after the old GAC in Stage 1 was 47 ng/L compared to the average after filtration with the new GAC in Stage 1 36.5 ng/L. In Stage 2, the average concentration of Total PFAS after old GAC filtration was 19.5 ng/L and for new GAC 18.5 ng/L. Regarding differences in removal efficiency after two stages between the GACs, the new had 1% higher RE of total PFAS, 2%

higher for PFAS 11 and 5% higher for PFCA. The new and old GAC had similar RE % of PFAS 4 and PFSA. Furhtermore, the RE % from the average concentrations of total PFAS for Stage 1 also suggests that the new GAC is more efficient than the old GAC, as depicted in Figure 13. The old GAC demonstrates two significant decreases in RE, while the new GAC has a flatter line. In Stage 2, as illustrated in Figure 14, the new and old GAC exhibit very similar RE through cumulative BV.

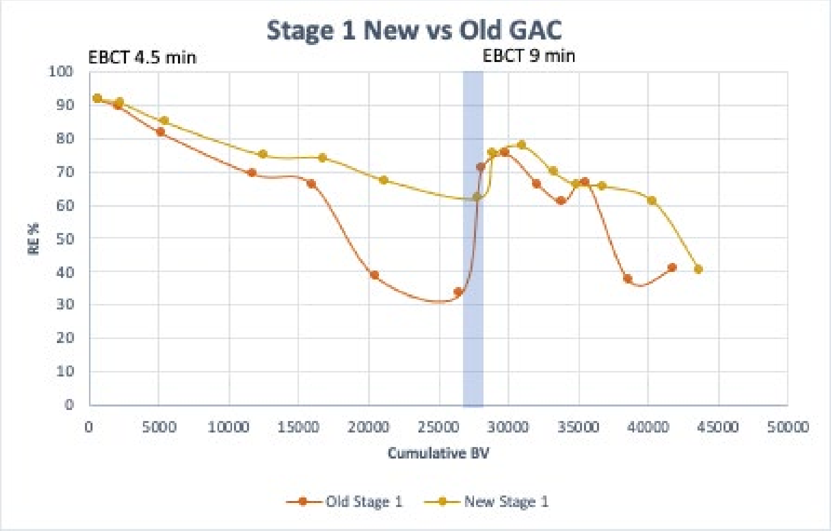


Figure 13. Comparison of removal efficiency between new and old GAC for Stage 1.

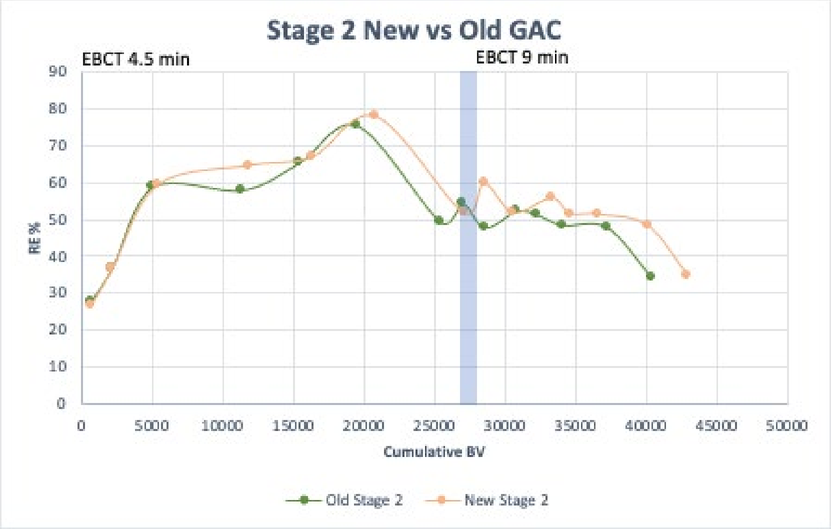
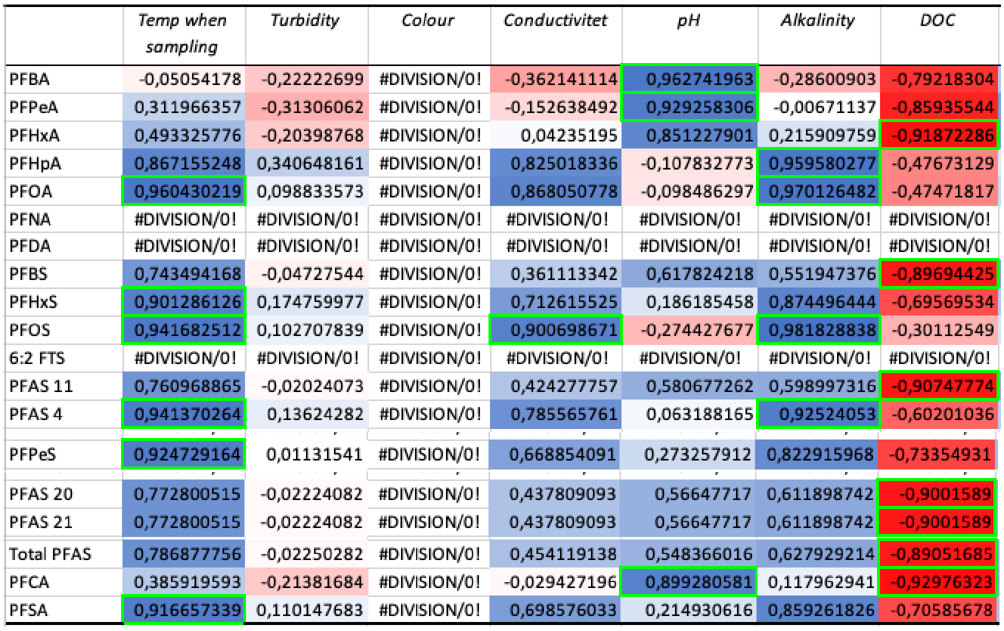


Figure 14. Comparison of removal efficiency between new and old GAC for Stage 2.

4.3 Statistic correlation

Pearson’s correlation coefficient along with t-statistic and p-value was used to determine any statistical correlation between the removal efficiency (RE %) of the analysed PFASs and chemical characteristics in the incoming water. The chemical characteristics that varied the most in the five samples that were included in the correlation, were alkalinity and uranium. Figure 20 in Appendix 4 illustrates the entirety of the correlation landscape whilst Figure 15 shows the selection of the significant ones. A significant positive correlation can be made between pH and PFASs PFBA (C3), PFPeA (C4) and PFCA (where both PFBA and PFPeA are included), as illustrated in Figure 15. In other words, as the pH increases, so does the RE for these particular compounds. Another significant correlation could be made between increasing temperature and increasing removal efficiency of PFOA (C7), PFHxS (C5), PFOS (C8), PFAS 4 (where the three previous PFASs are included) and PFPeS (C5). Significant positive correlations could also be made between conductivity and RE of PFOA (C7), as well as alkalinity and RE of PFHpA (C6), PFOA (C7), PFOS (C8) and PFAS 4, as presented in Figure 15. Furthermore, a significant negative correlation was found between the concentration of DOC (dissolved organic carbon) and the removal efficiency of PFHxA (C5), PFBS (C4), PFAS 11, 20, 21, Total PFAS and PFCA (Figure 15). Previous research indicates a possibly reduced capacity of GAC to adsorb PFAS, especially short-chained PFASs, when DOC is present in the water (Rahman et al. 2014; McCleaf et al. 2017). There is not enough research on the correlation between water's chemical substances and the removal of PFAS by GAC filtration to draw a general conclusion.



	Temp when sampling	*Turbidity*	*Colour*	*Conductivitet*	*pH*	*Alkalinity*	*DOC*
PFBA	-0,05054178	-0,22222699	#DIVISION/0!	-0,362141114	0,962741963	-0,28600903	-0,79218304
PFPeA	0,311966357	-0,31306062	#DIVISION/0!	-0,152638492	0,929258306	-0,00671137	-0,85935544
PFHxA	0,493325776	-0,20398768	#DIVISION/0!	0,04235195	0,851227901	0,215909759	-0,91872286
PFHpA	0,867155248	0,340648161	#DIVISION/0!	0,825018336	-0,107832773	0,959580277	-0,47673129
PFOA	0,960430219	0,098833573	#DIVISION/0!	0,868050778	-0,098486297	0,970126482	-0,47471817
PFNA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFBS	0,743494168	-0,04727544	#DIVISION/0!	0,361113342	0,617824218	0,551947376	-0,89694425
PFHxS	0,901286126	0,174759977	#DIVISION/0!	0,712615525	0,186185458	0,874496444	-0,69569534
PFOS	0,941682512	0,102707839	#DIVISION/0!	0,900698671	-0,274427677	0,981828838	-0,30112549
6:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFAS 11	0,760968865	-0,02024073	#DIVISION/0!	0,424277757	0,580677262	0,598997316	-0,90747774
PFAS 4	0,941370264	0,13624282	#DIVISION/0!	0,785565761	0,063188165	0,92524053	-0,60201036
PFPeS	0,924729164	0,01131541	#DIVISION/0!	0,668854091	0,273257912	0,822915968	-0,73354931
PFAS 20	0,772800515	-0,02224082	#DIVISION/0!	0,437809093	0,56647717	0,611898742	-0,9001589
PFAS 21	0,772800515	-0,02224082	#DIVISION/0!	0,437809093	0,56647717	0,611898742	-0,9001589
Total PFAS	0,786877756	-0,02250282	#DIVISION/0!	0,454119138	0,548366016	0,627929214	-0,89051685
PFCA	0,385919593	-0,21381684	#DIVISION/0!	-0,029427196	0,899280581	0,117962941	-0,92976323
PFSA	0,916657339	0,110147683	#DIVISION/0!	0,698576033	0,214930616	0,859261826	-0,70585678

Figure 15. Partial statistical correlation between PFASs and chemical characteristics where the colour scale blue to red illustrates if the correlation is positive, neutral or negative. The green border symbolises if the p-value was below 0.05, thus significant. The entirety of the correlation landscape can be seen in Figure 20, Appendix 4.

Discussion

This study aimed to evaluate the removal efficiency of PFAS with two-stage GAC filtration and determine the number of BV before individual PFAS breakthroughs and how the flow rate (EBCT) affects the time until GAC regeneration. The hypothesis of this study was that a two-stage GAC filtration will enhance removal efficiency and extend the time until GAC regeneration, compared to a single-stage GAC filtration. The assumption is that a two-stage process will approximately double the number of bed volumes before PFAS 4 breakthrough. As well as a reduction in flow rate will lead to higher removal efficiency by extending the empty bed contact time (EBCT), ultimately prolonging the time until breakthrough for both stages due to increased adsorption. The results from the pilot experiment showed an overall higher removal efficiency (RE %) with a two-stage GAC filtration compared to a single-stage process. The long-chained PFASs appear to have a higher level of adsorption than the short-chained ones, which has been confirmed by previous research (Brendel et al. 2018; Li et al. 2020). Additionally, the results indicated overall better removal of the PFAS classed as PFSA compared to the PFCA, which also has been confirmed by previous research (Appleman et al. 2014; McCleaf et al. 2017). To determine the difference in the removal of PFAS 4 between a one and two-stage system, values from Table 6 around 4 ng/L were first interpolated. To be able to compare the two systems, the bed volumes from the interpolation for Stage 1 were doubled. This since the hypothetical one-filter system needs to have the same amount of GAC as the two filters of the experiment, along with the assumption that the two stages have the same amount of GAC. Based on that assumption, the bed volumes for Columns 2 and 3 in Stage 1 are 5763 BV when the concentration of PFAS 4 reaches 4 ng/L, and 6693 BV for Columns 4 and 5. The average BV for Stage 2 Columns 9 and 10 was 20 876 and after the interpolation, the bed volume for Columns 7 and 8 was 20 586 BV. The two-stage filtration managed to treat approximately three and a half times as many bed volumes of water compared to single-stage filtration with the same amount of GAC. Which exceeded the hypothesis assumption of approximately double the amount of bed volumes. Observing the second breakthrough of PFAS 4 after the flow rate change in Stage 2 confirms that longer EBCTs result in greater amounts of water being treated until the PFAS 4 concentration limit of 4 ng/L is reached. The lower removal efficiency observed when the flow rate changed and the EBCT got longer is not relevant to the question of how EBCT affects removal efficiency, as the conditions for the two EBCTs were different. The conclusion that can be drawn from these results is that more bed volumes of water can be treated before the guideline limit of PFAS 4 has been reached at a lower flow rate. To improve the pilot experiment, the columns could have a consistent EBCT for the entire time period or change the flow rate after resetting it. This would allow for a more clear comparison of the effect of EBCT on the removal efficiency of PFASs.

Moreover, for the two-staged filtration system, the columns with new GAC treated 1.4 % more BV than the “old” GAC until breakthrough to 4 ng/L PFAS 4 was reached. In a single-stage filtration after Stage 1, was a difference of 1.6 % more BV in favour of the new GAC. The new GAC seems to also have a higher removal efficiency of Total PFAS than the “old” GAC during the first stage of filtration. The two had similar removal efficiency during the second stage for Total PFAS. These results are specific to Bäcklösa groundwater during this time period and PFAS concentration and removal efficiency may look different for different groundwater.

The statistical correlation made between the removal efficiency of individual PFAS and the chemical characteristics of the water showed positive correlations of some of the individual PFAS with temperature, conductivity, pH, and alkalinity, and a negative correlation with DOC. Considering the relatively limited sample size and the lack of prior research on these correlations, a well-supported conclusion cannot be drawn.

The results may also have been affected by the measurement uncertainty at the commercial lab doing the analysis varied between compounds and dates. The measurement uncertainty (MU) for PFPeA, PFHpA and PFOA often demonstrated the least range at approximately +/– 0.0006 µg/L. In contrast, PFAS 4 and 11 typically exhibit a MU of +/– 0.010 µg/L. PFAS 20 and 21 displayed more variation, with +/– 0.02 µg/L in some samples and +/– 0.010 µg/L in others. Given that the MU associated with PFAS 4 was +/– 10 ng/L and the regulatory limit of 4 ng/L, the results should be interpreted with this fact in mind. The lab’s limit of reporting, LOR, were specific for each PFAS compound. For example, the PFAS 4’ compound PFNA had a LOR of 0.00030 µg/L and when PFNA had a concentration below detection, the results stated it as “<0.00030 µg/L”. When Total PFAS was calculated, the limit of reporting was taken into account by halving the LOR concentration. This means that the concentration of total PFAS in this study may be shown as slightly higher than in reality. The removal efficiency should be aligned with reality since this also affected the calculation of Total PFAS in the incoming water.

Some data points deviated excessively from the adjacent data points, resulting in some of the supplementary samples being sent for analysis. The additional results were consistent with the rest of the data and the deviated data points were excluded. The reason for deviated data could be shifting in the natural compound composition of water making the levels of PFAS higher than usual at times. Another reason could be competition between PFASs to bind to GAC or less removal efficiency due to varying flowrate. The latter reason is probably less likely to the monitoring twice a week. In total three dates of results were excluded.

Franke et al. (2021) evaluated the cost-effectiveness of a combined treatment approach using nanofiltration, single-stage GAC, and anion exchange resins for removing PFASs from drinking water. Given the conclusion of this study regarding the effectiveness of a two-stage GAC filtration system, it would be interesting to further evaluate the economic perspective of a two-stage system as well.

Conclusion

The results of this thesis showed that a two-stage process is more effective in removing PFAS and extending the time until GAC regeneration compared to a single-stage process. The study also found that long-chained PFASs have a higher level of adsorption than short-chained ones, making it coherent with previous research. The study estimated that a two-staged GAC filtration can treat approximately three and a half times as many bed volumes as a single stage before the limit of PFAS 4, 4 ng/L were reached (with an EBCT of 4.5 min). The results showed a slightly higher removal capacity of the new Filtrasorb ® 400-E GAC compared to the regenerated "old" Filtrasorb ® 400-E GAC. The thesis also concluded that a lower flow rate (higher EBCT) resulted in a longer time until GAC regeneration. It is important to note that these results are specific to Bäcklösa groundwater during this time period and PFAS concentration and removal efficiency may look different for different groundwater.

This study provides valuable insights into the effectiveness of different GAC filtration systems for PFAS removal and highlights the need for further investigation into the economic aspects of these systems to fully evaluate two-stage GAC filtering in a drinking water treatment plant. The evaluation of removal techniques in drinking water production contributes to protecting human health from harmful water pollution.

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Populärvetenskaplig sammanfattning

Det här masterarbetet har kommit fram till att ett tvåstegs filtreringsystem med granulerat aktivt kol är mer effektivt i att ta bort PFAS ur dricksvatten än ett enstegssystem, som vanligtvis används i vattenverk.

PFAS är en grupp kemikalier skapade av människan som är kända för att vara otroligt stabila ämnen och därför använts i en rad olika industri och konsumentprodukter, bland annat brandsläckningsskum. PFAS ämnen bryts inte ner i naturen och det har dessutom visat sig att flera PFAS ämnen ackumuleras i bland annat människor, vilket kan leda till hälsorisker. Fler och fler studier ägnar sig åt att reda ut hur detta mer exakt påverkar människors hälsa. Exponering av PFAS kommer mestadels i från det vi konsumerar, däribland dricksvatten. Diskussionen hur vi som samhälle ska hantera PFAS ämnena är mycket aktuell med den senaste lagstiftningen för dricksvatten i Sverige trädde i kraft januari 2023 och berör flera av FN:s hållbarhetsmål i Agenda 2030. Med stramare lagstiftning om gränsvärden för PFAS i dricksvatten, behövs utvecklade reningstekniker för vattenverken.

Flera studier har utvärderat befintliga reningstekniker och även kombinationer av dem men hittills är det ingen som utvärderat kolfiltrering i två steg. Den här studien har tillsammans med Uppsala Vatten och Avfall AB skapat ett experiment att utvärdera just det i Bäcklösa vattenverk. Experimentet består av stora plaströr med granulerat aktivt kol i botten, samt några utan som kontroller. Plaströren är indelade i två steg där vattnet pumpas från kolfiltren i första steget vidare till de i andra steget, vattnet behandlas alltså av två kolfilter. Vattenprover tas efter båda stegen och skickas iväg till ett kommerisellt lab för analys. Dessutom används olika typer av granulerat aktivt kol, helt nytt och “gammalt” så kallad regenererat med värme för att återaktivera kolet igen. Experimentet började behandla vatten i juni 2022 och resultaten fram till januari 2023 är inkluderade i den här uppsatsen.

Förutom slutsatsen att en tvåstegs kolfiltrering verkar ta bort PFAS ur vattnet mer effektivt, verkar ett tvåstegssystem även kunna behandla mer vatten innan regenerering av kolet behövs än ett system med enbart ett kolfilter. Ytterligare slutsatser som har kunnat dras är att långkedjade PFAS lättare tas bort än kortkedjade och att en lägre flödeshastighet genom filtret ökar borttagningseffektiviten. Det är viktigt att notera att dessa resultat är unika till Bäcklösas grundvatten och att effektiviteten kan se annorlunda ut på andra platser. För att komplettera denna utvärdering skulle exempelvis en ekonomisk analys kunna göras för ett tvåstegssystem av kolfiltrering jämfört med andra reningstekniker.

Acknowledgements

I want to thank my enthusiastic supervisors Lutz and Philip for the opportunity to be a part of their ongoing project with SLU and Uppsala Water and Waste Ltd regarding PFAS and removal techniques. Thank you for your expertise and support during these months. Learning more about drinking water production and being part of a larger project that contributes to cleaner water has been very interesting and inspiring.

I am grateful for my family and friends who supported and encouraged me, especially during the tough times when I underwent knee surgery in December and the following rehabilitation. I could not have finished this thesis without them. An extra thanks to my study buddies, Sandra and Lisa, who could brighten any day.

Appendix 1: Experiment set-up

The following pictures depitets the set-up in the pilot experiment at Bäcklösa DWTP in Uppsala. In Figure 16, Column number 1 (sample point 1) is to the far left and Tank 3 (sample point 15) with outgoing water in white, is to the far right. Stage 1 is the six columns to the left and Stage 2 is the six columns to the right of the blank tanks in the middle (sample points 13 and 14 aka Tanks 2AOld and 2BNew).

Figure 16. Picture 1 of the expermental set-up showing the plastic columns with Stage 1 to the left and Stage 2 to the right.

In Figure 17, sample point 12 with untreated water, can be seen furthest in the corner. Sample points 13 and 14 are the two black tanks in the middle with Stage 1 to the left and Stage 2 to the right.

Figure 17. Picture 2 of the experimental set-up.

Closer picture of the foundation of the columns in stage 1 and their GAC filters (Figure 18).

Figure 18. Picture 3 of the experimental set-up showing the bottom of the plastic columns of Stage 2.

Appendix 2: Water quality

Average	Temp when sampling (°C)	Turbidity (FNU)	Colour (mg/L)	Conductivitet (mS/m)	pH	Alkalinity (mg/L)	DOC (mg/L)
12-Tank1Raw	10,34	0,135	2,6	45,16	8,22	95,32	2,28
13-Tank2AOld and 14-Tank2BNew	11,6	0,0615	2	45,35	8,05	97,18	2,02
15-Tank3	12,2	0,095	2	45,6	8,04	97	1,86
	Hardness (total °dH)	Ammonium, NH4 (mg/L)	Sodium, Na (mg/L)	Magnesium, Mg (mg/L)	Nitrite, NO2 (mg/L)	Aluminum, Al (mg/L)	Fluoride, F (mg/L)
12-Tank1Raw	8,08	0,035	29,4	16,2	0,005	0,04	0,986
13-Tank2AOld and 14-Tank2BNew	8,13	0,035	29,7	16,2	0,005	0,0365	0,987
15-Tank3	8,14	0,035	29,8	16,2	0,005	0,0346	0,988
	Calcium, Ca (mg/L)	Chloride, Cl (mg/L)	Manganese, Mn (mg/L)	Nitrate, NO3 (mg/L)	Iron, Fe (mg/L)	Sulphate, SO4 (mg/L)	Copper, Cu (mg/L)
12-Tank1Raw	31,2	58,68	0,001	3,74	0,0025	41	0,0025
13-Tank2AOld and 14-Tank2BNew	31,5	58,52	0,001	3,775	0,0025	41	0,0025
15-Tank3	31,6	58,58	0,001	3,775	0,0025	41	0,0025
	Uranium, U (µg/L)	UV-254nm	TOC (mg/L)	Potassium, K (mg/L)			
12-Tank1Raw	27,2	0,032	2,3	8,25			
13-Tank2AOld and 14-Tank2BNew	26,2	0,0235	1,9	8,2375			
15-Tank3	24,6	0,014	1,5	8,2			

Table 8. Averages of 25 chemical characteristics indicating water quality at sample points 12-15.

Appendix 3: Removal efficiency of PFBS (included in PFAS 11)

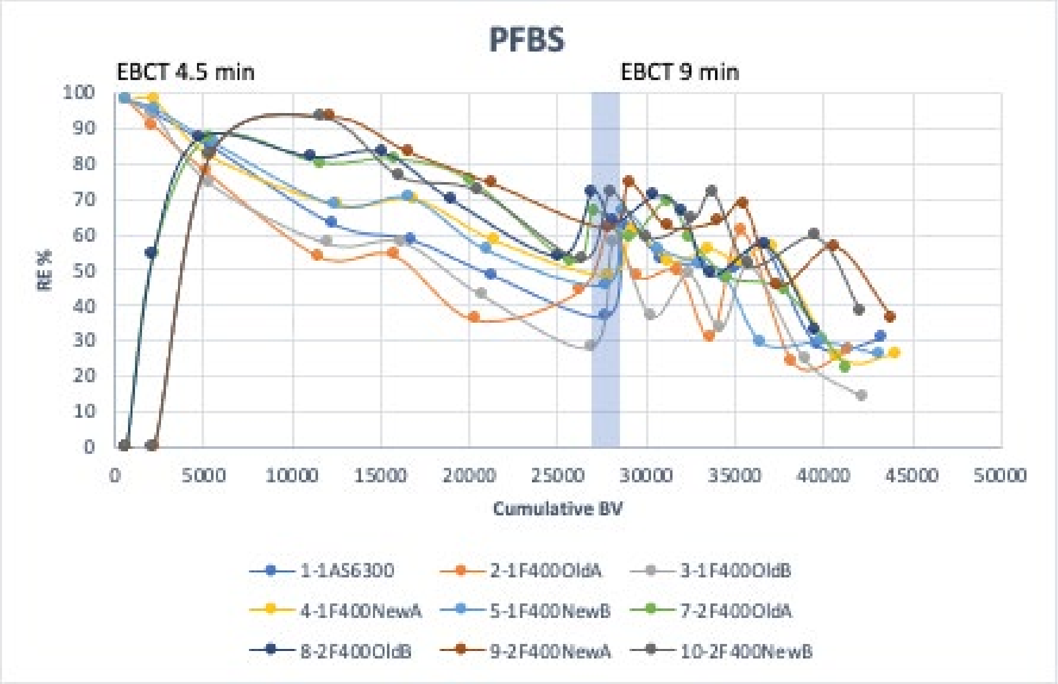

Figure 19. Removal efficiency of PFBS (included in PFAS 11).

Appendix 4: Statistical correlation

	Temp when sampling	Turbidity	Colour	Conductivitet	pH	Alkalinity	DOC	Hardness	Ammonium, NH4	Sodium, Na	Magnesium, Mg
PFBA	-0,05054178	-0,22222699	#DIVISION/0!	-0,362141114	0,962741963	-0,28600903	-0,79218304	0,410466882	#DIVISION/0!	-0,57106988	#DIVISION/0!
PFPeA	0,311966357	-0,31306062	#DIVISION/0!	-0,152638492	0,929258306	-0,00671137	-0,85935544	0,701696334	#DIVISION/0!	-0,58533514	#DIVISION/0!
PFHxA	0,493325776	-0,20398768	#DIVISION/0!	0,04235195	0,851227901	0,215909759	-0,91872286	0,707281698	#DIVISION/0!	-0,63430858	#DIVISION/0!
PFHpA	0,867155248	0,340648161	#DIVISION/0!	0,825018336	-0,107832773	0,959580277	-0,47673129	0,178145309	#DIVISION/0!	-0,40824829	#DIVISION/0!
PFOA	0,960430219	0,098833573	#DIVISION/0!	0,868050778	-0,098486297	0,970126482	-0,47471817	0,274079516	#DIVISION/0!	-0,49585614	#DIVISION/0!
PFNA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFBS	0,743494168	-0,04727544	#DIVISION/0!	0,361113342	0,617824218	0,551947376	-0,89694425	0,646856099	#DIVISION/0!	-0,64771997	#DIVISION/0!
PFHxS	0,901286126	0,174759977	#DIVISION/0!	0,712615525	0,186185458	0,874496444	-0,69569534	0,392736797	#DIVISION/0!	-0,56443692	#DIVISION/0!
PFOS	0,941682512	0,102707839	#DIVISION/0!	0,900698671	-0,274427677	0,981828838	-0,30112549	0,205043465	#DIVISION/0!	-0,37088104	#DIVISION/0!
6:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFAS 11	0,760968865	-0,02024073	#DIVISION/0!	0,424277757	0,580677262	0,598997316	-0,90747774	0,579874935	#DIVISION/0!	-0,69646064	#DIVISION/0!
PFAS 4	0,941370264	0,13624282	#DIVISION/0!	0,785565761	0,063188165	0,92524053	-0,60201036	0,357164739	#DIVISION/0!	-0,53274968	#DIVISION/0!
PFUnDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDoDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
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PFPeS	0,924729164	0,01131541	#DIVISION/0!	0,668854091	0,273257912	0,822915968	-0,73354931	0,49810295	#DIVISION/0!	-0,62037608	#DIVISION/0!
PFHpS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFNS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
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PFUnDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
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PFTrDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFAS 20	0,772800515	-0,02224082	#DIVISION/0!	0,437809093	0,56647717	0,611898742	-0,9001589	0,579917314	#DIVISION/0!	-0,69404623	#DIVISION/0!
PFAS 21	0,772800515	-0,02224082	#DIVISION/0!	0,437809093	0,56647717	0,611898742	-0,9001589	0,579917314	#DIVISION/0!	-0,69404623	#DIVISION/0!
4:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
8:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
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Total PFAS	0,786877756	-0,02250282	#DIVISION/0!	0,454119138	0,548366016	0,627929214	-0,89051685	0,579682093	#DIVISION/0!	-0,68934239	#DIVISION/0!
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PFSA	0,916657339	0,110147683	#DIVISION/0!	0,698576033	0,214930616	0,859261826	-0,70585678	0,440078723	#DIVISION/0!	-0,57971117	#DIVISION/0!

	Nitrite, NO2	Aluminum, Al	Fluoride, F	Calcium, Ca	Chloride, Cl	Manganese, Mn	Nitrate, NO3	Iron, Fe	Sulphate, SO4	Copper, Cu	Uranium, U	UV-254nm	TOC	Potassium, K
PFBA	#DIVISION/0!	0,838123065	0,769763225	0,445896986	-0,69215686	#DIVISION/0!	-0,41908819	#DIVISION/0!	-0,4451452	#DIVISION/0!	0,61557848	#DIVISION/0!	#DIVISION/0!	-0,79669367
PFPeA	#DIVISION/0!	0,681274954	0,811603792	0,715342829	-0,41294811	#DIVISION/0!	-0,33886433	#DIVISION/0!	-0,35598281	#DIVISION/0!	0,382081272	#DIVISION/0!	#DIVISION/0!	-0,45414403
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PFHpA	#DIVISION/0!	-0,04199194	-0,32936831	0,097128586	0,761736809	#DIVISION/0!	0,776749826	#DIVISION/0!	0,718421208	#DIVISION/0!	0,029285371	#DIVISION/0!	#DIVISION/0!	0,474685037
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PFHxS	#DIVISION/0!	0,151349186	-0,0403526	0,336269775	0,556464123	#DIVISION/0!	0,579929715	#DIVISION/0!	0,51643709	#DIVISION/0!	0,116761895	#DIVISION/0!	#DIVISION/0!	0,385384886
PFOS	#DIVISION/0!	-0,3155912	-0,37448554	0,206335107	0,780343268	#DIVISION/0!	0,65691172	#DIVISION/0!	0,57369022	#DIVISION/0!	-0,26206235	#DIVISION/0!	#DIVISION/0!	0,316109834
6:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFAS 11	#DIVISION/0!	0,450271158	0,350214038	0,553726558	0,166758654	#DIVISION/0!	0,246060227	#DIVISION/0!	0,187383931	#DIVISION/0!	0,292984319	#DIVISION/0!	#DIVISION/0!	0,200112414
PFAS 4	#DIVISION/0!	0,015554242	-0,12787046	0,323387106	0,623529498	#DIVISION/0!	0,600904472	#DIVISION/0!	0,52718774	#DIVISION/0!	0,000543478	#DIVISION/0!	#DIVISION/0!	0,344681681
PFUnDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDoDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFTrDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFPeS	#DIVISION/0!	0,154834137	0,087502842	0,482576451	0,449043588	#DIVISION/0!	0,432450153	#DIVISION/0!	0,358886499	#DIVISION/0!	0,06345913	#DIVISION/0!	#DIVISION/0!	0,257742731
PFHpS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFNS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFUnDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFDoDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFTrDS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFAS 20	#DIVISION/0!	0,434234025	0,339461058	0,554902521	0,181408511	#DIVISION/0!	0,253690056	#DIVISION/0!	0,194087962	#DIVISION/0!	0,278696318	#DIVISION/0!	#DIVISION/0!	0,201641749
PFAS 21	#DIVISION/0!	0,434234025	0,339461058	0,554902521	0,181408511	#DIVISION/0!	0,253690056	#DIVISION/0!	0,194087962	#DIVISION/0!	0,278696318	#DIVISION/0!	#DIVISION/0!	0,201641749
4:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
8:2 FTS	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
FOSA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
MeFOSA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
EtFOSA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
MeFOSE	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
EtFOSE	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
FOSAA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
MeFOSAA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
EtFOSAA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
HPFHpA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PF37DMOA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
PFTeDA	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!	#DIVISION/0!
Total PFAS	#DIVISION/0!	0,414672447	0,325496396	0,555088703	0,201045319	#DIVISION/0!	0,264531378	#DIVISION/0!	0,204282252	#DIVISION/0!	0,261573647	#DIVISION/0!	#DIVISION/0!	0,207373178
PFCA	#DIVISION/0!	0,706380702	0,710731302	0,642217798	-0,33266628	#DIVISION/0!	-0,19168465	#DIVISION/0!	-0,22743132	#DIVISION/0!	0,452758864	#DIVISION/0!	#DIVISION/0!	-0,4545204
PFSA	#DIVISION/0!	0,144811848	0,008591748	0,397843856	0,524214299	#DIVISION/0!	0,524304482	#DIVISION/0!	0,458286282	#DIVISION/0!	0,086520548	#DIVISION/0!	#DIVISION/0!	0,34378596

Figure 20. The entyrity of the Pearson correlation between PFASs and chemical characteristics where the colour scale blue to red illustrates if the correlation is positive, neutral or negative. The green border symbolises if the p-value was below 0.05 and therefore significant.

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