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Is two stage GAC better than one stage GAC for removing PFAS at a DWTP?

Investigation of PFAS removal from drinking water using two stage granular activated carbon (GAC) filter

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Abstract

The removal of 34 per- and polyfluoroalkyl substances (PFAS) were compared in a 1 stage granular activated carbon (GAC) filtration to a 2 stage GAC filtration in a pilot study at a drinking water treatment plant (DWTP). The PFASs that were present in the water were perfluorooctanoic acid (PFOA), perfluorooctane sulfonic acid (PFOS), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorobutane sulfonic acid (PFBS), perfluoropentane sulfonic acid (PFPeS) and perfluorohexane sulfonic acid (PFHxS). A cost comparison for the operation of a one stage GAC to a two stage GAC was compared for PFAS₄ (sum of PFOA, PFNA, PFHxS and PFOS) at treatment goals ranging from 2 - 10 ng/L. The pilot was operated at three different flows and the three different bed volumes (BV)s resulting in three different empty bed contact times (EBCTs) at three different times. Therefore, the Lin & Huang adsorption model (1999) was used to model the concentrations at each EBCT. It was found that the model worked good for PFBS, PFPeS, PFHxS, PFOS and PFOA but not for PFPeA, PFHxA and PFHpA (except for PFPeA and PFHxA during EBCT 5 min) and did not work for desorbing PFASs. The removal comparison of PFASs was made, partly by comparing removal efficiencies between the first stage and the second stage GAC filter and by comparing the removal per weight of GAC per BV 1 stage and 2 stages. It was found that the removal efficiency decreases with decreasing chain length and increasing treated BVs for both the first stage and the second stage. The short chain PFCAs were also desorbing after a number of BVs. The removal per weight of GAC showed that the removal does not increase when comparing a one stage GAC to a two stage GAC for any the PFAS. The cost comparison was made using the adsorption model. It showed that it was cheaper to operate a 2 stage GAC for the EBCT of 5 minutes and 8 minutes for the whole range of treatment goals. However, for the EBCT of 15 minutes the costs for the second stage was decreasing with decreasing treatment goal which is unrealistic result. This was caused by too few data points available for the model to predict reliable results.

Keywords: GAC, PFAS, drinking water treatment, two stage GAC

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Sammanfattning

Avlägsning av 34 per- och polyfluorerade ämnen (PFAS) från vatten jämfördes i 1 stegs aktivt granulärt kolfilter (GAC) mot 2 stegs GAC i en pilotanläggning på ett dricksvattenverk i Uppsala. De PFAS-ämnen som fanns i vattnet var perfluoroktansyra (PFOA), perfluoroktansulfonsyra (PFOS), perfluoropentansyra (PFPeA), perfluorhexansyra (PFHxA), perfluorheptansyra (PFHpA), perfluorbutansulfonsyra (PFBS), perfluoropentansulfonsyra (PFPeS) och perfluorhexansulfonsyra (PFHxS). En kostnadsjämförelse mellan 1 stegs och 2 stegs GAC genomfördes för PFAS₄ (summan av PFOA, PFNA, PFHxS och PFOS) för målkoncentrationer i dricksvattnet på 2-10 ng/l. På piloten användes 3 olika flöden vilket resulterade i tre olika kontakttider i bädden (EBCT) på 5, 8 och 15 minuter. Dessa EBCT var vid olika tider och därmed olika bäddvolym (BV). Därför modellerades koncentrationer med hjälp av Lin & Huang's adsorptionsmodell (1999) för att ta reda på vilka koncentrationer som hade varit om samma EBCT hade varit under hela pilotens livslängd. Det konstaterades att modellen fungerade bra för PFBS, PFPeS, PFHxS, PFOS och PFOA men inte för PFPeA, PFHxA och PFHpA (förutom PFPeA and PFHxA för EBCT 5 min) och inte heller för desorberande PFAS. Jämförelsen av avlägsning bestod dels i en jämförelse av avlägsningseffektivitet mellan första steg 1 och steg 2, samt en jämförelse av avlägsning per vikt av GAC per BV mellan 1 steg och 2 stegs GAC. Det visade sig att avlägsningseffektiviteten minskar med minskande längd på PFAS och ökande BV och att samma sak skedde för steg 1 och steg 2. De kortaste PFAS-ämnena desorberade efter ett antal BV. Jämförelsen av avlägsning per vikt av GAC visade att det inte var en mer effektiv avlägsning för två stegs jämfört med ett stegs GAC för något av PFAS-ämnena. Kostnadsjämförelsen genomfördes med hjälp av adsorptionsmodellen. Den visade på att det var billigare med 2 stegs GAC jämfört med ett stegs GAC för EBCT 5 minuter och 8 minuter för målkoncentrationerna 2-10 ng/L. Men kostnadsberäkningen för EBCT 15 minuter visade på minskande kostnader med minskande målkoncentrationer vilket är ett orealistiskt resultat orsakat av för få datapunkter tillgängliga för att kunna modellera tillförlitliga resultat.

Nyckelord: GAC, PFAS, dricksvatten, två stegs GAC

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Populärvetenskaplig sammanfattning

PFAS är en typ av kemikalie skapad av människor som idag finns i naturen genom produktion och användning av produkter innehållande PFAS. Sådana produkter är Teflon-stekpannor, brandskum, skidvalla, regnkläder och mycket mer. Kemikalien innehåller kol-fluor-bindningar i olika långa kedjor vilket ger upphov till dess önskade egenskaper av att vara vatten- och fettavstötande samt att klara av höga temperaturer. Men det gör också att den är svårnedbrytbar och är därmed persistent i naturen. PFAS hittas i dricksvatten på alla kontinenter på jorden och även i Uppsala. Det har visat sig hälsoskadligt att konsumera PFAS och att dricksvatten är det huvudsakliga intaget av för människor. Därför måste halten PFAS minska i dricksvattnet innan det når konsumenterna.

Att ta bort PFAS från vatten har visat sig vara svårt men det finns en handfull metoder. Den mest välanvända metoden är kolfiltrering genom granulärt aktivt kol (GAC) och denna används i Uppsala. Vattnet passerar genom ett GAC-filter, där PFAS-ämnen kan fastna, innan det blir dricksvatten. En fördel med denna metod är att de PFAS-ämnen som fastnat på kolet kan förångas i en process som kallas regenerering och då kan kolet användas igen. Ett problem med denna metod är att även om den är bra på att ta bort långkedjade PFAS så är den inte så bra på att ta bort kortkedjade PFAS₄. Livsmedelsverket har sänkt gränsvärdet på PFAS₄ (de 4 vanligaste PFAS-ämnena) i dricksvatten till 4 ng/l. Detta är ett lågt gränsvärde som blir svårt för dricksvattenverken att nå. En förändring av dricksvattenproduktionen som skulle kunna underlätta för vattenverket att nå gränsvärdet skulle kunna vara att låta vattnet passera genom två kolfilter. Ett pilotanläggning med sådan 2 stegs kolfiltrering har låtit byggas på ett dricksvattenverk i Uppsala.

I det här examensarbetet jämfördes effektivitet av borttagning av PFAS mellan 1stegs och 2 stegs kolfiltrering för långkedjade och kortkedjade PFAS. Dessutom beräknades kostnaden för att nå det nya gränsvärdet med 1 stegs och 2 stegs kolfiltrering med hjälp av modellering. Alla jämförelser och beräkningar baserades på de PFAS-ämnen som förekommer i råvattnet till Uppsalas dricksvattenverk. Det konstaterades att det inte blir mer effektiv borttagning av två steg för vare sig långkedjade eller kortkedjade PFAS. Effektiviteten av borttagning var som störst för de långkedjade PFAS och som lägst för de kortkedjade PFAS. Tre olika flöden på vattnet testades vid olika tidpunkter och dess påverkan utvärderades i den ekonomiska beräkningen. De tre flödena modellerades för hela testperioden för att ta reda på hur koncentrationen PFAS₄ som lämnade kolfilterna skulle utveckla sig om endast ett flöde hade använts för hela testperioden. För det högsta och mellersta flödet var det billigare med 2 steg än 1 steg men för det lägsta flödet kunde ingen jämförelse med säkerhet göras på grund av för lite data.

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Glossary

AC Activated carbon.

AIX Anion exchange.

DWTP Drinking water treatment plant.

EBCT Empty bed contact time.

GAC Granular activated carbon.

Legacy PFAS The most produced and most common PFAS in the environment: PFOA and PFOS.

PAC Powdered activated carbon.

PFAS Per- and polyfluoroalkyl substances.

PFAS precursors longer chained PFASs that can degrade into shorter chained.

PFAS₁₁ The sum of PFAS₄ plus PFBA, PFPeA, PFHxA, PFHpA, PFDA, PFBS and 6:2 FTS.

PFAS₂₀ The same as PFAS₂₁ but without 6:2 FTS.

PFAS₂₁ The sum of PFAS₁₁ plus PUnDA, PDoDA, PTrDA, PFPeS, PFHpS, PFNS, PFDS, PUnDS, PDoDS and PTrDS.

PFAS₄ The sum of PFOS, PFOA, PFNA and PHF_xS.

PFBS Perfluorobutane sulfonic acid.

PFCA Peryfluoroalkyl substances with carboxyl acid as functional group.

PFHpA Perfluoroheptanoic acid.

PFHxA Perfluorohexanoic acid.

PFH_xS Perfluorohexane sulfonic acid.

PFOA Perfluorooctanoic acid.

PFOS Perfluorooctane sulfonic acid.

PFPeA Perfluoropentanoic acid.

PFPeS Perfluoropentane sulfonic acid.

PFSA Peryfluoroalkyl substances with sulfonic acid as functional group.

RO Reverse osmosis.

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1 Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of more than 9000 chemicals made by humans (EPA 2021). They consist of one or more C-atoms where the hydrogen atoms have been replaced by F-atoms and are connected to a functional group (Buck et al. 2011). Because of their wanted properties of being water and oil repellent and withstanding high temperatures, they have been widely used in industrial processes and products. However, they are also difficult and once released into nature they will hardly breakdown making them persistent (Dickman & Aga 2022). Therefore, they have been coined 'forever chemicals'. The widespread usage of these chemicals has led to that they can now be detected in the environment, animals and humans. This is due to environmental release at production factories, from disposal of PFAS-containing products and from biotic and abiotic degradation of PFAS precursors.

The PFAS released into the environment can reach drinking water sources and is now found in drinking water all over the world (Domingo & Nadal 2019, Lapworth et al. 2018, Boiteux et al. 2017, Bartolomé et al. 2017, Schwanz et al. 2016, Wilhelm, Bergmann, & Dieter 2010, Lu et al. 2018, T. Wang et al. 2015, Kaboré et al. 2018, Liu et al. 2017, Eriksson et al. 2013). Humans can be exposed to PFAS through drinking water, inhalation of air, food and exposure to contaminated ground (Li et al. 2020). However, drinking water is considered one of the most significant sources of PFAS to the general public (D'Hollander et al. 2010). During the last decade, the number of toxicological studies on PFAS has increased (Pelch et al. 2022). Especially toxicology studies on humans. Most of these studies has been done on PFOA and PFOS, the so called Legacy PFAS (Dickman & Aga 2022). Results from epidemiological studies of humans being unintentionally exposed to PFAS shows linkage to different kinds of cancers, higher cholesterol levels, decreased liver function and significant birth defects. PFAS alters the immune system and makes it harder for people to respond to vaccines (Beans 2021). The short-chained PFASs however, are not as bioaccumulative as the long-chained (Zaggia et al. 2016) and have shorter half-lives in humans (Nicole 2020).

More strict regulations on PFAS are emerging on the production of drinking water (Livsmedelsverket 2022). This poses a great challenge for drinking water treatment plants (DWTP) to remove the PFAS before sending the water to its consumers. Conventional treatment at DWTPs such as coagulation/flocculation, filtration and/or disinfection does not remove PFAS efficiently (Appleman et al. 2014). Therefore there is need for efficient techniques such as anion exchange (AIX), activated carbon (AC) and reverse osmosis (RO) in order to decrease the concentration of PFAS in drinking water. In this master's thesis, PFAS adsorption onto granular activated carbon (GAC) was investigated. GAC is better at adsorbing long-chained PFASs than short chained PFASs (Karbassiyazdi et al. 2023). However, the short chain removal might be increased if a two-stage GAC filter is used and it might also be a cheaper option when reaching the new water treatment goals on drinking water.

1.1 Aim

- Compare the removal of long and short chain PFAS at a two stage GAC pilot to a one stage pilot.
- Compare the costs of operating a full scale one stage GAC to a two stage GAC at a DWTP with respect to different treatment goals for the EBCT of 5, 8 and 15 minutes.

2 Background

2.1 What is PFAS?

Per- and polyfluoroalkyl substances (PFAS) are a family of anthropogenic persistent organic pollutants which have been produced since the 1940s (Z. Wang et al. 2017). They have been and are used in many products thanks to their appreciated qualities of being water and oil repellent and withstanding high temperatures. Today they can be found in firefighting foam, food contact materials, cosmetics, inks, household products, water repellent clothing and many more (Swedish Chemicals Agency 2015).

As introduced earlier, PFASs are molecules that contain at least one perfluoroalkyl moiety C_nF_{2n+1} (Buck et al. 2011). Fluorine is the atom with the highest electronegativity in the periodic table (Silberberg 2013) making the C-F bond found in PFAS very strong resisting oxidation and thereby conserves electrons (Katam & Bhattacharyya 2022). To break this bond requires a lot of energy. PFASs can be divided into two groups; Polymeres and Non-polymers (Buck et al. 2011). The non-polymers consists of Perfluoroalkyl substances and Polyfluoroalkyl substances.

The water analyzed in this thesis only consisted of Perfluoroalkyl substances and they can be divided into two subgroups; perfluoroalkyl carboxylic acids (PFCA)s and perfluoroalkyl sulfonic acids (PFSA)s.

The PFCAs contain a carboxylic acid functional group and the PFSAs contain a sulfonate as its functional group. An example of a PFCA is perfluorooctanoic acid (PFOA) that has a fluorinated chain length of 7 and an example of a PFSA is perfluorooctane sulfonic acid (PFOS) that has a fluorinated chain length of 8 carbons. Their chemical structure can be seen in table 1 where their functional group can be seen in the right end of the PFAS.

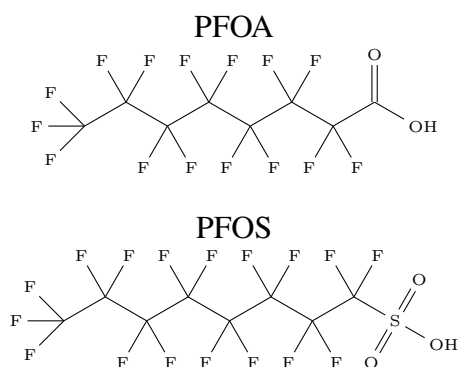


Table 1: Chemical structure of PFOA and PFOS

Except PFOA, the rest of the PFCAs found in the raw water of this study were Perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), Perfluoroheptanoic acid (PFHpA) and except PFOS the rest of the PFSAs were perfluorobutane sulfonic acid (PFBS), perfluoropentane sulfonic acid (PFPeS) and perfluorohexane sulfonic acid (PFHxS). Franke et al.

(2021) found that the PFAS with the highest concentration in the raw water in Uppsala is PFHxS, followed by PFOS and PFPeS.

2.2 PFAS in drinking water

Since 2016 the limit on PFAS in Swedish drinking water has been 90 ng/L for PFAS₁₁ (Enander 2015). However, as of the 1st of January 2026 the new limit in drinking water will be 4 ng/L for PFAS₄ (sum of the 4 most common PFASs) and 100 ng/L for PFAS₂₁ (sum of the 21 most common PFASs) (Livsmedelsverket 2022). This is a stricter limit than what is decided by the European parliament and of the council in the directive 2020/2184 on the quality of water intended for human consumption. The limit there is 500 ng/L for the sum of all PFASs and 100 ng/L for PFAS₂₀. In Denmark, however, they are lowering the limit even more on PFAS₄ to 2 ng/L (Miljøstyrelsen 2021).

Conventional treatment steps at DWTPs have been proven ineffective in removing PFAS where ozonation even resulted in an increase in PFAS concentrations (Boiteux et al. 2017). Therefore, there is a need for advanced techniques, and some of these are ion exchange, adsorption, advanced oxidation process and membrane based separation (Yadav et al. 2022). However, even if the advanced oxidation have showed promising results there is no full removal of PFAS but rather reduction of chain length which results in shorter PFASs that are more difficult to oxidize (Phong Vo et al. 2020). The membrane based separation produces a retentate of high PFAS concentration which is in need of further treatment. For the adsorption and the ion exchange there is the opportunity of incineration when the adsorption media is spent and thereby reaching the end of life for the captured PFAS. This decomposition is thought to occur through mineralization to fluorine (Wang et al. 2022). Both have its drawbacks of having lower removal efficiencies compared to nanofiltration but the advantage of not creating a retentate. The advantage of GAC over anion exchange (AIX) is that it is reliable, straight forward to operate have been used in DWTPs for decades and has a commercially available method for regeneration without creating an additional waste stream (Sonmez Baghirzade et al. 2021).

2.3 What is GAC?

GAC stands for granular activated carbon and is a type of activated carbon. In the process of making activated carbon two main steps are involved; carbonization and activation (Chang & Zainal 2019). The carbonization process, or pyrolysis as it is also called, occurs through heating in an oxygen free environment. The activation process can be divided into the thermal or physical activation and the chemical activation. (Marsh, Rodríguez Reinoso, & Rodríguez-Reinoso 2006). The goal of the activation process is to enhance the porosity and thereby maximize the adsorption potential. The thermal or physical reactivation is done by gasifying the carbon with either carbon dioxide or water vapor. The chemical activation is done by adding a chemical agent to the carbonization step such as phosphoric acid or potassium hydroxide. Activated carbon can be made into granular form (GAC) and powdered form (PAC) and can be made from nutshells, coals, woods, cloths and felts. In 1901, a commercially available method for producing activated carbons were introduced by Rapfael von Ostreyko (Dąbrowski 2001). In 1929, the first activated carbon filter were used for water treatment and in the 1970s they were used to remove a wide range of synthetically made pollutants from water and gases (Inglezakis & Pouloupoulos 2006).

2.3.1 Adsorption using GAC

PFASs in water that passes through a GAC filter adheres to the GAC through a process called adsorption. The PFASs dissolved in the liquid phase thereby stays onto the solid GAC. Adsorption occurs through electrostatic interaction and van der Waals forces (hydrophobic interaction) when there is adhesion of molecules dissolved in the liquid to the surface of the solid media (Katam & Bhattacharyya 2022). The PFASs can also be trapped using pore blockage when natural organic matter plugs open pores in the GAC. These processes can be seen in figure 1.

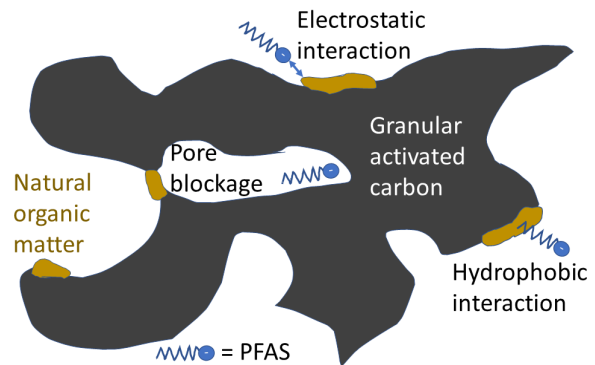


Figure 1: Adsorption and pore blockage of PFAS onto GAC. Figure modified from Katam & Bhattacharyya (2022)

PFASs have been shown to be more easily adsorbed onto GAC than PFCAs (Chen et al. 2022). Long chained PFASs can be involved in the formation of aggregates on the surface which can improve the adsorption through GAC (Gagliano et al. 2020). However, if PFASs of different chain length co-exist in the water, the longest out compete the shorter due to stronger hydrophobic interactions. This can lead to desorption which occurs when the outgoing concentration of a PFAS is greater than the incoming concentration. If a short chained PFAS has been adsorbed to the surface of GAC, a longer chained PFAS can push it away and take its place on the GAC which leads to desorption. The effect of natural organic matter (NOM) in the water can both increase the adsorption and decrease the adsorption. It seems as most studies has found an negative effect due to competition of the active sites on the GAC and that the effect is not as strong for longer chained PFASs. While long chained PFASs are found both in the dissolved phase but also found on the organic matter, short chained PFASs are only found in dissolved phase (Ahrens et al. 2010).

An advantage of operating a 2 stage GAC at the DWTP is that it is easier to achieve plug flow in the GAC container. Plug flow is an idealized flow where all the particles going through an adsorption media have the same velocity (Coker 2015) and can be seen in the left part of figure 2. This is more likely to occur at the top of the GAC. The benefit of plug flow is that it means that all of the GAC would be utilized and no PFAS would reach breakthrough earlier than the others. The opposite of plug flow can be seen in the right part of figure 2 where the water flows faster in certain areas of the GAC. This means that not all the GAC is utilized for PFAS adsorption and that the breakthrough of certain PFASs would occur earlier than with the plug flow. In a two stage GAC, the water is collected after the first stage and then led through another GAC stage. Thereby, eventual breakthrough in the first stage can be monitored and since the water passes through another step, this allows for a more confident removal of PFAS.

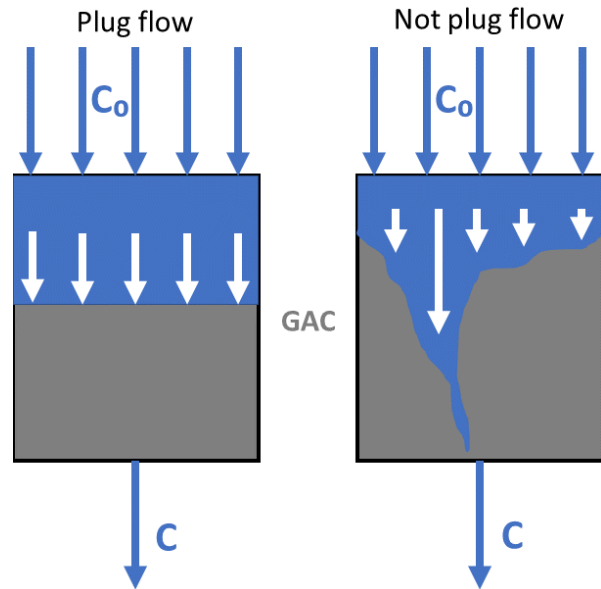


Figure 2: Visualisation of plug flow through a container of GAC on the left and the unwanted flow of breakthrough of water in certain areas of the GAC on the right. C_0 is the incoming concentration to the GAC and C is the outgoing concentration. The white arrows show how fast the water is flowing.

2.4 Model

The Lin & Huang adsorption model (1999) has been used by McCleaf et al. (2017), by Cantoni et al. (2021) and Franke et al. (2019) to model PFAS adsorption onto GAC. The model is based on adsorption of benzene, toluene, ethylbenzene and xylene (BTEX) onto macroreticular resins. The concentration leaving an adsorption media can be explained by equation 1

$$C = \frac{C_0}{1 + \exp[k_c(\tau - BV)]} \quad (1)$$

or

$$BV = \tau + \frac{1}{k_c} \ln \frac{C}{C_0 - C} \quad (2)$$

where C is the outgoing concentration, C_0 is the incoming concentration, k_c and τ are adsorption parameters determined for every case and BV is the number of bed volumes treated. This is an empirical model that assumes steady state mass transfer for one substance in water to the adsorbent. A graph of the cumulative BVs on the y-axis and $\ln \frac{C}{C_0 - C}$ on the x-axis is created. An example of this can be found in figure 14A. After that, trendlines in the data are found and using equation 2 the parameters k_c and τ can be determined. They are then used in equation 1 to model the concentration for a range of BVs using an incoming concentration (C_0). An example of a modeled concentration using this equation can be seen in figure 14B.

2.5 Previous studies on PFAS adsorption onto GAC?

McCleaf et al. (2017) found no desorption for PFBS within 50 000 BVs treated for a one stage GAC. The PFASs had a higher removal efficiency compared to the PFCAs and that the removal efficiency for the short chained PFCAs was the worst. They furthermore modeled the concentration of PFPeA, PFHxA, PFHpA, PFOA, PFBS, PFHxS and PFOS with an EBCT of 6.1 minutes using the Lin & Huang adsorption model (1999). They found a change in adsorption trends after 50 days of operation and called this first period phase 1. This phase got its individual k_c and τ values and was followed by a phase 2 with other k_c and τ values. The phase 1 k_c and τ values were then used to model the concentration for the first 50 days and the phase 2 k_c and τ values were used to model the following days.

A two stage GAC filtration study was made by Vahala (2002) for organic matter removal. They found that neither installing a two stage GAC nor increasing the EBCT (10-15 min) resulted in a more effective use of GAC in terms of NOM removal. Another two-stage GAC filtration was used by Eschauzie et. al (2011). The finished drinking water contained several short chained PFASs while the long chained were removed. Furthermore, it showed that most conventional treatment processes in DWTPs does not remove PFASs.

Franke et al. (2019) investigated AIX and GAC adsorption on both raw water and the retentate from nanofiltration. They found that nanofiltration removed >99% of PFAS for the water in Uppsala. For both the raw water and retentate adsorption, the AIX had higher adsorption than GAC but with a higher difference for the retentate treatment. Chow et al. (2022) found that breakthrough curves for PFAS follows an asymmetric sigmoidal shape that shows that the removal efficiency is high at first, then decreases rapidly and eventually reach saturation where the removal efficiency is zero due to all active sites on the GAC being occupied. In cases of short chained PFASs with less than 4 carbons (<4 C), desorption occurred. They furthermore used a similar model to that used in this report to model breakthrough of different PFCAs and PFASs of different chain-lengths. They found that that short chained PFCAs C_4 , C_3 and C_2 and PFASs C_2 experience desorption within 60000 bed volumes. They adjusted their model to exclude the desorbing data points in order to get a functional model for breakthrough of PFAS.

Belkouteb et al. (2020) found that conventional treatment techniques are insufficient in removing PFAS. They also found that GAC is a straight forward and reliable treatment method for the removal of PFAS and stated that there is an existing competitive GAC market that can be used to minimize operational costs. Furthermore they saw that PFASs were removed more easily than PFCAs and that lowering the flow resulted in higher removal efficiencies. Especially for GAC that has been in operation for a while. They also made a cost analysis that showed that treatment goals has an overwhelming effect on the cost and that the dominant treatment cost was the regeneration cost. Son et al. (2010) made a comparison of virgin and regenerated GAC. They compared the breakthrough of dissolved organic carbon (DOC) in virgin GAC to regenerated GAC that has been regenerated 1-3 times. The results were that the adsorption capacity of the virgin GAC was equal to that of all the regenerated GAC.

3 Method

Two main comparisons were made to compare operating a one stage GAC to two stage GAC. A comparison of removal by chain length and a cost comparison for PFAS₄.

3.1 Pilot operations

The setup of the pilot can be seen in figure 3 which includes a sketch of Bäcklösa DWTP at the top of the figure. This is included to show that the pilot received the same type of water as the full scale GAC filters at the DWTP is doing. The pilot consisted of pumps in two locations in order to pump the water from the raw water tank to the first stage GAC and from the collection tanks after the first stage to the second stage. The first stage filters consisted of a virgin Aquasorb 6300 GAC (Jacobi 2012), two regenerated Filtrasorb 400 GAC (CalgonCarbon 2019), two virgin Filtrasorb 400 GAC and a blank column without any filter. The second stage filters consisted of two regenerated Filtrasorb 400 GAC, two virgin Filtrasorb 400 GAC and a blank. The water leaving the virgin Aquasorb 6300 GAC and the two virgin Filtrasorb 400 GAC were collected in a tank and were pumped to the two virgin Filtrasorb 400 GAC and the blank in the second stage. The water leaving the two regenerated Filtrasorb 400 GAC were collected in another tank and were then pumped to the two regenerated Filtrasorb 400 GAC in the second stage. The water leaving the two regenerated and virgin Filtrasorb 400 GAC in the second stage were then collected in a collection tank but the water leaving the two blanks was not collected. They were included in the experiment as a reference to the columns with GAC.

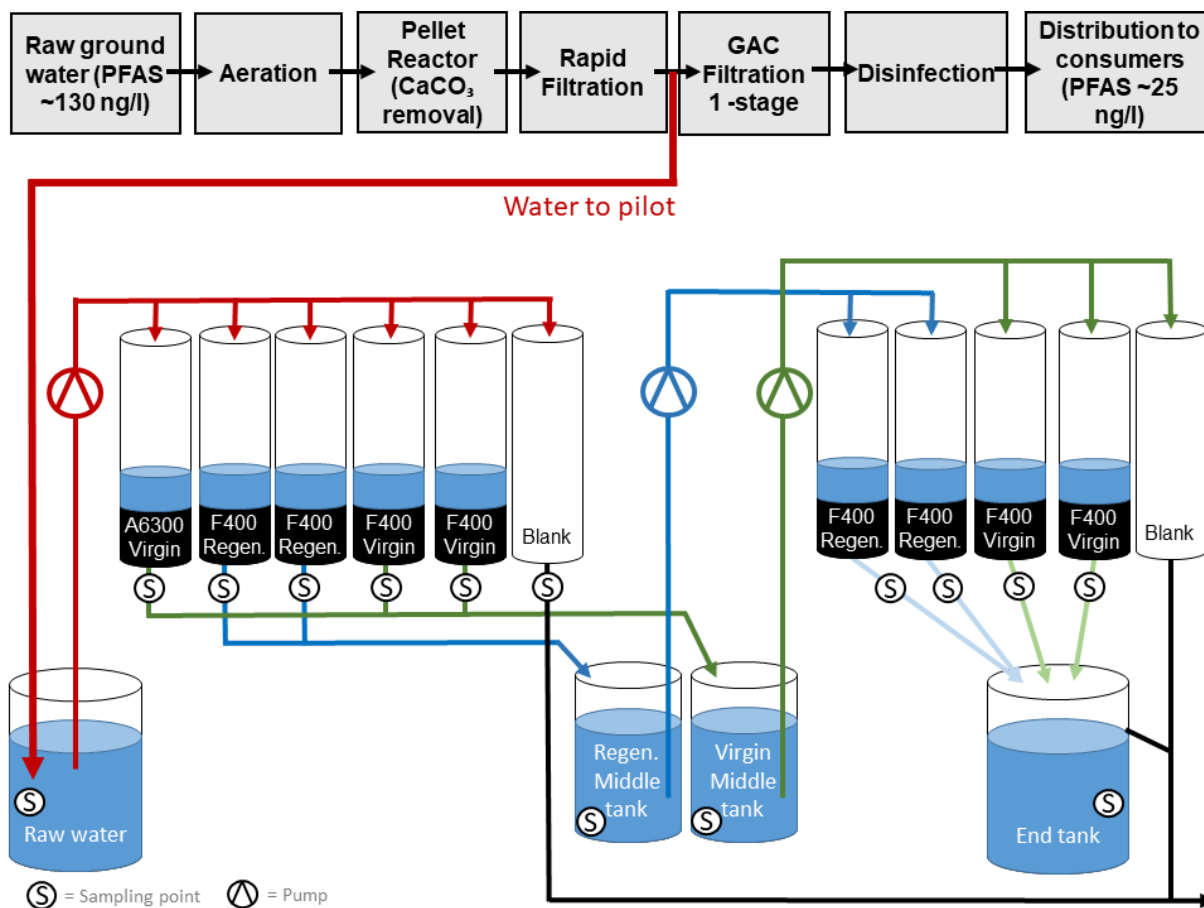


Figure 3: Scheme of two-stage GAC for PFAS removal. In grey at the top of the figure is the treatment scheme for Bäcklösa DWTP. The red arrow is the Raw water to the pilot, the dark blue color is the water that has passed by one stage of regenerate GAC and the lighter blue color is the water that has passed by two stages of regenerated GAC. The darker green is the water that has passed by one stage of virgin GAC and the lighter green is the water that has passed by two stages of virgin GAC. The black lines are water going to the sewage. The 15 sampling points in the pilot and the pumps are shown in two stages.

The pilot was visited twice a week from 2022-07-01 until 2023-02-28 before the setup of the pilot was changed. The data between these dates is what is included in this thesis and consisted of three periods that can be seen in table 2. The first period, until 2022-09-27 had a flow of around 100 ml/min resulting in an empty bed contact time (EBCT) of 5 min. The second period lasted until 2023-01-17 with a flow of around 50 ml/min and an EBCT of 8 min. The last period had a flow of around 25 ml/min with an EBCT of 15 min. The EBCT was calculated using equation 3.

Table 2: Pilot operations.

	Period 1	Period 2	Period 3
EBCT [min]	5	8	15
Flow [ml/min]	100	50	25
Start date	2022-07-01	2022-09-27	2023-01-17
End date	2022-09-27	2023-01-17	2023-02-28
BVs treated	0-26000	26000-45000	45000-50000

The EBCT was calculated using equation 3

$$EBCT[min] = \frac{V_{BV}[ml]}{flow[ml/min]} \quad (3)$$

where V_{BV} is the bed volume in ml and $flow$ is the flow coming out of the column in ml/min. The three flows that were used and the corresponding calculated EBCTs can be seen in table 2.

Every week, 15 PFAS samples were gathered from the 15 sampling points shown in figure 3. The samples from every other week were sent to the lab for PFAS analysis. Occasional samples, that were not sent in, were later sent to the lab to confirm or discard surprising values in the results. For each sampling, the height of the water inside the column as well as the height of the GAC was noted. Furthermore, the flow, pH, conductivity and temperature was measured and recorded. The whole sampling period with concentrations of PFAS₄ can be seen in figure 4.

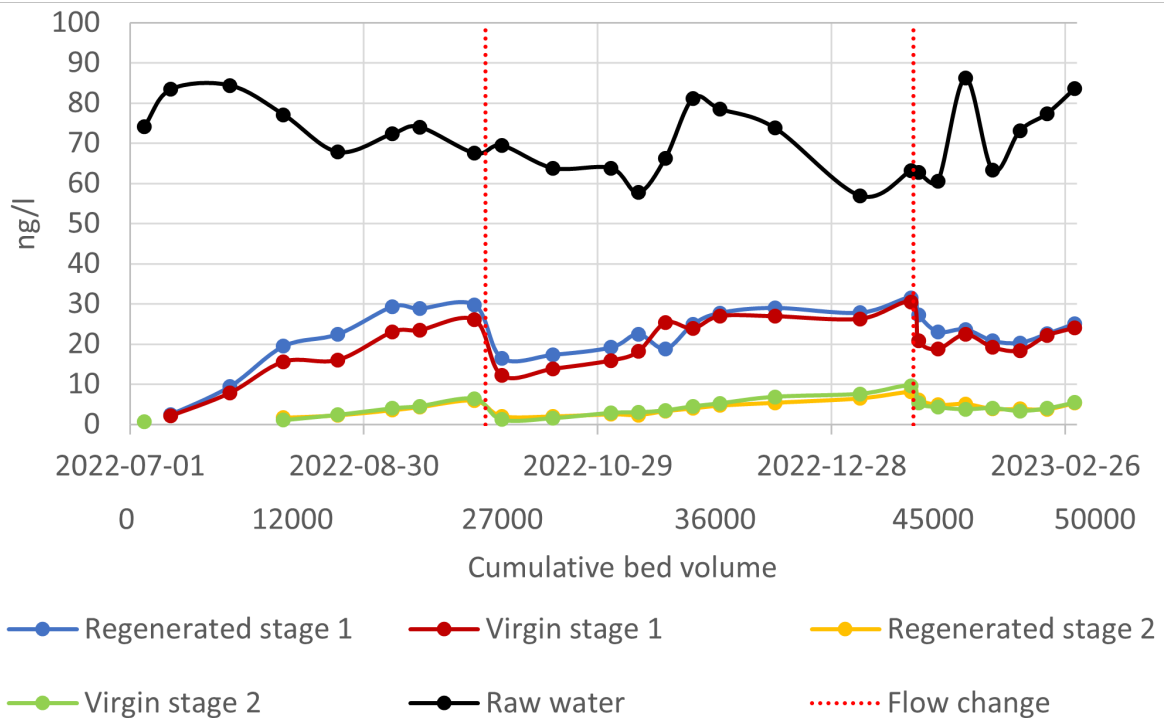


Figure 4: PFAS₄ concentrations for the whole pilot operation period with concentrations in the (black) raw water, (blue) regenerated stage 1, (red) virgin stage 1, (yellow) regenerated stage 2 and (green) virgin stage two. The flow changes (see table 2) are also visible in the figure by the red vertical dotted lines.

3.2 Data handling

The data consisted of concentrations of 34 different PFASs as well as PFAS₄, PFAS₁₁, PFAS₂₀ and PFAS₂₁. These can be seen in appendix in table A1. If a data point was below the level of detection, it was assumed to be zero. This resulted in gaps in the data set. The data from 2022-09-20 was excluded from all the calculations because it was considered an outlier. The PFASs that existed in more than half of the samples can also be seen in table 3 where their individual C-F chain length can be seen.

Table 3: Existing PFASs in Uppsala's untreated groundwater.

CF-chain length	4	5	6	7	8
PFASs	PFBS	PFPeS	PFHxS		PFOS
PFCAs	PFPeA	PFHxA	PFHpA	PFOA	

3.3 Bäcklösa DWTP

A two stage pilot was installed at Bäcklösa DWTP. Today, the plant has a full scale one stage GAC which operates at a flow resulting in an EBCT of 15 minutes. 10 GAC filters are placed in parallel and 6 of them are usually operated simultaneously. The filters are of the same type as in the pilot; Aquasorb 6300 GAC (Jacobi 2012) and Filtrasorb 400 GAC (CalgonCarbon 2019). The treatment scheme at Bäcklösa DWTP can be seen in figure 3. First the raw water is aerated, and then the $CaCO_3$ is removed in a pellets reactor. This is followed by a rapid filtration and then the water goes to the GAC filter before being disinfected. The plant produces approximately 7000000 m^3 of water each year and has a current treatment goal of 25 ng/L for PFAS₁₁. The GAC filters contain a GAC volume of 40 m^3 .

3.4 Sieving analysis

A dry sieving analysis was performed on the first stage GAC-filters after the pilot was stopped. The reason for this was to determine the difference in particle size between the regenerated and virgin GAC and to determine the apparent density. A potential difference in particle size between two samples of GAC could be an explanation for difference in adsorption capacity and thereby removal efficiency. The apparent density can be used to calculate the weight of GAC for samples that has not been analyzed in a sieving analysis but has a known volume.

Before the sieving analysis could be started, the 5 first stage columns were emptied. The wet GAC was dried in an oven and then on the top of the radiator in order to achieve complete dryness. The machine used for the sieving analysis was a vibration sieving machine and in order to choose the appropriate sizes for analysis, test results from an earlier sieving analysis of GAC was investigated. The sizes <0.2 mm, 0.2 mm, 0.4 mm, 0.6 mm, 0.8 mm, 1.0 mm, 1.25 mm and 1.6 mm were chosen to resemble those test results. In order to determine the appropriate time for each analysis, a sieving test was performed before the actual sieving analysis was started. This was done using a sample of 353 g of virgin Aquasorb 6300 GAC (Jacobi 2012). The sieving time of 20 minutes was chosen appropriate for the sieving analysis.

For each sample, the total weight of the sample and the individual weight of each size exclusion was found. An effective size (the size of the smallest 10 % of the sample) and the Uniformity coefficient (the size of the smallest 60 % of the sample divided by the effective size) could be calculated for the samples in the test. They can be seen in table 4.

Table 4: Effective size (10 % diameter) and uniformity coefficient (60 % diameter / 10 % diameter) from the sieving analysis.

	Virgin Aquasorb 6300	Regen. Filtrisorb 400	Regen. Filtrisorb 400	Virgin Filtrisorb 400	Virgin Filtrisorb 400
Effective size (mm)	0.685	0.67	0.665	0.74	0.75
Uniformity coefficient	1.87	1.74	1.74	1.74	1.72
Total weight (g)	267	292	289	318	314

The average total weight for the first stage regenerated and virgin GAC was calculated from table 4. The average bed volume in the regenerated and virgin first stage was used to calculate the average apparent density for the regenerated and the virgin GAC. This number was then used, together with the average bed volume for the regenerated and virgin second stage to calculate the average weight of the GAC in the second stage. These numbers can be seen in table 5.

Table 5: Sieving analysis average results, apparent density and weight of GAC in the pilot.

	Regenerated GAC		Virgin GAC	
	Stage 1	Stage 2	Stage 1	Stage 2
Average apparent density [g/ml]	0.69		0.78	
Average weight of GAC [g]	291	299	316	328
Average bed volume [ml]	419	430	405	420

3.5 Removal efficiency calculations

The removal efficiency, RE , was calculated using equation 4

$$RE = \frac{C_0 - C}{C_0} \cdot 100\% \quad (4)$$

where C_0 is the incoming concentration to the GAC filter and C is the concentration coming out of the filter. It was calculated for the PFASs found in Uppsala's groundwater presented, in table 3. A comparison of removal efficiency over time with respect to chain length was made. In the graphs with the comparison gaps occurred. These were caused by some of the data being below the detection limit. For the removal efficiency, only the regenerated GAC data is presented. This is because the regenerated and the virgin graphs looked similar. In order to prove that they are not significantly different a T-test was performed and in order to do the T-test, the series of data needed to be detrended. The detrended series and the results from the T-test can be seen in figure 10 and table 8.

The removal efficiencies based on the data from the pilot contains the removal efficiencies for the three EBCTs spread over different BVs. This makes the comparison of a removal efficiency for the BVs of 0-26000 to another removal efficiency at a bed volume of >26000 BVs unfair. In order to find a fair comparison of removal efficiency for an increasing number of bed volumes, modeling was made using Lin & Huang adsorption model (1999) for each EBCT individually. This was done for each of the PFASs and PFCAs present in the water.

3.6 Removal comparison of 1 stage to 2 stages

The removal comparison of 1 stage to 2 stages was made by dividing the removal by the weight of GAC and by the number of BVs treated. It was calculated using equation 5

$$\frac{Removed[ng] / GAC[g]}{BV} = \frac{(C_0 - C) \cdot V_{time} / W_{GAC}}{BV} \quad (5)$$

where V_{time} is the volume in Liters that has passed through the column between each sampling time, W_{GAC} is the weight in gram of the GAC in the column and BV is the number of bed volumes that has passed. The reason for dividing by the weight of GAC was to make a fair comparison between the removal of one stage and removal of two stages and by dividing by the number of BVs the comparison of the three EBCTs could be made. The removal for each EBCT, and thereby an interval of bed volumes, was placed into one bar with visible average, top and lower quartile as well as visible potential outliers. These results can be seen in figure 13. A comparison of the removal between stage one and stage two can be seen in appendix in figure A1.

3.7 Cost calculation

The cost calculation was performed for PFAS₄ removal for the EBCT of 5, 8 and 15 minutes individually. No cost calculation was made for PFAS₂₁ since the concentrations after the first and the second stage never went above the limit of 100 ng/L during the pilot. Therefore, the PFAS₄ limit was considered the most important limit from a cost perspective since the PFAS₄ concentrations went above the limit of 4 ng/L. Because the pilot was operated at three different EBCTs, a modeling using Lin & Huang adsorption model (1999) was done to allow comparison of the different EBCT data. By using this model, the concentrations of PFAS₄ for the BVs of 0-60000 could be estimated. In figure 4, the concentrations for in the incoming tank, the first stage and the second stage could be seen. The EBCT of 5 and 8 minutes can be distinctly seen with increasing trends in the first and second stage between the flow changes. For the EBCT of 15 minutes, there is only an increasing trend in the last three points of the data. Therefore, the only points that were included were the last three points of that EBCT. Moreover, the data from 2022-09-20 was also excluded from the modeling of EBCT 5 minutes because it was considered an outlier.

The modeling was done by plotting $\ln \frac{C}{C_0 - C}$ on the x-axis and BVs on the y-axis resulting in figure on the left of figure 5. The trendlines represents equation 2 and from there the parameters k_c and τ can be found for each column. These parameters can then be used to calculate the modeled concentration using equation 1 for each bed volume and incoming concentration resulting in a graph looking like the right part of figure 5. The figure could then be used to find the number of bed volumes where each modeled line crossed the concentration of 2, 4, 6, 8 and 10 ng/L.

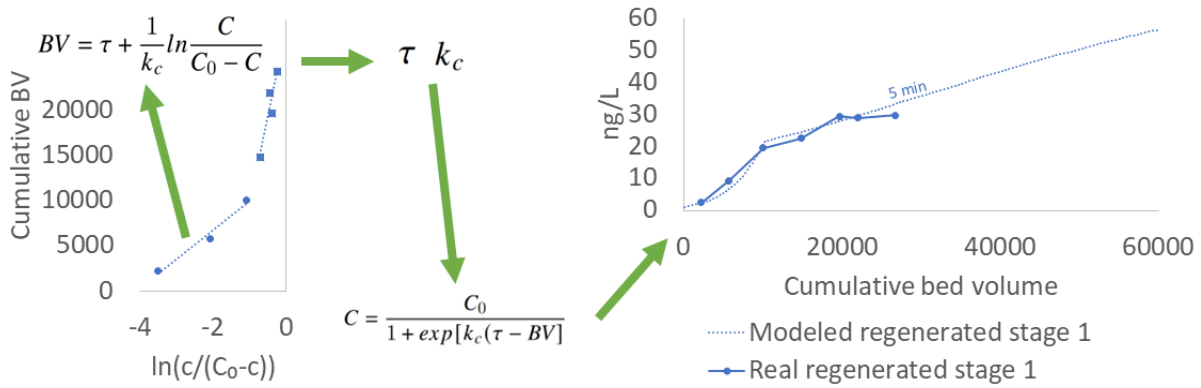


Figure 5: A visualization on how the modeling was made for the EBCT of 5 minutes for PFAS₄. The green arrows shows how the process was conducted step by step.

The parameters for the calculation comparison between one stage and two stages are shown in table 6. The differences between 1 stage and 2 stages included in the calculation are the double investment cost of virgin GAC, the double pumping cost, the number of bed volumes between each regeneration and the weight of one stage vs two stages of GAC.

Table 6: Examples of cost calculation parameters for the regenerated EBCT 5 minutes. Only the treatment goal of 2 and 4 ng/L is included in the figure.

Parameter	GAC filter Filtrasorb® 400			
	2		4	
Treatment goal [PFAS ₄ , ng/L]	1	2	1	2
Treatment stages	1	2	1	2
Reaches target after certain BV ^a	1600	12000	3350	20000
BVs treated per regeneration cycle ^b	1600	10400	3350	16650
Service volume (m ³) ^c	64000	416000	134000	666000
Number of needed regenerations per year	109	17	52	11
Yearly regeneration cost (euro) ^d	3120000	481000	1490000	300000
Yearly pumping cost (euro) ^e	14000	28000	14000	28000
Virgin GAC purchase (euro) ^f	5920	11800	5920	11800
Total cost (euro) ^g	3140000	520000	1510000	340000
Total cost/GAC [euro/kg GAC]	110	9.4	54	6.1

^a From the intercept shown in figure 14B.

^b Calculated according to the concept shown in figure 6.

^c The number of BVs multiplied by the volume of one BV.

^d GAC regeneration cost 714 euro m⁻³ wet GAC

^e 0.002 euro/m³ concentrate

^f Virgin GAC cost 1142 euro m⁻³ wet GAC and service life 10 years with interest rate 5%

^g Based on the apparent density calculated from the sieving analysis.

In order to determine the number of BVs between each regeneration cycle a concept was needed (figure 6). The blue dots stand for the concentration in the first stage and the orange for the second stage. When the target concentration is about to be exceeded in the second stage (in figure 6 this is 4 ng/L) the process is stopped. The second stage is moved and placed as the first stage and the original first stage is regenerated and placed as the new second stage. Thereby, the orange dots turn into blue dots when the new regeneration cycle starts and the blue dots turn into orange, newly regenerated dots. The figure is based on real values until the second stage

reaches 4 ng/L. The rest is based on the assumption that the first stage is going to experience the same concentration increase by BVs as it did in the pilot while the second stage is calculated from the removal efficiency from when the first stage reaches 4 ng/L until the second stage reaches 4 ng/L. Therefore, the number of BVs will be the same for each regeneration cycle. The calculation is based on the modeled values for the virgin and regenerated filters. Therefore, the the number of BVs between each regeneration for the two stage system is calculated by subtracting the number of BVs when the second stage reaches the treatment goal with the number of BVs when the first stage reaches the treatment goal. For the one stage system, the number of BVs between each regeneration cycle is just the number of BVs it takes for a regenerated or virgin GAC to reach the treatment goal.

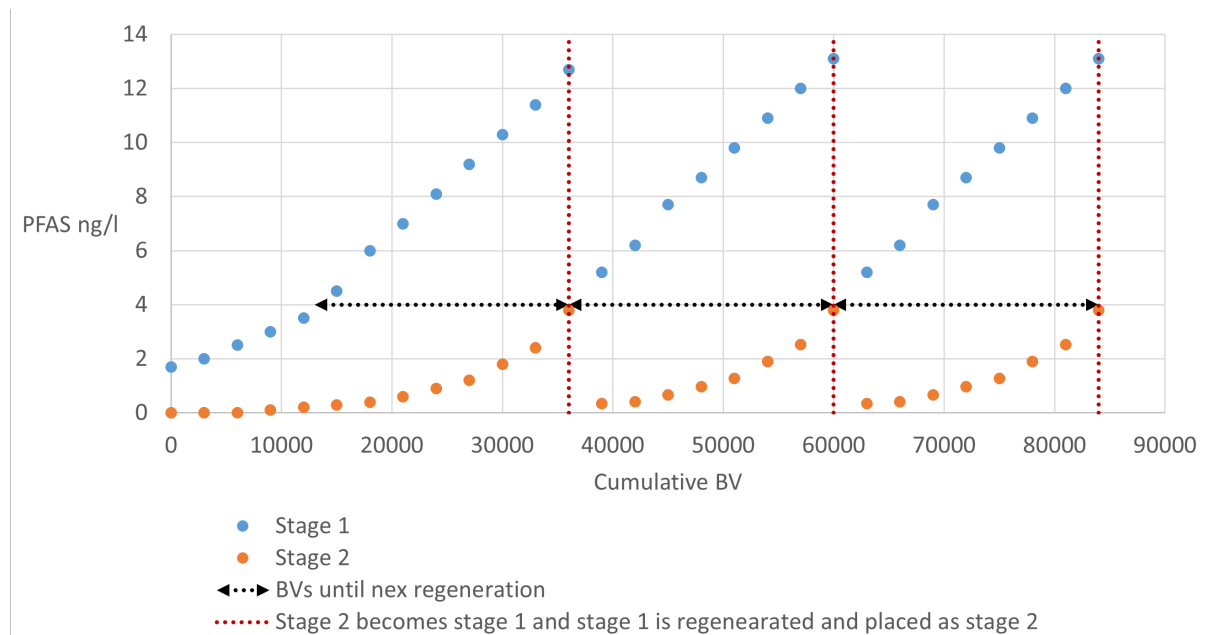


Figure 6: The concept behind the economic calculations

According to Lin & Huang (1999) there is a linear relation between k_c and τ for different flows. Thereby, it should be a linear trend between k_c and τ in this experiment but instead of plotting time and flow on the y-axis and x-axis, the cumulative BV and the EBCT was plotted on the y-axis and x-axis (figure 15). The finding of k_c and τ was based on the incoming concentration in the raw water tank for stage 1. For stage 2 it was based on the middle tanks with the water that had passed the regenerated filters in the first stage for the second stage regenerated filters and likewise for the virgin filters. However the modeling using those parameter could not be done using the middle tank concentrations. This is because the concentration in the middle tanks were highly affected by the different flows and therefore it reflects the unwanted flow changes in the modeling. The concentrations in the middle tanks can be seen in figure 7A. There are clear trends in the middle tank concentrations and the result is that the concentration decreases more than what is suspected in the modeling. This can be seen in the light red color in figure 7B. The dotted line in figure 7B is the modeled concentration using the modeled concentrations as the incoming concentration from the first stage in equation 1. The modeled concentrations were calculated using the average concentration for the first stage and the modeled concentrations for the second stage.

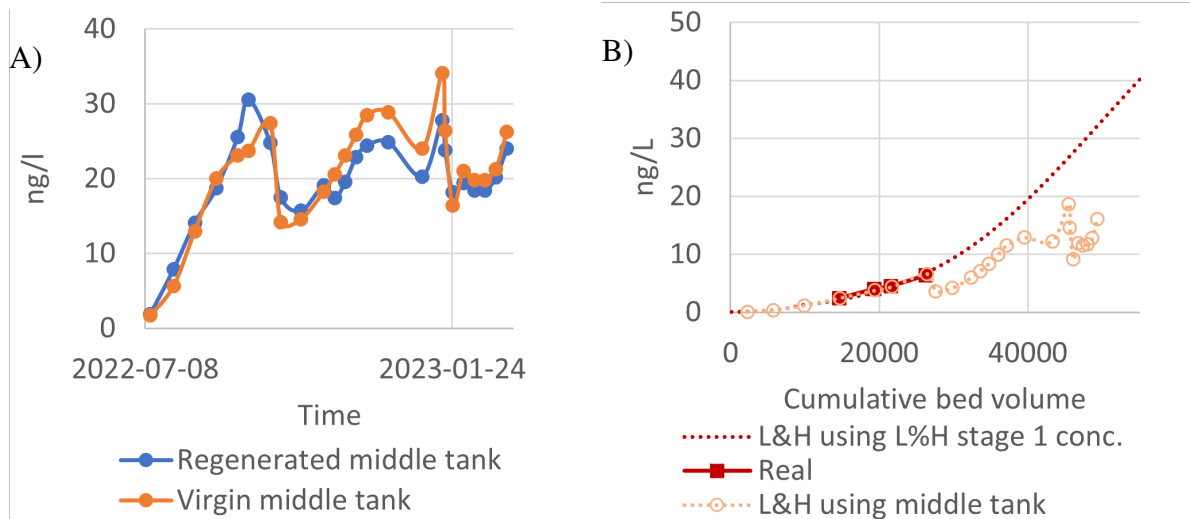


Figure 7: (A) The concentrations of the middle tanks and (B) the modeling using the middle concentrations (red circles), the modeling using the first stage modeled concentrations (dotted line) and the actual concentrations (squares)

In order to visualize the costs per weight of GAC graphs were made with the treatment goal on the x-axis and the cost per weight of GAC on the y-axis for each EBCT and the virgin and regenerated GAC individually. Also a calculation of the cost difference between 1 stage and 2 stages was made. The difference in cost between 1 stage and 2 stages was calculated using equation 6

$$\%difference = \frac{Cost_1 - Cost_2}{Cost_1} \cdot 100\% \quad (6)$$

where $Cost_1$ is the cost in €/kg GAC for 1 stage and $Cost_2$ is the cost in €/kg GAC for 2 stages. However, for those treatment goals where the cost of having a 2 stage GAC was more expensive than having a 1 stage GAC, the calculation was flipped. In those cases $Cost_1$ changed places with $Cost_2$.

4 Result

The results section includes a segment with the raw water concentrations, a segment with the removal efficiencies in the first and section stage with a T-test comparison for the removal efficiencies for the virgin and regenerated GAC, a display of the results for modeling the PFASs found in the raw water, then a comparison for the 1 stage and 2 stages removal per weight of GAC per BV is included and finally a section with the economic comparison between 1 stage and 2 stages.

4.1 Raw water and PFAS₂₁

The concentrations for the PFASs in the raw tank can be seen in table 7. The concentrations for all the individual PFASs are higher than for the PFCAs and that the PFSA with the highest concentrations is PFHxS and the PFCA with the highest concentration is PFHxA. The total number of samples included in the experiment was 25 and in the table it is shown how many of those points are above the level of detection. All included PFAS have more than half of the points included and the PFAS with the least points is PFHpA which also has the lowest average concentration of all the PFASs. The average concentration of PFAS₄ and PFAS₂₁ in table 7 is the average concentration of the black line in figure 4 and figure 8 respectively.

Table 7: Average raw water concentrations for PFASs present in Uppsala's groundwater during the whole pilot period, the number of data points used and the standard deviation.

	Average concentration (ng/L)	Data points used/ total number	Standard deviation (ng/L)
PFCA (C-length)			
PFPeA (<i>c</i> ₄)	1.8	24 / 25	0.3
PFHxA (<i>c</i> ₅)	4.8	25 / 25	0.8
PFHpA (<i>c</i> ₆)	1.3	16 / 25	0.2
PFOA (<i>c</i> ₇)	3.7	25 / 25	0.8
PFCA (C-length)			
PFBS (<i>c</i> ₄)	5.9	25 / 25	0.7
PFPeS (<i>c</i> ₅)	6.1	25 / 25	0.9
PFHxS (<i>c</i> ₆)	49	25 / 25	6.3
PFOS (<i>c</i> ₈)	19	25 / 25	2.4
Sum of PFASs			
PFAS ₄	71	25 / 25	8.5
PFAS ₂₁	91	25 / 25	11

The concentrations of PFAS₂₁ in the raw water (black) and after the regenerated stage 1 (blue), regenerated stage 2 (yellow), virgin stage 1 (red) and virgin stage 2 (green) can be seen in figure 8. The black line can be compared to the new limit in drinking water of 100 ng/L. It can be seen that even though the raw water is above the limit the water coming out of the first and the second stage is well below that limit and therefore the economic calculation was only performed for PFAS₄.

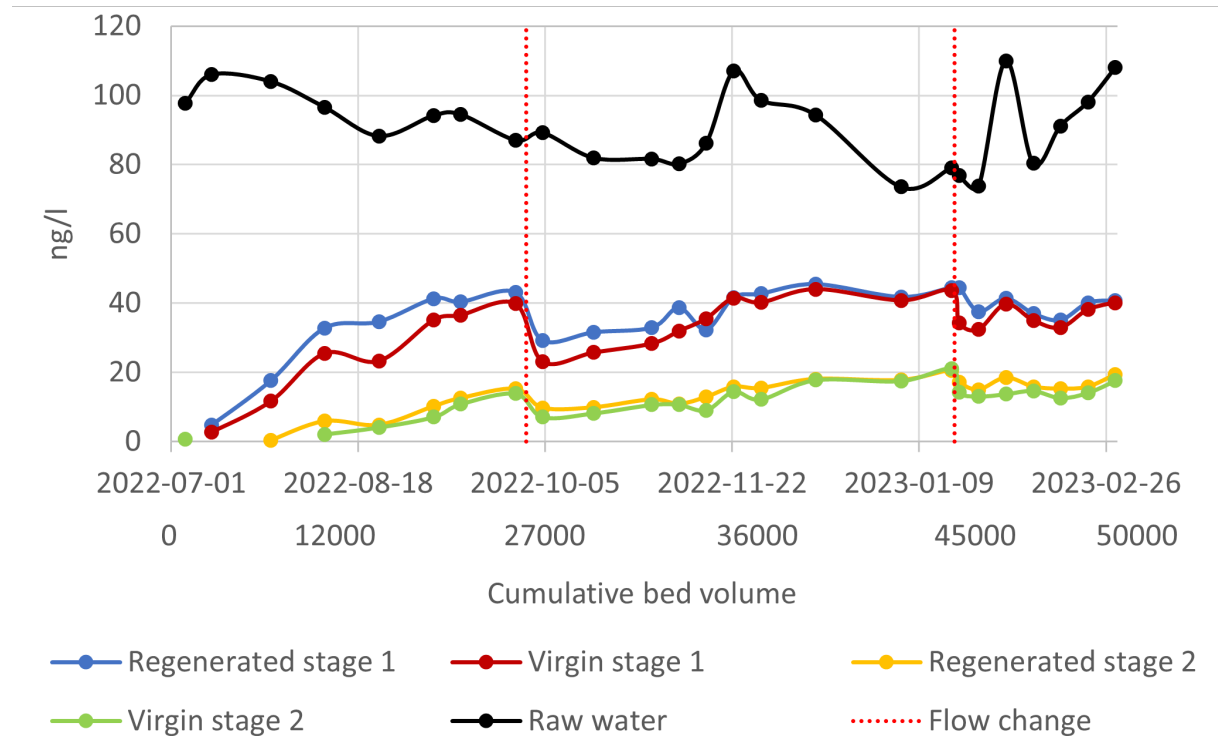


Figure 8: PFAS₂₁ concentrations for the whole pilot operation period. The flow change is shown in table 2.

4.2 Removal efficiencies

The removal efficiencies for the PFASs in the first stage can be seen in figure 9A and the second stage in figure 9B. It can be seen that no desorption occurs except for PFBS in the first stage at roughly 46000 bed volumes. The removal efficiencies for the PFCAs in the first stage can be seen in 9C and for the second stage in figure 9D. Desorption occurs for both PFPeA and PFHxA in both the first and the second stage. For all the removal efficiencies shown in figure 9 it can be seen that even though all PFASs start with a high removal efficiency, the removal efficiency decreases faster, for the short chains than for the longer chains.

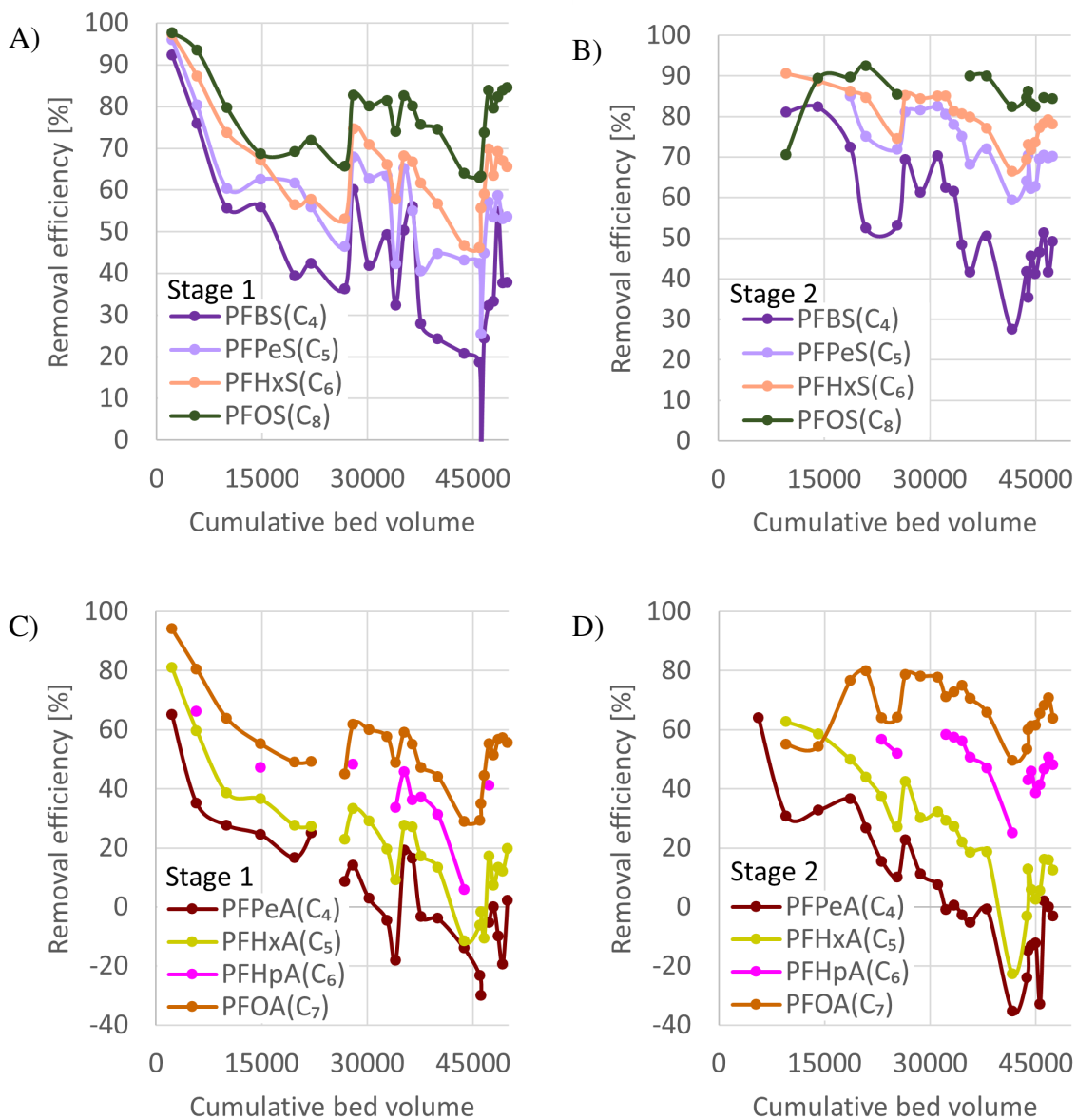


Figure 9: Removal efficiencies for the regenerated columns at stage 1 and stage 2 for the PFASs in (A) and (B) and for the PFCAs in (C) and (D). The PFASs chain lengths are presented in parenthesis.

The detrending of the removal efficiencies was done in order to make a T-test between the virgin GAC and the regenerated GAC. An example of how the original data looked like for PFOA can be seen in figure 10A and the detrended series can be seen in figure 10B.

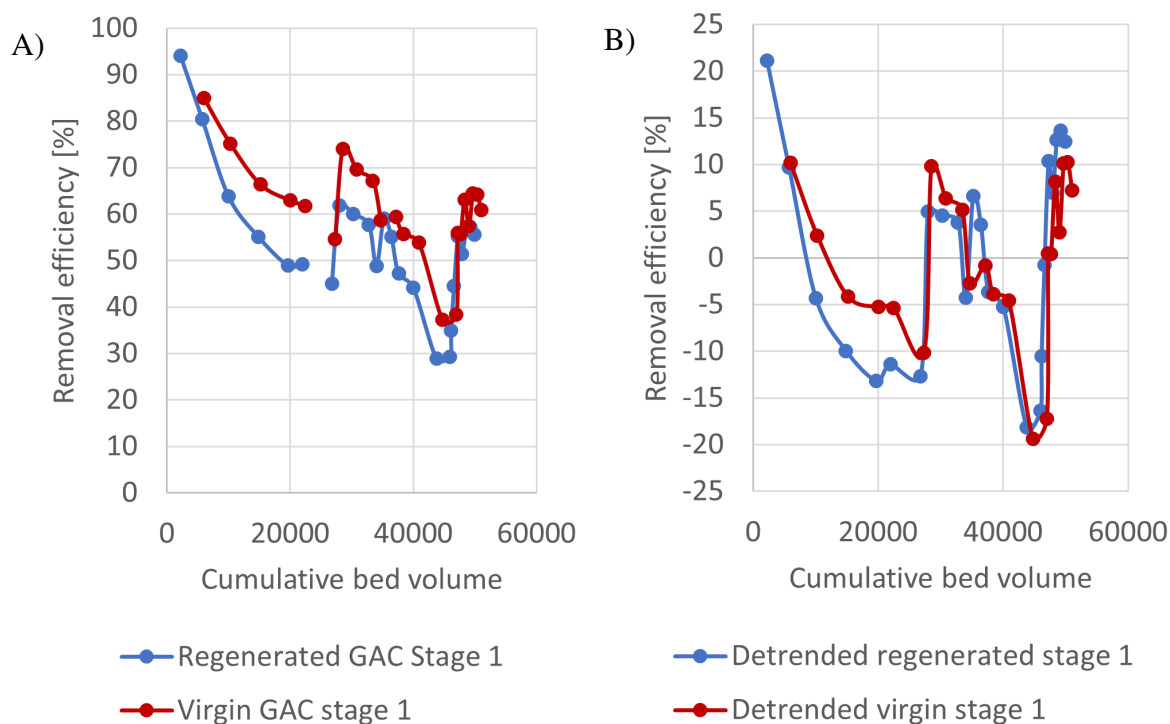


Figure 10: An illustration of the (A) actual removal efficiency of the regenerated filters and the virgin filters and (B) the detrended series used for the T-test.

The detrended series was made by subtracting each data point from its trendline. The results from the T-test can be seen in table 8. The difference between values in detrended virgin GAC and the detrended regenerated GAC for each PFAS was executed in the T-test. The difference for the removal efficiency was determined to not be of significance which can be seen in table 8 where none of the p-values goes below 0.05.

Table 8: P-values from the T-test of the detrended removal efficiencies for the regenerated vs virgin GAC for stage 1 and stage 2 efficiencies for each PFAS in the table.

	PFPeA	PFHxA	PFHpA	PFOA	PFBS	PFPeS	PFHxS	PFOS
Stage 1	1.00	1.00	1.00	0.61	0.84	1.00	1.00	0.87
Stage 2	0.77	1.00	0.94	0.90	0.84	1.00	0.70	0.84

4.3 Modeling of PFASs and PFCAs

The Lin & Huang adsorption model (1999) was used for the PFASs present in the water. The PFASs can be seen in figure 11. It can be seen that all the PFASs have linear trends for the three EBCTs of 5, 8 and 15 minutes. The EBCT of 5 minutes is also separated into phase 1 and phase 2 as was done by McCleaf et al. (2017).

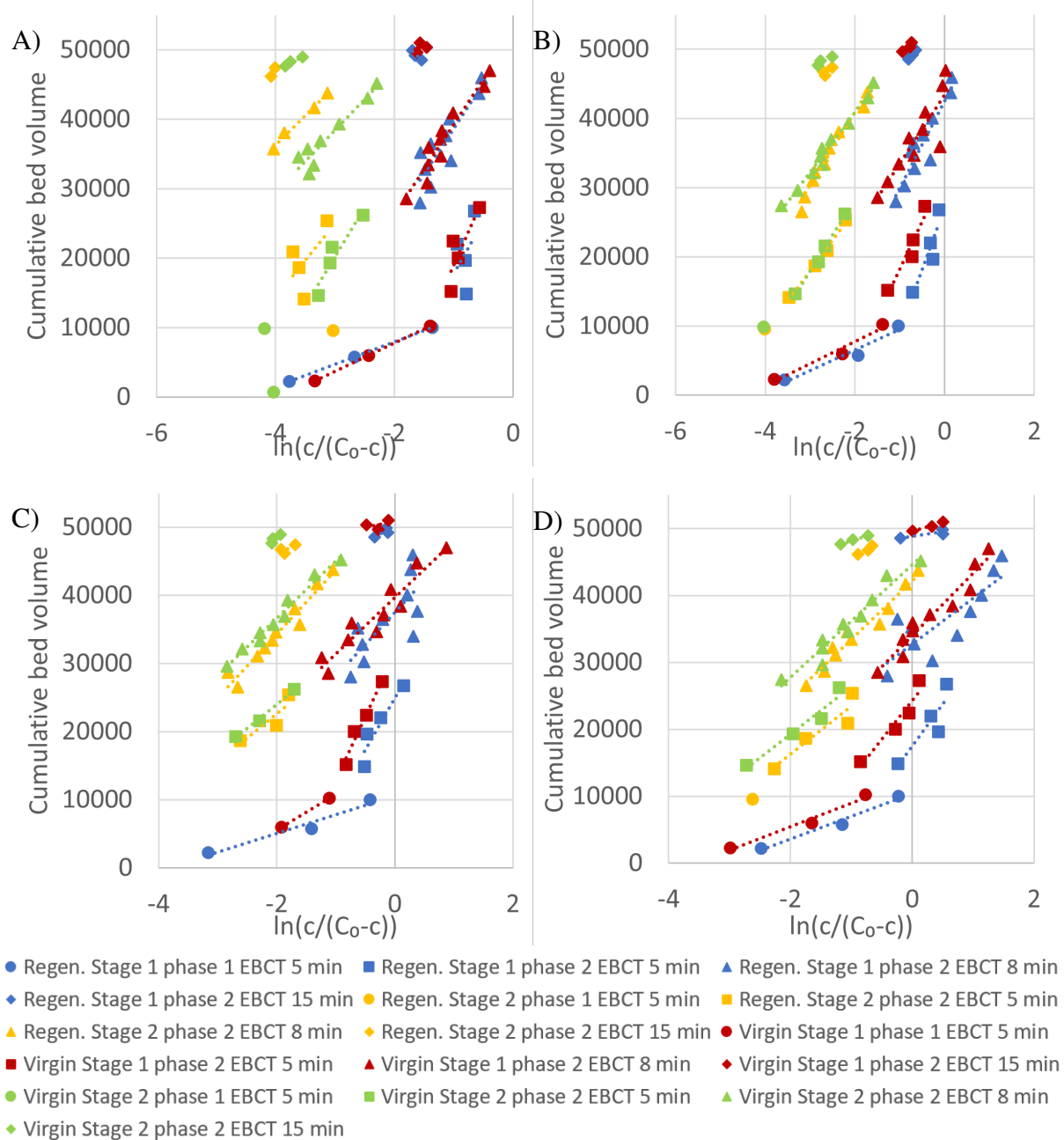


Figure 11: (A) PFOS, (B) PFHxS, (C) PFPeS and (D) PFBS. Blue is regenerated stage 1, yellow is regenerated stage 2, red is virgin stage 1 and green is virgin stage 2. Circle dots are EBCT 5 min phase 1, squares are EBCT 5 min phase 2, triangles are EBCT 8 minutes and dimonds are EBCT 15 min.

The PFCA's Lin & Huang adsorption model (1999) can be seen in figure 12. The model results looks similar to the PFSA's for PFOA but not for the other PFCA's. It can be seen that when the incoming concentration is less than the outgoing concentration, the value of $\ln \frac{C}{C_0-C}$ is zero. This is especially prominent in figure 12D for the shortest chained PFCA where there are many points equal to zero from 30000 BVs and above.

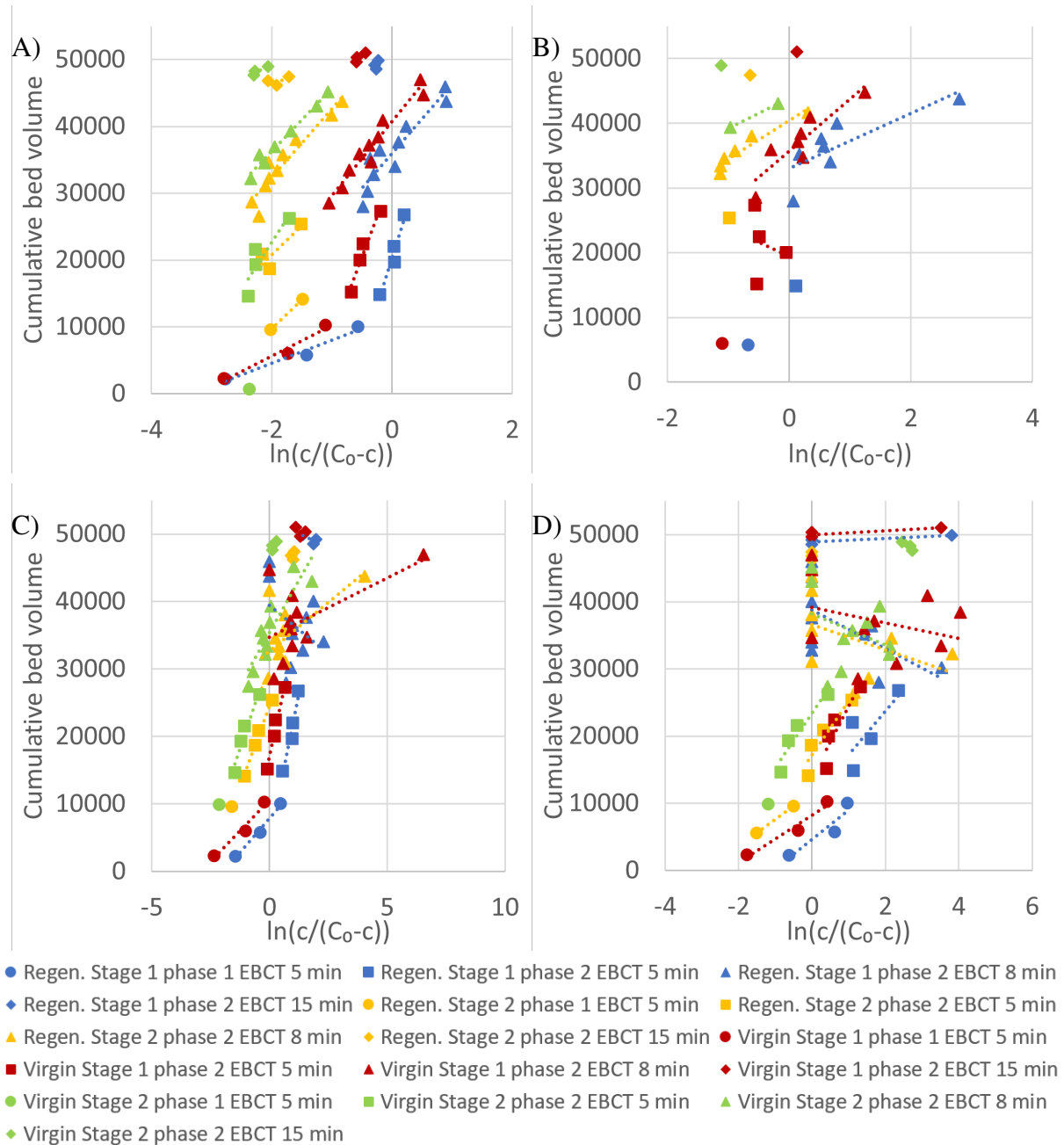


Figure 12: (A) PFOA, (B) PFHpA, (C) PFHxA and (D) PFPeA. Blue is regenerated stage 1, yellow is regenerated stage 2, red is virgin stage 1 and green is virgin stage 2. Circle dots are EBCT 5 min phase 1, squares are EBCT 5 min phase 2, triangles are EBCT 8 minutes and diamonds are EBCT 15 min.

4.4 Removal per weight of GAC per BV for 1 stage and 2 stages

The removal of PFSA and PFCA for 1 stage and 2 stages were compared for the three EBCT periods (figure 13). The removal per weight of GAC per BV for 1 stage is shown in the blue bars and the removal for 2 stages are shown in the grey bars. It can be seen in that the grey bars are never higher than the blue bars indicating that the removal is not more efficient for a two stage system than for a one stage system per mass of GAC. In the figure, the height of each bar is directly correlated to the amount of that PFAS in the raw water and the highest removal can therefore be seen for PFHxS (C_6) in figure 13A, 13B and 13C.

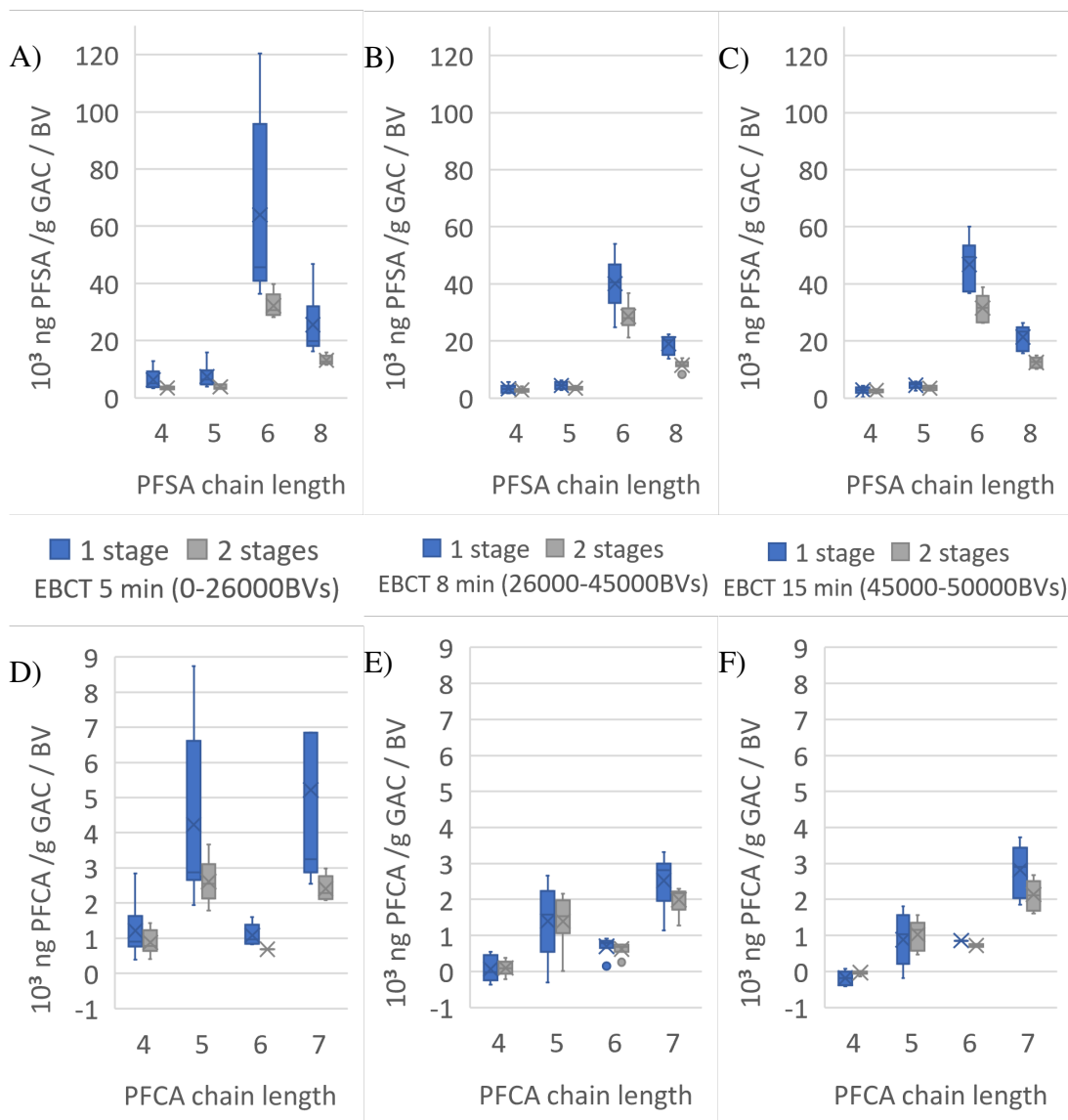


Figure 13: (A) PFSA removal EBCT 5 min, (B) PFSA removal EBCT 8 min, (C) PFSA removal EBCT 15 min (D) PFCA removal EBCT 5 min, (E) PFCA removal EBCT 8 min and (F) PFCA removal EBCT 15 min. EBCT 5 min represents 0-26000 BVs, EBCT 8 min represents 26000-45000 BVs and EBCT 15 min represents 45000-50000 BVs. Blue is removal in stage 1 and grey is the total removal. The removal is represented by ng PFAS removed/ g GAC / BV so the the total removal (grey) is divided by the double weight of GAC.

4.5 Economic comparison of 1 stage to 2 stages for PFAS₄

The Lin & Huang (1999) modeling of PFAS₄ can be seen in figure 14. Figure 14A shows the linear fitting for the virgin GAC for stage 1 and stage 2. For EBCT 5 minutes, stage 1 is separated into phase 1 (circles) and phase 2 (squares). Stage 2 however, only contains phase 2 because there are too few data points for phase 1. From the linear trends, the parameters k_c and τ can be found where k_c is 1/slope and τ is the intercept with the y-axis. These are then used according to equation 1 to generated figure 14B where the modeled concentrations and the real concentrations can be seen. The intercept of the modeled concentrations with 2, 4, 6, 8 and 10 ng/L can be seen in figure 14B.

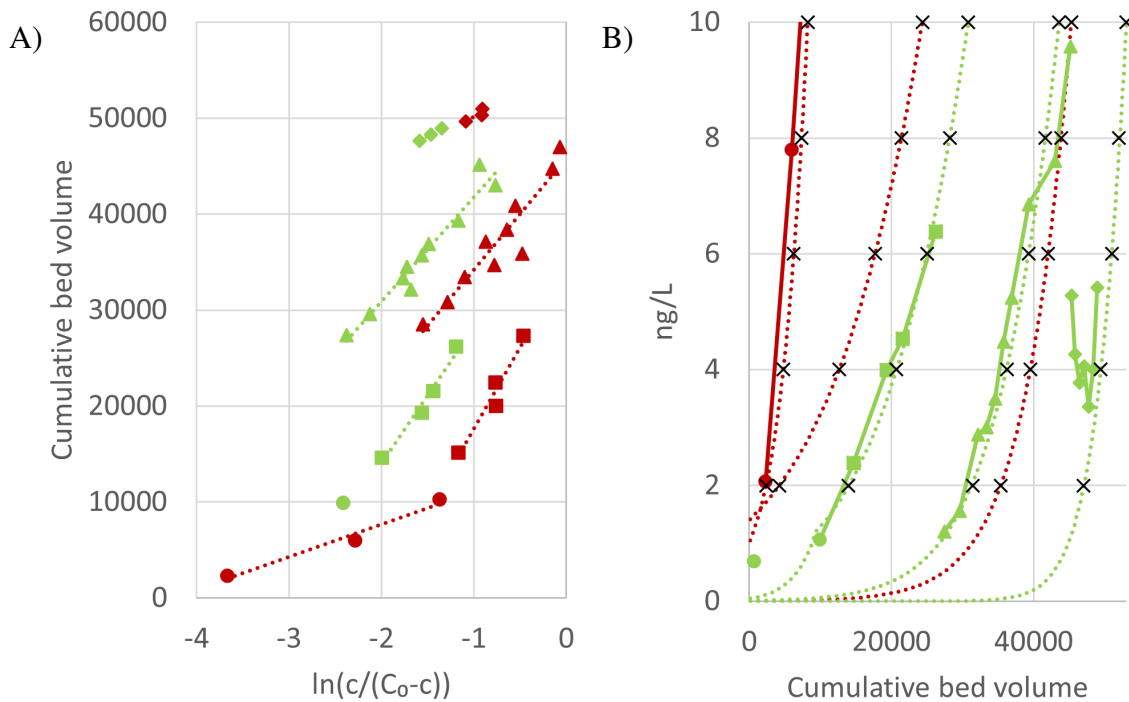


Figure 14: (A) The Lin & Huang linear fitting (1999) of PFAS₄ for the virgin GAC where red is stage 1 and green is stage 2. Circle dots are EBCT 5 min phase 1, squares are EBCT 5 min phase 2, triangles are EBCT 8 minutes and diamonds are EBCT 15 min. (B) The modeled concentrations (dotted lines), the real concentrations (solid line) and the intercepts of 2, 4, 6, 8, 10 ng/L (X marks). In the figure, EBCTs of 5, 8 and 15 minutes can be seen from left to right for the first stage (red) and the second stage (green) individually.

The modeling parameters τ and k_c for the stage 1 and two regenerated and virgin GAC can be seen in table 9. It can be seen that no values are presented for phase 1 in the second stage. That is because there are too few data points. It can furthermore be seen that the R^2 values for the EBCT of 5 and 8 minutes lies between 0.72-0.97. For the EBCT of 15 minutes, there is one value within that range which is that for the regenerated stage 1, the other values are between 0.21 and 0.55.

Table 9: Values of τ and k_c used for modeling of concentrations seen in figure 14B. Also $1/k_c$ is presented as well as the trendline R^2 value together with the number of points included and the total number of points within that period.

EBCT (min)	Phase	$1/k_c$ (BVs)	k_c ($BVs^{-1} \cdot 10^3$)	τ (BVs)	R^2	N.O. points included / total points
Regenerated stage 1						
5.1	1	3164	0.32	13046	0.97	3 / 4
5.1	2	23917	0.04	31438	0.87	4 / 5
8.0	2	12949	0.08	44526	0.83	10 / 10
14	2	12155	0.08	60148	0.97	3 / 7
Virgin stage 1						
4.9	1	3408	0.30	14511	0.97	3 / 4
4.9	2	16819	0.06	34528	0.94	4 / 5
7.8	2	10822	0.09	45302	0.84	10 / 10
14	2	3608	0.29	53835	0.55	3 / 7
Regenerated stage 2						
5.3	1	-	-	-	-	-
5.3	2	9996	0.10	36757	0.72	4 / 5
8.3	2	11884	0.08	52611	0.93	10 / 10
15	2	2596	0.49	50411	0.21	3 / 7
Virgin stage 2						
5.1	1	-	-	-	-	-
5.1	2	14363	0.07	42520	0.97	4 / 5
8.1	2	10869	0.09	52682	0.94	10 / 10
15	2	4745	0.26	55191	0.53	3 / 7

The relationship between τ and k_c for different EBCTs for the first and second stage of the virgin and regenerated columns can be seen in figure 15 and in table 10. It can be seen that there is a strong linear trend for all the parameters except for the k_c of the regenerated first stage and τ for the regenerated second stage.

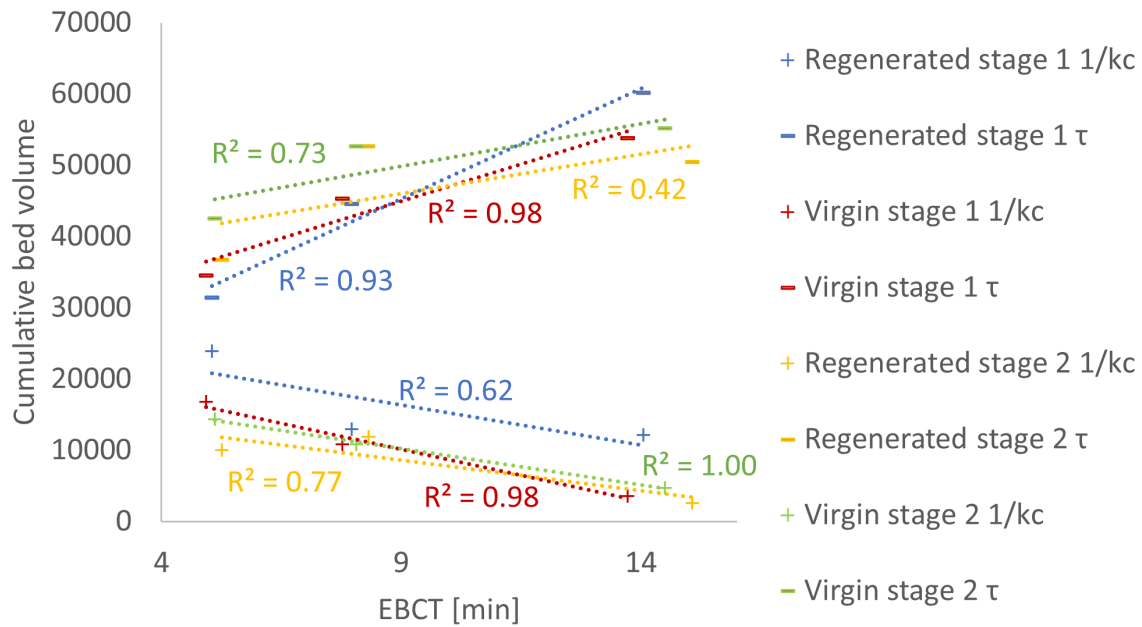


Figure 15: The linear relationship of τ and $1/k_c$ over EBCT for the regenerated and virgin stage 1 and 2. Only Phase 2 is visualized in this figure where "+" stand for $1/k_c$ and "-" stand for τ .

Table 10: R^2 values for the regenerated and virgin stage 1 and stage 2.

Stage 1	$1/k_c (R^2)$	$\tau (R^2)$	Stage 2	$1/k_c (R^2)$	$\tau (R^2)$
Regenerated	0.62	0.98	Regenerated	0.77	0.42
Virgin	0.98	0.93	Virgin	1.00	0.73

The result from comparing the modeled concentrations to the real concentrations can be seen in figure 16. The real values and the modeled values are matching for EBCT 5 min and EBCT 8 minutes. For the EBCT of 15 min, the modeling is suppose to match the three last data points. However, the pumps in the second stage were set to a slightly lower flow making the cumulative BV calculation lower than what was modeled.

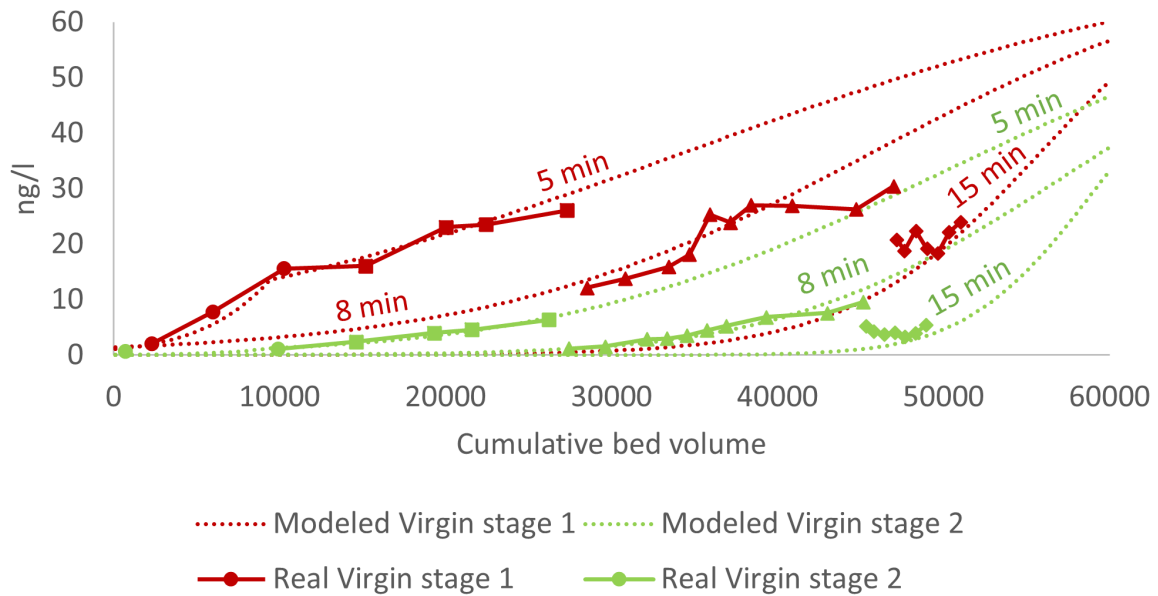


Figure 16: The modeled concentrations and the real concentrations for the whole period. "5 min", "8 min" and "15 min" stand for each corresponding EBCT for the stage 1 and stage 2 concentrations. For the real concentrations, circle dots are EBCT 5 min phase 1, squares are EBCT 5 min phase 2, triangles are EBCT 8 minutes and diamonds are EBCT 15 m. The red color stands for virgin stage 1 and the green for virgin stage 2.

A comparison of the number of needed BVs per regeneration cycle explained in figure 6 can be seen in table 11. The number of bed volumes are presented for the virgin and regenerated cost calculation and for each treatment goal. It can be seen that there are no BVs for the treatment goal of 2 ng/L in the EBCT 8 minutes stage 1. This is because the modeling for this stage and EBCT never reached below 2 ng/L. The number of BVs in 2 stages are higher than for 1 stage for the EBCT of 5 minutes for all the treatment goals. For the EBCT of 8 minutes the number of BVs are higher for the second stage for the regenerated GAC but not for the virgin GAC which has a lower number of BVs for the treatment goals of 8 and 10 ng/L. For the EBCT of 15 minutes the only treatment goal with more BVs per regeneration cycle is 2 ng/L for the regenerated GAC. All of the other treatment goals in the regenerated and virgin GAC calculations has more BVs in the first stage. A decrease in the number of BVs with increasing treatment goal is unexpected. The reason why this is occurring is because the distance in BVs between 1 stage and 2 stages is decreasing with increased treatment goals. This can be seen in figure 14B where the distances between the "X" for 1 stage and 2 stages is getting smaller when going from 2 ng/L to 10 ng/L.

Table 11: BVs between each regeneration cycle for each treatment goal, stage and EBCT.

Treatment goal (ng/L)	EBCT 5 min		EBCT 8 min		EBCT 15 min	
	1 stage (BVs)	2 stages (BVs)	1 stage (BVs)	2 stages (BVs)	1 stage (BVs)	2 stages (BVs)
Regenerated GAC						
2	1720	10300	-	28700	16400	29000
4	4000	15000	6890	34200	25200	21700
6	5380	17600	12600	37600	30600	17300
8	6390	19900	16900	40200	34500	14200
10	7190	21500	20200	42500	37600	11900
Virgin GAC						
2	2340	11600	4210	27200	35400	11600
4	4780	15900	12600	23600	39500	9900
6	6260	18700	17700	21600	42000	9000
8	7340	20900	21400	20200	43800	8170
10	8200	22600	24300	19200	45300	7710

In figure 17 the cost per weight of GAC for reaching the treatment goal of 2, 4, 6, 8 and 10 ng/L. Figure 17A and 17B contains the cost per weight of GAC for the EBCT of 5 minutes for a 1 stage and a two stage GAC for the regenerated GAC and the Virgin GAC respectively. Figure 17C and 17D contains the cost per weight of GAC for the EBCT of 8 minutes and figure 17E and 17F for the EBCT of 15 minutes. It can be seen in figure 17A, 17B, 17C and 17D that the cost of operating a two stage GAC is cheaper than operating a one stage GAC system for the EBCT of 5 and 8 minutes for both the regenerated GAC and the virgin GAC. However, for the EBCT of 15 minutes it is more expensive to operate a two stage GAC for the whole range of treatment goals in figure 17F for the virgin GAC. For the regenerated GAC with EBCT of 15 minutes in figure 17E, it is cheaper to operate a two stage system for the treatment goals 2-6 ng/l but more expensive for the treatment goals 8-10 ng/L. It can be noted that the costs for the second stage virgin GAC EBCT 8 minutes in figure 17D and both the regenerated and virgin second stages in figure 17E and 17F the the cost is decreasing with decreasing treatment goal.

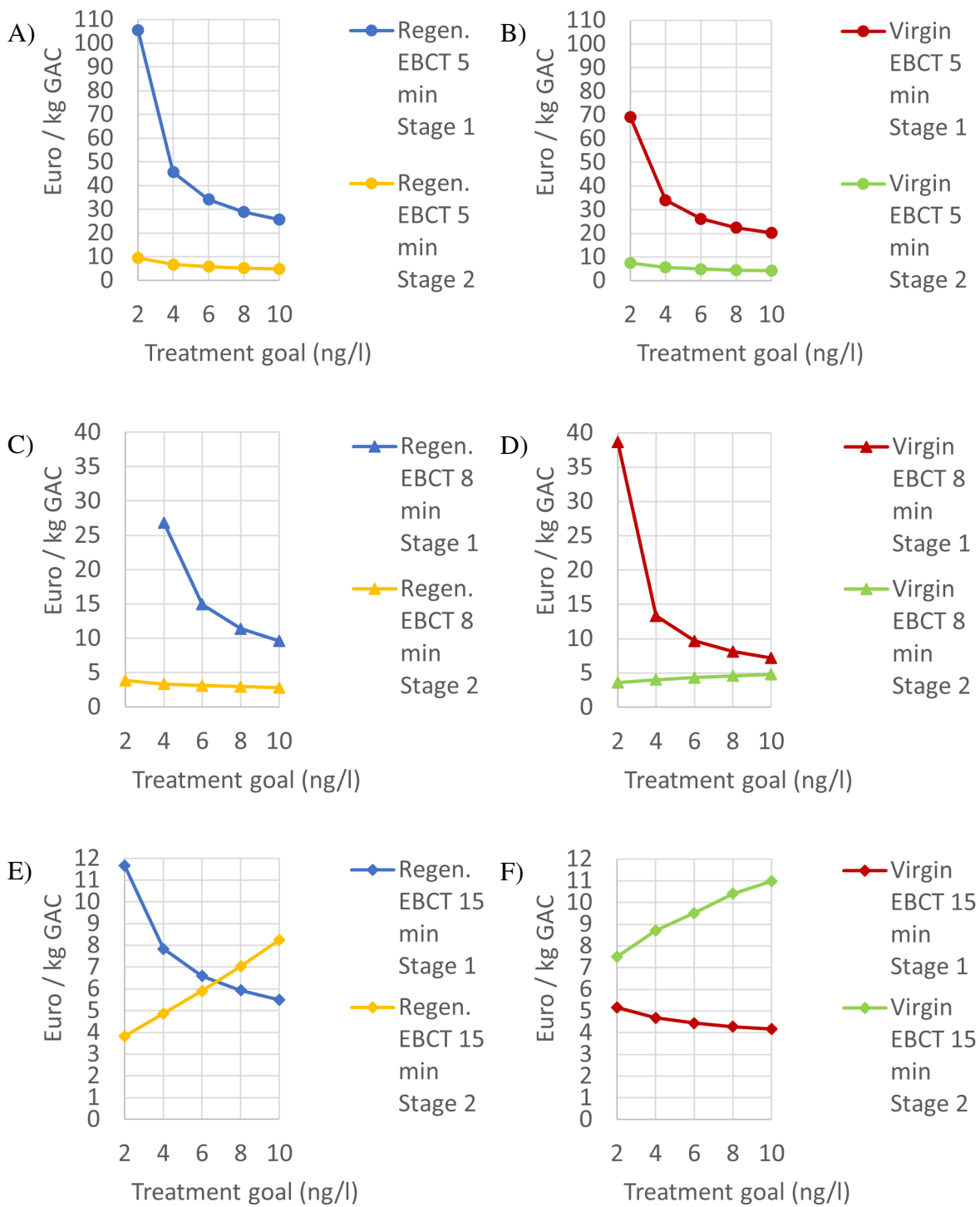


Figure 17: The cost of reaching a PFAS₄ target concentration in the range 2-10 ng/L for (A) Regenerated GAC EBCT 5 min (B) Virgin GAC EBCT 5 minutes (C) Regenerated GAC EBCT 8 minutes (D) Virgin GAC EBCT 8 minutes (E) Regenerated GAC EBCT 15 minutes and (F) Virgin GAC EBCT 15 min.

The cost / weight of GAC for 1 stage and 2 stages can also be seen in table 12. Also the % cost difference between 1 stage and 2 stages can be seen. It can be seen that during EBCT 5 min the reduced cost was between 81%-91% and 79%-89% for the regenerated and virgin GAC calculations respectively. For the EBCT of 8 minutes, it was between 71%-88% and 33%-91% for the regenerated and virgin GAC calculations respectively. It needs to be noted however, that there is no cost calculation for the treatment goal of 2 ng/L for the regenerated GAC because the Lin & Huang (1999) modeling never went below 2 ng/L. During EBCT 15 minutes the difference between 1 and 2 stages was between 11%-67% for the regenerated GAC calculations and the treatment goal range of 2-6 ng/L. For the range 8-10 it was 16%-33% cheaper with two stages compare to one. For the EBCT of 15 minutes and virgin GAC calculations the cost was 31%-62% cheaper with 1 stage compared to 2 stages.

Table 12: Costs in € / kg GAC for reaching a treatment goal of 2-10 ng/L in 1 stage or 2 stages as well as the % difference between 1 stage and 2 stages. The % difference was calculated using equation 6.

Treatment goal (ng/L)	Regenerated GAC					Virgin GAC				
	2	4	6	8	10	2	4	6	8	10
EBCT 5 min										
1 stage € / kg GAC	105	46	34	29	26	69	34	26	22	20
2 stages € / kg GAC	9.5	6.7	5.8	5.2	4.9	7.5	5.7	4.9	4.5	4.2
% difference 1-2 stages	91	85	83	82	81	89	83	81	80	79
EBCT 8 min										
1 stage € / kg GAC	- ^a	27	15	11	9.6	39	13	9.7	8.1	7.2
2 stages € / kg GAC	3.8	3.3	3.1	3.0	2.8	3.6	4.0	4.3	4.6	4.8
% difference 1-2 stages	-	88	79	74	71	91	70	55	43	33
EBCT 15 min										
1 stage € / kg GAC	12	7.8	6.6	5.9	5.5	5.2	4.7	4.4	4.3	4.2
2 stages € / kg GAC	3.8	4.9	5.9	7.0	8.3	7.5	8.7	9.5	10	11
% difference 1-2 stages	67	38	11	16 ^b	33 ^b	31 ^b	46 ^b	53 ^b	59 ^b	62 ^b

^a The modeling could not be done for this treatment goal.

^b For the calculation of these % numbers, the calculation using equation 6 was flipped because the second stage was more expensive than the first stage. This shows how much cheaper the 1 stage is compared to 2 stages.

5 Discussion

5.1 Raw water and PFAS₂₁

The three most abundant PFASs in the raw water were PFHxS, PFOS and PFPeS and can be seen in table 7. The same result was also found by Franke et al. (2021). The average PFAS₂₁ concentration in the raw water was below the upcoming limit in drinking water of 100 ng/L but occasional samples were above as can be seen in figure 8. The average PFAS₂₁ concentration was 91 ng/L which is 9 ng/L below the limit. The average PFAS₄ concentration was 71 ng/L and is 67 ng/L above the limit of 4 ng/L. Therefore it is much more difficult to reach the PFAS₄ limit than the PFAS₂₁ which is already met in the raw water average concentration. It can furthermore be noted that the PFASs that makes up PFAS₄ contributes to the majority(78%) of the PFAS₂₁ concentration. Meaning that if the PFAS₄ concentration gets lower using GAC, so does the majority of PFAS₂₁. Therefore, the focus in this report on the economic calculation was put towards PFAS₄.

5.2 Removal efficiencies

The removal efficiency is higher for long chained PFASs than short chain PFASs and also higher for PFASs than for PFCAs which is shown in figure 9. This is in line with the findings by McCleaf et al. (2017). However, even though PFBS experienced desorption at one sample in the first stage, which can be seen in figure 9, none of the other PFASs experience desorption. For the PFCAs, the shortest chained, PFPeA, experienced desorption in the first stage and second stage after 31000 BVs and PFHxA after 42000 BVs. This can be compared to McCleaf et al. (2017) who found desorption of PFPeA after 37000 BVs in a one stage GAC with an EBCT of 6.1 minutes.

The removal efficiency seems to follow the same pattern for the first and second stage for the PFASs and the PFCAs. This can be seen in all the figures in figure 9 but is extra clear for PFOA and PFOS in the first stage. A straight line of decreasing removal efficiencies for the first EBCT until 26000 BVs and after that a rapid increase in removal efficiencies followed by a new decrease in a less steep straight line until 45000 BV which is at the end of the EBCT of 8 minutes. After that a rapid increase in removal efficiencies followed by an unchanged or slow decrease in removal efficiency. This pattern was confirmed by Belkouteb et al. (2020) for the longer chained PFASs when the flow was changed from a faster flow to a slower flow.

With increasing BVs, the removal efficiency is still relatively high (>55% for PFOA and >80% for PFOS at the end of the experiment) in the first and second stage for the longest-chained PFCA and PFSA. This can be compared to the removal efficiencies of the shortest chained PFCA and PFSA which are 0% for PFPeA and 40-50% for PFBS. The removal efficiency seems to decrease with decreasing chain length for increasing treated BVs. This is in agreement with McCleaf et al. (2017) who found that for both the PFCAs and the PFASs, the compound with longer carbon chain showed a smaller decrease in removal efficiency over the course of the experiment.

The removal efficiency is not significantly different in regenerated GAC compared to virgin GAC, which is shown by the p-values from the T-test in table 8. The differences between the virgin and

regenerated GAC were investigated in the sieving analysis. The effective size (the 10% diameter) of the regenerated GAC was 0.67 mm and for the virgin GAC it was 0.74 and 0.75 mm. This means that the distribution of particle sizes was towards slightly more smaller particles in the regenerated GAC. The Uniformity coefficient (60% diameter / 10% diameter) was 1.74 for the regenerated GAC and 1.74 and 1.72 for the virgin GAC. These numbers are very close indicating that the difference between the regenerated GAC and the virgin GAC is very small. This is in line with the finding in Son et al. (2010) that the adsorption capacity is equal in virgin and regenerated GAC. Therefore, the average removal of the virgin and the regenerated can be used to compare the removal in a one stage system to a two stage system as was done in figure 13.

5.3 Modeling of PFASs and PFCAs

The Lin & Huang adsorption model (1999) can be used with success for PFOA, PFOS, PFHxS, PFPeS and PFBS which can be seen in figure 12A, 11A, 11B, 11C and 11D. However, for PFHpA, PFHxA and PFPeA it cannot be used accurately because the trendlines are not right which can be seen in figure 12B, 12C and 12D. In figure 12C and 12D it can be seen however, that the trendlines are accurate for the EBCT of 5 minutes for PFPeA and PFHxA and therefore the modeling could be done for that EBCT. In figure 12B however, it can be seen that the modeling could not be done for any EBCT. This can be related to McCleaf et al. (2017) who could model PFPeA, PFHxA, PFHpA, PFOA, PFBS, PFHxS and PFOS with an EBCT of 6.1 minutes.

It can be seen in figure 12B, 12C and 12D that when the the removal efficiency is close to zero but positive, the $\ln \frac{C}{C_0-C}$ gets too high (values to the far right in the figures). Also, when desorption occurs $\ln \frac{C}{C_0-C}$ gets zero proving that the model does not work for PFASs that are desorbing PFASs. This proves that the modeling need adjustments in order to work around the occurring. One way could be to exclude the desorption as was done by Chow et al. (2022) and another way would be to come up with a model taking the desorption into account. For PFHpA there is furthermore to few data points for the model to be accurate. The Lin & Huang adsorption model (1999) is developed to model a substance adsorbing to some kind of adsorption media but can be used to model the adsorption of PFAS₄ which is proved by figure 16. However, when desorption occurs, or when the removal efficiency is positive but close to zero it does not work.

5.4 Removal per weight of GAC per BV for 1 stage and 2 stages

When comparing the removal of PFAS per weight of GAC per BV in figure 13 it can be seen that the removal is never higher in the two stage GAC compared to the one stage GAC. This shows that a two stage GAC is not more efficient in removing PFASs than a one stage GAC. However, the difference between the one stage GAC (blue) and the two stage GAC (grey) seems to decrease with an increase in EBCT and more BVs treated. The scale in figure 13A, 13B and 13C is higher than the scale in figure 13D, 13E and 13F which is because the removal of PFASs is much higher (more than 10 times) the removal of PFCAs. This is because they are more abundant in the raw water which can be seen in table 7 but could also be partly explained by the higher removal efficiency for PFASs than for PFCAs.

Among the PFASs the removal was highest for PFHxS (C_6) during the whole period. An explanation for this can be that the raw water concentration of PFHxS is the highest among all the PFASs which can be seen in table 7. It can be seen in table 7 that the PFCAs with the highest concentrations in the raw water are PFHxA (C_5) and PFOA (C_7). In figure 13D the removal is the highest for those two among the PFCAs in the EBCT of 5 minutes. However, in figure 13E and 13F the removal of PFHxA (C_5) is decreasing relative to PFHpA (C_6) and PFOA (C_7) indicating that the removal of the shortest chained PFCAs gets worse than for the long chained PFCAs the longer the experiment runs. This can furthermore be confirmed by earlier findings in this report.

5.5 Economic comparison of 1 stage to 2 stages for PFAS

For the EBCT 5 minutes it is 81-91% and 79-89% cheaper to operate a two stage GAC system for the range of treatment goals of 2-10 ng/L and for the EBCT 8 minutes it is 71-88% and 33-91% cheaper. This can be seen in table 12. For the EBCT of 15 minutes however, the two stage is only cheaper (11-67%) for 2-6 ng/L with the regenerated GAC. For the virgin GAC it is not cheaper within the range of 2-10 ng/L. It was 16-33% cheaper to operate a 1 stage system of the EBCT of 15 minutes and the range of treatment goals of 8-10 ng/L with the regenerated GAC and for the range of 2-10 ng/L with 31-62%.

A reason why it was cheaper to operate a 2 stage GAC compared to 1 stage GAC for the EBCT of 5 and 8 minutes lies with the number of BVs between each regeneration. The numbers were higher for 2 stages compared to 1 stage which can be seen in table 11. However, increasing the number of bed volumes means that the removal efficiency in the first and second stage decreases even more. It could be seen in figure 9 that with increasing BVs treated, the removal efficiency decreased more with shorter chain length, which was also confirmed in McCleaf et al. (2017). Therefore, an increased number of BVs between each regeneration will lead to lower adsorption capacities for short chained PFASs. This will then lead to more short-chained PFASs being released into the drinking water even if the PFAS₄ limit will be met.

The actual full scale DWTP has an EBCT of 15 minutes and for the limit on 4 ng/L of PFAS₄ it would be cheaper with a two stage GAC for the results of the regenerated GAC (5.9 € / kg GAC) but more expensive with the virgin GAC (9.5 € / kg GAC). However, there are a number of sources of errors related to this. On the one hand, the regenerated GAC better represents the continuous regeneration cycles presented in the concept in figure 6 that would apply since the GAC implemented at the plant would always be regenerated GAC. However, in the economic calculation, virgin GAC is bought at the start of implementing a one stage or two-stage GAC system and therefore the virgin GAC calculations better apply for the first regeneration cycle. Moreover, the concept itself is not validated and needs to be confirmed before these calculations can be fully trusted.

It can be seen in figure 4 that the first 4 samples of PFAS₄ after the first and second stage at EBCT 15 minutes have generally decreasing concentrations over time and BV. For the last three samples of EBCT 15 minutes after the first and second stage however, there is an increase in concentrations over time and BV. This is an indication that the GAC needs a number of BVs before it reaches the mass transfer equilibrium for the new flow. When the flow was changed

from having an EBCT of 5 minutes to 8 minutes this period of decreasing concentrations could not be seen in the figure. An explanation for this could be that the needed number of BVs to reach the new mass transfer equilibrium was lower when going from an EBCT of 5-8 minutes than going from an EBCT of 8-15 minutes. Furthermore, the period of decreasing concentrations between EBCT 5-8 minutes was never visualized in the figure because the time between the samples was too long.

If the increase in concentration for the last three samples of PFAS₄ after the first and the second stage is a result of the GAC reaching the new mass transfer equilibrium this would mean that this represents the increase for these BVs that would have occurred if the EBCT of 15 minutes would have been for the whole experiment. However, it cannot be fully confirmed that this is the case. It could also be a result of the PFAS₄ raw water concentration increasing during those three samples. It can be seen during the whole period in figure 4 that when the concentration in the raw water is increased for a point, such as the concentration in the 5th to last point, the concentrations after the first and second stage also increase slightly. Since the Lin & Huang adsorption modeling (1999) for EBCT 15 minutes is only based on the last three samples of EBCT 15 min, it is therefore unsure if the behaviour of the GAC at the EBCT of 15 minutes actually would behave as it is shown in figure 16.

The decreasing costs per weight of GAC with decreasing treatment goals seen in figure 17E and 17F are unreasonable. Therefore, any conclusion drawn from observing those graphs will be flawed. In order to confirm or disregard those conclusions a new 2-stage pilot study with a longer period of EBCT 15 minutes needs to be carried out. Also, a 2-stage pilot study that runs for a while and then replaces the first filter with the second filter and replaces the newly moved second filter with a newly regenerated filter needs to be done in order to confirm that the concept explained in figure 6 developed in this thesis for the economic calculation is accurate in real life.

6 Conclusion

The first aim of this Master's thesis was to compare the removal of long and short chain PFAS at a two stage GAC pilot to a one stage pilot. The second aim was also to compare the costs of operating a full scale one stage GAC to a two stage GAC at a DWTP with respect to different treatment goals for the EBCT of 5, 8 and 15 minutes. This was done by concluding that the limit on PFAS₄ concentration was the most difficult to reach and therefore the economic calculation was done on reaching the PFAS₄ limit and not on reaching the PFAS₂₁ limit.

The removal efficiency was compared for the PFASs and the PFCAs in the first and second stage. It was concluded that the removal efficiency decreases the shorter the chain length and the more BVs gets treated in both the first and the second stage. It was determined that the removal efficiency for the regenerated and the virgin GAC was not significantly different by detrending the series and comparing them with a T-test.

The modeling using The Lin & Huang adsorption model (1999) of the PFASs and the PFCAs was made for the average first and second stage for the regenerate and the virgin GAC for the three EBCTs. The model worked good for the all PFASs and PFOA but not for the shorter chained PFCAs. This lead to the conclusion that the model works well for all PFASs until desorption occurs.

The removal of PFAS per weight of GAC with respect to chain lengths of PFASs and PFCAs in a 1 stage and a 2 stage GAC was performed for the three EBCTs in the experiment. The removal per weight of GAC per BV for the PFASs included is not increased in a two stage GAC system compared to a one stage GAC filtration.

The economic calculation was based on the Lin & Huang adsorption model (1999) for PFAS₄. This was done on the average concentrations from the virgin and regenerated GAC for the three EBCTs of 5, 8 and 15 minutes respectively. It was found to be cheaper to operate a two stage GAC filtration compared to a one stage GAC when reaching the target of <4 ng/L for PFAS₄ at both the EBCT of 5 and 8 minutes. For 15 minutes, the cost per weight of GAC was decreasing for the second stage which is an unrealistic result and should be the object of further research. Additional future research should focus on modeling desorption of PFASs in order to fully understand the relationship between the GAC and the short chained PFASs.

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Appendix

The analyzed PFASs, PFAS₄, PFAS 11, PFAS 20 and PFAS 21 can be seen in table A1.

Table A1: PFAS.

	Analyzed PFAS	PFAS ₄	PFAS 11	PFAS 20	PFAS 21
1	PFBA	PFOA	PFBA	PFBA	PFBA
2	PFPeA	PFOS	PFPA	PFPA	PFPA
3	PFHxA	PFHxS	PFHxA	PFHxA	PFHxA
4	PFHpA	PFNA	PFHpA	PFHpA	PFHpA
5	PFOA		PFOA	PFOA	PFOA
6	PFNA		PFNA	PFNA	PFNA
7	PFDA		PFDA	PFDA	PFDA
8	PFBS		PFBS	PFUnDA	PFUnDA
9	PFHxS		PFHxS	PFDoDA	PFDoDA
10	PFOS		PFOS	PFTTrDA	PFTTrDA
11	6:2 FTS		6:2 FTS	PFBS	PFBS
12	PFUnDA			PFPS	PFPS
13	PFDoDA			PFHxS	PFHxS
14	PFTTrDA			PFHpS	PFHpS
15	PFPeS			PFOS	PFOS
16	PFHpS			PFNS	PFNS
17	PFNS			PFDS	PFDS
18	PFDS			PFUnDS	PFUnDS
19	PFUnDS			PFDoDS	PFDoDS
20	PFDoDS			PFTTrDS	PFTTrDS
21	PFTTrDS				6:2 FTS
22	4:2 FTS				
23	8:2 FTS				
24	FOSA				
25	MeFOSA				
26	EtFOSA				
27	MeFOSE				
28	EtFOSE				
29	FOSAA				
30	MeFOSAA				
31	EtFOSAA				
32	HPFHpA				
33	PF37DMOA				
34	PFTeDA				

The removal of PFAS per weight of GAC for the first stage and the second stage can be seen in figure A1.

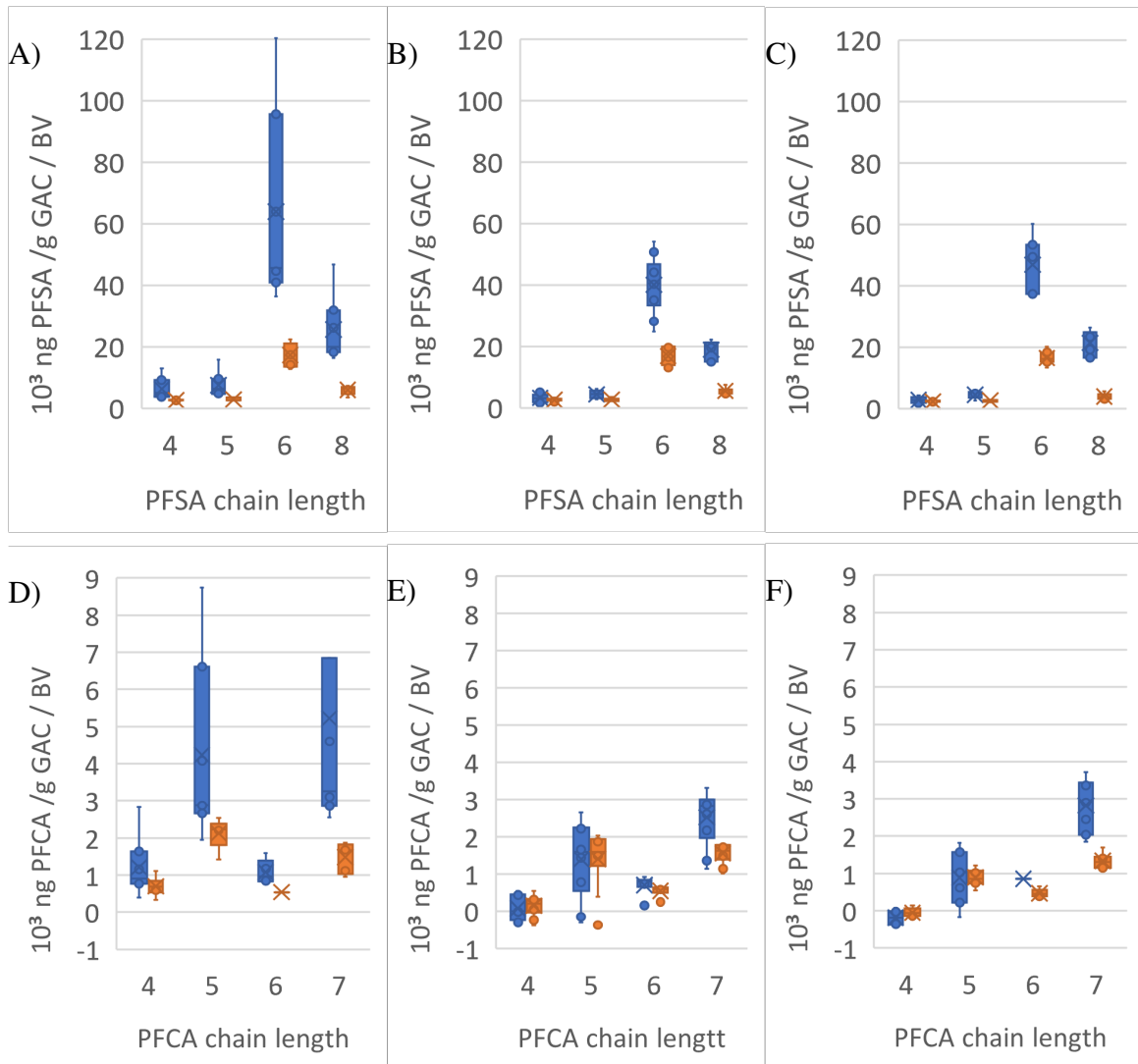


Figure A1: (A) PFSA removal EBCT 5 min, (B) PFSA removal EBCT 9 min, (C) PFSA removal EBCT 15 min (D) PFCA removal EBCT 5 min, (E) PFCA removal EBCT 9 min and (F) PFCA removal EBCT 15 min. EBCT 5 min represents 0-26000 BVs, EBCT 9 min represents 26000-45000 BVs and EBCT 15 min represents 45000-50000 BVs. Blue is removal in stage 1 and orange is removal in stage 2. The removal is represented by ng PFAS removed/ g GAC / BV.

The visualisation for the regenerated filters can be seen in figure A2.

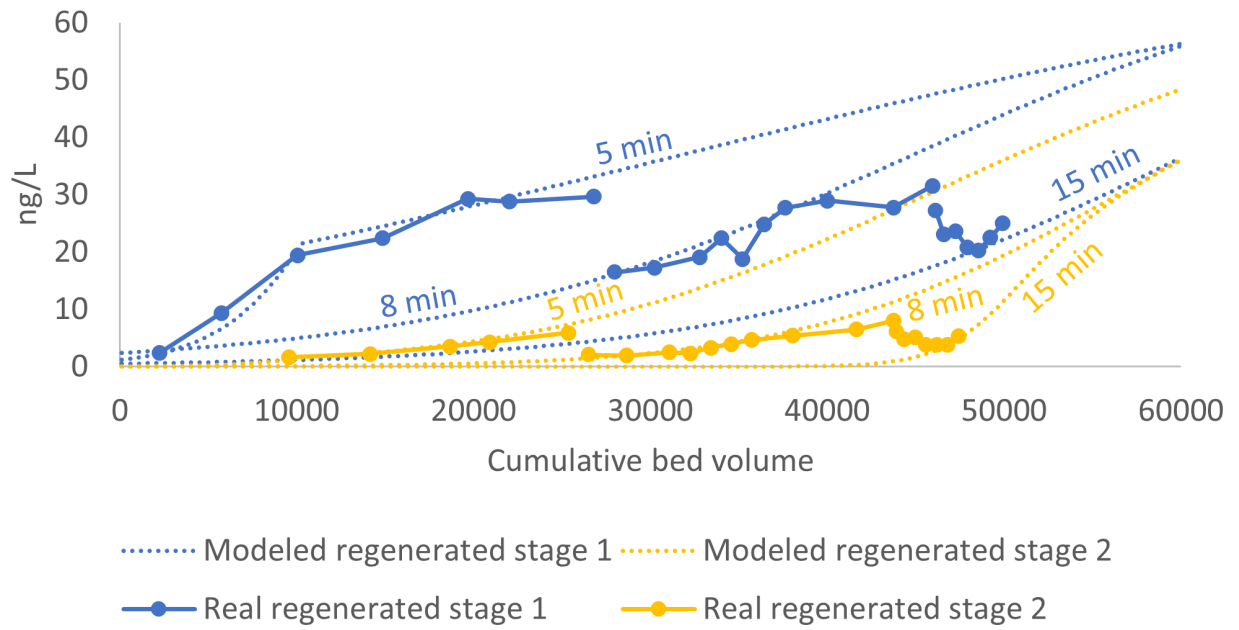


Figure A2: The modeled concentrations and the real concentrations for the whole period.

The linear fitting the modeled concentrations for the regenerated filters can be seen in figure A3

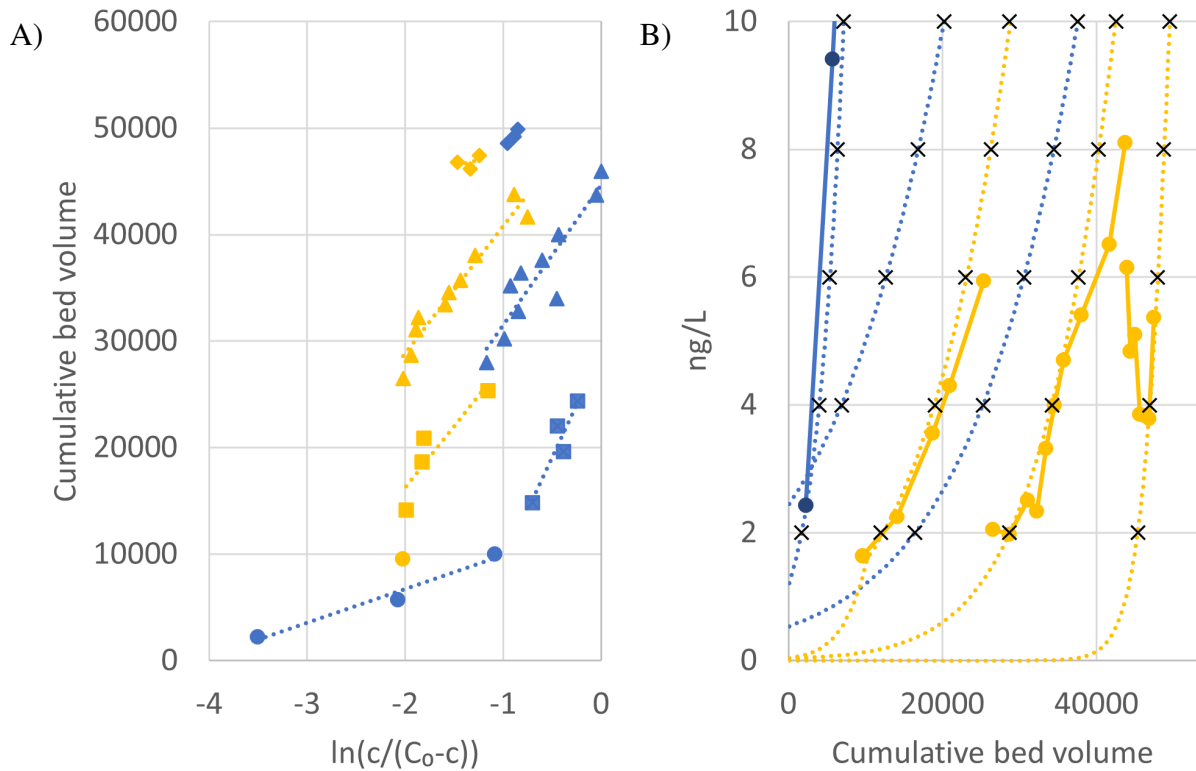


Figure A3: (A) The Lin & Huang linear fitting (1999) for the regenerated GAC where blue is stage 1 and yellow is stage 2. Circle dots are EBCT 5 min phase 1, squares are EBCT 5 min phase 2, triangles are EBCT 9 minutes and diamonds are EBCT 15 min. (B) The modeled concentrations (dotted lines), the real concentrations (solid line) and the intercepts of 2, 4, 6, 8, 10 ng/L (X marks). In the figure, EBCTs of 5, 9 and 15 minutes can be seen from left to right for the first stage (red) and the second stage (green) individually.