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Removal of per- and polyfluoroalkyl substances (PFAS) from contaminated leachate using aeration foam fractionation

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Referat

Rening av per- och polyfluorerade alkylsubstanser (PFAS) från kontaminerat lakvatten med hjälp av skumfraktionering

Axel Krögerström

Lakvatten från deponianläggningar är förorenade på många sätt och per- och polyfluorerade alkylsubstanser (PFAS) är en av dessa föroreningar. Nyligen utförda studier har påvisat människans och naturens negativa inverkan av PFAS. Därmed blir en behandling och rening av lakvattnet viktigt. En teknik för att minska PFAS-koncentrationerna är skumfraktionering med hjälp av luftning.

Hovgården är en deponianläggning nordöst om Uppsala som i tidigare mätningar har uppmätt höga nivåer av PFAS. Tidigare framgångsrika småskaliga experiment har utförts med skumfraktionering som reningsmetod för PFAS-kontaminerat lakvatten men i och med kommande lagar och regleringar av PFAS-koncentrationer uppkommer behovet av att undersöka om en uppskalning är möjlig.

I denna studie har detta undersökts genom att PFAS-kontaminerat lakvatten från deponianläggningen har pumpats in i en plastcylinder med volymen 46 liter och vattnet har luftats med ett flöde på tio liter per minut. Totalt har sex experiment utförts där kontakttiden och skumhalten har varit parametrar som varierats. Fyra experiment gjordes med en kontakttid på tio minuter och en skumhalt på fem, tio, tjugo och trettio procent och två experiment gjordes med skumhalt på fem procent och en kontakttid på tjugo respektive trettio minuter.

Den genomsnittliga Σ PFAS-reduceringen, d.v.s. den procentuella minskningen i Σ PFAS-koncentrationen från råvattnet till det behandlade vatten i de olika körningarna varierade mellan 31 % och 66 %. Reduceringen av PFAS med långa kolkedjor ($\geq C_6$) var högre än reduceringen för korta kolkedjor ($\leq C_5$) även om alla experiment visade på en reduktion av PFAS-koncentrationen. Kolkedjor med en funktionsgrupp bestående av en karboxylsyra heter perfluorkarboxylsyror (PFCA) och motsvarande för funktionsgrupper bestående av sulfonsyra heter perfluoroktansulfonsyra (PFSA). Precursors är PFAS som efter reaktion degraderas till de PFSA och PFCA. Den genomsnittliga reduceringen av PFCA var 48 %, PFSA var 59 % och precursors 78 %. Den högsta reduceringen uppmättes i experimentet med en trettio minuters kontakttid och 5 % skumhalt. Experimentet hade en genomsnittlig Σ PFAS-reduktion på 58 % och en genomsnittlig reduktion på långkedjade PFAS på 92 %. Den lägsta uppmätta reduceringen var på experimentet med en tio minuters kontakttid och tjugo procents skumhalt med en genomsnittlig Σ PFAS-reduktion på 41 % och en genomsnittlig reduktion på långkedjade PFAS på 67 %.

En slutsats som kan dras är att det finns ett samband mellan ökad kontakttid och ökad reduktion men att inga tydliga samband mellan skumhalt och reduktion kan göras. Däremot kan det inte ställas bortom rimligt tvivel att det är just kontakttiden som är den avgörande faktorn. En annan slutsats är att det sker en högre berikning av kortkedjade PFAS i vattnet och långkedjade PFAS i skummet. Generellt så ledde en ökad koncentration av PFAS i råvattnet till en högre PFAS-reduktion.

Nyckelord: PFAS, Skumfraktionering, Lakvatten, Reducering, Vattenrening

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Abstract

Removal of per- and polyfluoroalkyl substances (PFAS) from contaminated leachate using aeration foam fractionation

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Leachate from landfills is contaminated in many ways and per- and polyfluoroalkyl substances (PFAS)-contamination is one of them. Recent studies have demonstrated the environmental and human concerns of PFAS. Therefore, the treatment of leachate is important. One technique to reduce the PFAS concentration is by using aeration and foam fractionation.

Hovgården is a landfill northeast of Uppsala, where previous measurements have shown high levels of PFAS in the leachate. Earlier small-scale experiments using aeration foam fractionation as a treatment technique for PFAS removal have been done successfully, but with upcoming requirements of PFAS concentrations there is a need to investigate whether an upscaling is possible or not.

In this study, this has been investigated by pumping PFAS contaminated leachate from the landfill in to a 0.046 square metre plastic cylinder and aerated the leachate with an airflow of ten L min⁻¹. A total of six experiments were conducted where the contact time and fraction of extracted foam was parameters that was varying. Four experiments were done with a contact time of ten min and foam fraction of five, ten, twenty and thirty percent and two experiments were made with a foam fraction of five percent and a contact time of twenty respectively thirty min.

The average Σ PFAS removal, i.e., the percentage difference in Σ PFAS concentrations between the influent and effluent water in the different runs varied between 31 % and 66 %. The removal of long chained PFAS ($\geq C_6$) was higher than the removal of short chained PFAS ($\leq C_5$) even if all experiments did reduce the PFAS concentration. Carbon chains with a functional group containing a carboxylic acid is called perfluorocarboxylic acids (PFCA) and with a functional group containing a sulfonic acid is called perfluoroalkane sulfonic acids (PFSA). Precursors are PFAS that after a reaction degrades into PFCA and PFSA. The average removal efficiency of PFCA were 48 %, of PFSA 59 % and of precursors 78 %. The highest removal efficiency was discovered in the experiment with a 30-min contact time and five percent foam fraction with an average Σ PFAS removal of 58 % and an average Σ long chained PFAS removal of 92 %. The lowest removal efficiency was discovered in the experiment with a 10-min contact time and 20 percent foam fraction with an average Σ PFAS removal of 41 % and an average Σ long chained PFAS removal of 67 %.

In conclusion a connection between increased contact time and increased removal efficiency was discovered but no clear connections between foam fraction and removal efficiency were found. However, it cannot be stated beyond reasonable doubt that the contact time is the decisive factor. Another conclusion is that the enrichment of short chained PFAS are higher in the water and a higher enrichment of long chained PFAS in the foam. In general, a higher inlet concentration of PFAS in the influent water resulted in a higher removal efficiency.

Keywords: PFAS, Foam Fractionation, Leachate, Removal Efficiency, Wastewater Treatment

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Preface

This study was conducted as a Master's thesis as part of the Master's Programme in Environmental and Water Engineering at the Swedish University of Agricultural Sciences (SLU) and Uppsala University (UU). The thesis is the final part of the master program holding 30 credits and lasted for twenty weeks from January to June 2021. My supervisor was Philip McCleaf from Uppsala Vatten and my academic supervisor was Lutz Ahrens from the department of Aquatic Sciences and Assessments, Section for Organic Environmental Chemistry and Ecotoxicology.

I'm very grateful and would like to take the opportunity to thank Lutz Ahrens and Philip McCleaf for their educative way of helping me with this thesis. They have been patient and understanding and without their dedication of helping me and their outstanding knowledge of the subject this project would never have been done.

I would also like to send an extra thank to Sanne Smith that has been as a co supervisor for me. She has been a sounding board with a lot of knowledge within the subject, she has taught me the SPE-method and had a lot of experience that helped me approach upcoming problems.

Without the support of Uppsala Vatten och Avfall that provided the equipment for the experiments and access to Hovgården Avfallsanläggning this would never have been possible. Thank you, Elin Eriksson, head of the POPs lab at SLU for letting me use the lab and all its equipment and thank you Björn Bonnet for giving me approval to publish your map of Hovgården.

Axel Krögerström

Uppsala, June 2021

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Populärvetenskaplig sammanfattning

Att vatten kan innehålla ämnen som är dåliga för både människan och miljön råder det konsensus om. Mängden och sammansättningen av dessa ämnen varierar i olika vatten. Då vatten ska drickas och användas i matlagning finns det krav på vad vatten får innehålla och inte innehålla.

Per- och polyfluorerade alkylsubstanser (PFAS) är mänskligt framtagna ämnen som används i bland annat kläder, matförpackningar och brandskum och då det är både fett och vattenavskiljande så har det använts mycket. Dessa ämnen har bevisats ha en lång livslängd och vara vanligt förekommande på vissa ställen i naturen och även i människokroppen. En plats där PFAS är vanligt och finns i höga koncentrationer är i vattnet på deponianläggningar, detta då det avfallet som finns där i sig innehåller mycket PFAS. Det vatten som regnar eller kommer på det deponerade kallas för lakvatten. Hovgården är en deponianläggning utanför Uppsala där lakvattnet samlas upp och renas i ett reningsverk. Vattenreningen på Hovgården är anpassad efter de krav som finns och snart kan det komma krav på rening av PFAS så detta måste då undersökas.

Denna studie har undersökt möjligheten att rena vatten från PFAS genom att i en kolonn lufta vatten underifrån och ta bort det skum som bildas vid ytan. Detta heter skumfraktionering och skulle fungera då PFAS är ett ytaktivt ämne som är vattenavstötande och trivs bättre i luftbubblorna än i vattnet och skulle därmed följa med bubblorna ut. I studien har både uppehållstiden för vatten i kolonnen och mängden skum som tagits ut undersökts. Experimentet gick till genom att det smutsiga lakvatten pumpades in i kolonnen med ett membran som satt i botten som pumpade ut luft. Från kolonnen gick en slang med vatten från botten ut till avloppet medan en slang tog skum från ytan och ledde det till en behållare som samlade upp skummet. Vattenprover togs på vattnet som pumpades in i kolonnen, det utgående vattnet och på skummet. Vattenproverna togs sedan till ett labb där mängden PFAS mättes och jämförelser mellan PFAS-halter i in och utflödena gjordes.

Totalt gjordes sex experiment där varje experiment bestod av två eller tre identiska försök för att se att de inte skiljde sig allt för mycket. Från resultaten kunde man konstatera att skumfraktionering fungerar bättre för de stora PFAS-molekylerna och sämre för de små men att alla experiment visade sig minska PFAS-halterna. Det försöket som fick mest procentuell minskning av PFAS var det försöket med längst uppehållstid, ett experiment med 30 minuters kontakttid och fem procent skum. Då var den sammanlagda PFAS-borttagningen ungefär 58 procent men för de långa molekylerna var det 92 procent. Det experimentet med lägst PFAS-borttagning var ett försök med tio minuters kontakttid och 20 procent skum, då var den totala PFAS-borttagningen 41 procent och borttagningen av långa molekyler 66 procent.

Denna studie kunde visa att en längre uppehållstid i vattnet ledde till en större borttagning av PFAS-molekylerna men inget samband mellan ökad skumhalt och större borttagning kunde bevisas. Studien kunde också visa att en högre PFAS-koncentration i vattnet resulterade i en högre borttagning av PFAS-molekylerna.

Abbreviations:

- AFFFs - Aqueous film-forming foams
- APGs - Alkyl polyglycosides
- C^F₄- Tetrafluoromethane
- CT - Contact Time
- Dm – Dry matter
- EEA - European Environment Agency
- EFSA - European Food Safety Authority
- ET-FOSAA - N-ethylperfluorooctanesulfonamido acid
- FOSA - Perfluorooctanesulfonamide 1
- FF - Foam fractionation/fraction
- FTS - Fluorotelomer sulfonate
- GAC - Granular activated carbon
- GW – Groundwater
- HWPO-DA - Hexafluoropropylene oxide-dimer acid
- IX - Anion exchange/Ion exchange
- LSG - Less sensitive ground
- Me-FOSAA - N-methylperfluorooctanesulfonamido acid
- NaDONA - Sodium dodecafluoro-3H-4, 8-dioxanonanoate
- NF - Nanofiltration
- PCB - Polychlorinated biphenyl
- PerFAS - Perfluoroalkyl substances
- PFAS - Per- and polyfluoroalkyl substance
- PFBA - Pentafluorobenzoic acid
- PFBS - Perfluorobutanesulfonic acid
- PFCA - Perfluorocarboxylic acid
- PFDA - Perfluorodecanoic acid
- PFDS - Perfluorodecanesulfonic acid
- PFDoDA - Perfluoro-n-dodecanoic acid
- PFECHS - Perfluoro-4 ethylcyclohexane-sulfonate
- PFHpA - Perfluoroheptanoic acid
- PFHpS - Perfluoroheptanesulfonic acid
- PFHxA - Perfluorohexanoic acid
- PFHxS - Perfluorohexane sulfonic acid
- PFNA - Perfluorononanoic acid
- PFNS - Perfluorononane sulfonic acid
- PFOA - Perfluorooctanoic acid
- PFOS - Perfluorooctane sulfonate
- PFPE - Perfluoropolyethers
- PFPeA - Perfluoropentanoic acid
- PFPeS - Perfluoropentane sulfonic acid
- PFSA - Perfluoroalkane sulfonic acids
- PFTeDA - Perfluoro-n-tetradecanoic acid

- PFTriDA - Perfluoro-n-tridecanoic acid
- PFUnDA - Perfluoro-n-undecanoic acid
- PolyFAS - Polyfluoroalkyl substances
- SBT - Sequencing Batch Reactors
- SG - Sensitive ground
- SGI - Swedish Geotechnical Institute
- SPE - Solid phase extraction
- STDAV – Standard deviation
- US EPA - United States Environmental Protection Agency
- UV - Ultraviolet
- 4:2 FTSA - Fluorotelomer sulfonic acid
- 6:2 FTSA - Fluorotelomer sulfonate
- 8:2 FTSA Fluorotelomer sulfonate
- 9CI-PF3ONS - Potassium 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate
- 11CI-PF3OUds - 11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid

List of key concepts

- Air flow rate – The air flow divided by the influent flow of leachate.
- Contact time – The average time the water is in the column.
- Enrichment – The increase of concentration in the foam and underwater compared to the influent water.
- Foam Fraction – The percentage of the inlet flow that exits the column as foam instead of as effluent water.
- Foam Fractionation – The process in which hydrophobic molecules (in this case PFAS) are removed using foam.
- Leachate – Liquid (in this case mostly rain) that has percolated through a solid and leached out some of the constituents.
- Long chained PFAS – PFAS with a carbon chain of six or longer.
- Mass balance – A fraction between the foam or effluent flow divided with the influent flow.
- Removal Efficiency – The percentage difference in PFAS concentration between influent and effluent flow.
- Short chained PFAS – PFAS with a carbon chain shorter than six carbons.
- Underwater – The water sampled from approximately 5 cm under the foam.

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1 Introduction

Per- and polyfluoroalkyl substances (PFAS) are the “PCB of our time” as the Swedish Society for Nature Conservation states while begging the government to impose a permanent ban of PFAS in products aimed for the consumer (Naturskyddsföreningen 2016). PFAS are a large group of manmade chemicals that are water and oil repellent and withstand high temperatures, which makes them suitable for use in for example kitchen ware, clothes, fire foam and food packages (Livsmedelsverket 2021a). PFAS have carbon-fluorine bonds, which are very strong and therefore PFAS are very persistent and accumulates in the environment (ECHA n.d.) and the human body (US EPA 2016). PFAS are proven to have negative effects on humans and the environment and are therefore something that needs to be addressed (European Environment Agency 2020).

Landfill leachate is a source of PFAS to the environment (Benskin et al. 2012) but there is generally no treatment of leachate in Sweden (Swedish Environmental Protection Agency 2019). The treatment of PFAS contaminated leachate currently used is for example ion exchange (IX) and active carbon, and they are expensive and there is a need for investigating other water treatment techniques (Swedish Environmental Research Institute 2020).

One alternative technique is removal of PFAS using aeration foam fractionation, a technique where the water is aerated and PFAS adsorbs to and accumulates in the rising bubbles and is removed in the created foam (Meng et al. 2018). At Hovgården, a waste facility with a landfill, PFAS have been detected in the leachate but it has no specific treatment step suited for PFAS (Uppsala Vatten och Avfall AB 2018b). The applicability of aeration foam fractionation as a treatment step at Hovgården is further investigated in this report.

1.1 Aim and research questions

One aim of this master thesis is to continue Kjellgren (2020) thesis to examine if foam fractionation is a method that works towards removal of PFAS from contaminated water. Another aim is to investigate how different parameters e.g. contact time (CT) and foam fraction (FF) affect the removal efficiency (RE). The three research questions are presented below:

1. Does aeration foam collection reduce PFAS levels in leachate from Hovgården?
2. How does the contact time and collected amount of foam affect reduction of PFAS in leachate from Hovgården using aeration foam collection?
3. Does the removal of PFAS using aeration foam collection depend on the perfluoroalkyl carbon chain length and functional group?

1.2 Hypothesis

The hypothesis is that an increased contact time will result in an increased reduction of Σ PFAS. In addition, it is hypothesized that a higher foam percentage will result in a higher removal efficiency and there will be an enrichment of short chained PFAS in the samples taken under the foam. Finally, it is hypothesized that foam fractionation will work better for long chained PFAS in comparison to short chained PFAS.

2 Background and theory

2.1 What are PFAS?

PFAS exist in the water that we drink, the food that we eat, the kitchen tools we use and other products that we consume (Ding et al. 2020). PFAS are a group of manmade molecules that are both dirt repellent and hydrophobic. These characteristics makes PFAS useful in surface treatment on cloths as well as cooking tools (Naturskyddsforeningen 2015). The molecules are made up of chains of linked fluorine and carbon atoms, which creates strong bonds and make them very stable (NIH 2020).

2.1.1 PFAS classification

There are more than 4700 different types of PFAS molecules and it is an increasing number due to industries that creates new types of PFAS (NIH 2020). PFAS can be divided into three different groups or classes, perfluoroalkyl substances (PerFAS), polyfluoroalkyl substances (PolyFAS) and fluorinated polymers (Ahrens & Bundschuh 2014). The PerFAS have an alkyl chain that is fully fluorinated (i.e., fluorines have replaced all hydrogens on the carbons except for the carbons associated with functional groups) and has the formula $C_nF_{2n+1}R$ (where R is the functional group). The PolyFAS alkyl chain is partly fluorinated and contains at least one fluorine atom. The third class, Fluorinated polymers includes a great variation of atoms and molecules and is divided into three subclasses, perfluoropolyethers (PFPE), fluoropolymers and side-chain fluorinated polymers (Ahrens & Bundschuh 2014).

- Perfluoroalkyl substances (PerFAS)
- Polyfluoroalkyl substances (PolyFAS)
- Fluorinated polymers
 - Perfluoropolyethers (PFPE)
 - Fluoropolymers
 - Side-chain fluorinated polymers

PFAS can also be divided into long and short chains, depending on the length of the fluorinated carbon chain. The term *long chains* refers to three different groups: (OECD n.d).

- Perfluorocarboxylic acids (PFCA), which has a carbon chain length greater than eight. This section includes perfluorooctanoic acid (PFOA).
- Perfluoroalkane sulfonic acids (PFSA), which has a carbon length greater than six. This section includes perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonic acid (PFHxS).
- Precursors of these substances (substances that after a reaction will degrade into the substances above).

Two of the most common molecules within the PFAS group is PFOA and PFOS (US EPA 2016).

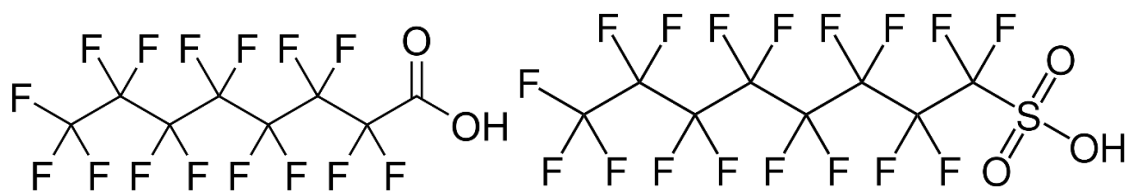


Figure 1 To the left the structural formula of PFOA and to the right the structural formula of PFOS.

Figure 1 shows the structural formula of PFOA and PFOS (both i.e. C₈ after its number of carbon atoms) that are both part of the PerFAS group (US EPA 2017). Both PFOA and PFOS are surfactants with a hydrophilic head and a fluorinated tail. The difference between these are their functional groups where PFOA has a carboxylic acid and PFOS has a sulfonic acid. A PFAS molecule with a carboxylic acid as a functional group is called a carboxylate and a PFAS molecule with a sulfonic acid is called a sulfonate (Lee et al. 2017).

2.1.2 Occurrence of PFAS in landfill leachate and groundwater at contaminated sites

A common characteristic of high fluorinated subjects is their ability to spread, both through air and water, which explains the existence of PFAS where there is no production or utilization. In Sweden, the largest emission source of PFAS are from fire foam at fire drill sites (Kemikalieinspektionen 2021). There are some preliminary guidelines for PFOS (which was seen as the most relevant PFAS molecule due to its occurrence and danger) in Swedish groundwaters developed by Swedish Geotechnical Institute (SGI). Dry matter (DM) is the material that is left after a complete dry out. The guidelines are for PFOS in less sensitive ground (LSG), sensitive ground (SG) and groundwater (GW) and are shown in Table 1.

Table 1 Preliminary guidelines of PFOS in sensitive ground (SG), less sensitive ground (LSG) and groundwater (GW) (Swedish Geotechnical Institute 2015)

	LSG	SG	GW
Preliminary guideline	0.020 [mg kg ⁻¹ DM ⁻¹]	0.003 [mg kg ⁻¹ DM ⁻¹]	45 [ng L ⁻¹]

Generally, in Sweden there is no treatment of the leachate from landfills designed to handle PFAS but is stated by the Swedish Environmental Protection Agency as something that is important to do to prevent the spread of PFAS (Swedish Environmental Protection Agency 2019).

The Swedish Waste Management Association (Avfall Sverige) has measured different PFAS molecules in treated and untreated leachate at eighteen landfills in Sweden. The treated leachate is treated primarily by wetlands, natural filtration, sequencing batch reactors (SBR) and different types of aeration. As seen in the Table 2 and 3 the concentrations are higher in the untreated water, than in the treated water and based on the average concentrations the Σ PFAS (Σ PFAS is the sum of all PFAS) removal efficiency is 32 % (Larsson 2020). PFOS and PFOA were two of the three most frequently found PFAS among with PFHxS in the leachate and the concentrations are

higher than guidelines (Avfall Sverige 2018). The results from the measurements are shown in Table 2 and Table 3.

Table 2 Measured concentration (ng L⁻¹) of Σ PFAS in untreated leachate at eighteen landfills in Sweden (Larsson 2020)

Compounds	Maximum concentration [ng L⁻¹]	Average concentration [ng L⁻¹]	Median concentration [ng L⁻¹]
PFBA	6 000	510	230
PFPeA	4 500	1 000	410
PFHxA	5 600	1 000	480
PFHpA	1 800	360	180
PFOA	4 200	500	300
PFNA	76	25	15
PFDA	220	23	11
PFBS	11 000	730	140
PFHxS	8 900	380	120
PFOS	9 600	340	120
6:2 FTSA	14 000	790	42

Table 3 Measured concentrations (ng L⁻¹) of Σ PFAS in treated leachate at eighteen landfills in Sweden (Larsson 2020)

Compounds	Maximum concentration [ng L⁻¹]	Average concentration [ng L⁻¹]	Median concentration [ng L⁻¹]
PFBA	7 200	1 000	140
PFPeA	690	240	240
PFHxA	2 900	570	310
PFHpA	260	120	120
PFOA	2000	430	230
PFNA	100	25	8.2
PFDA	110	32	3.4
PFBS	920	340	63
PFHxS	1 800	300	84
PFOS	1 500	450	64
6:2 FTSA	2000	320	110

In 2015, 161 samples from groundwater in Sweden were analysed to determine levels of 26 PFAS. The highest detected concentration of Σ PFAS in groundwater in the study were 6400 ng L⁻¹ (Gobelius et al. 2016) and the average concentration of Σ PFAS in the groundwater were 49 ng L⁻¹ (Ahrens et al. 2016).

2.1.3 Environmental and human concern of PFAS

PFOA and PFOS are non-degradable, which results in that they are very persistent in the body as well as in nature. Since the molecule does not degrade it will accumulate and the concentrations will increase over time. The high residence time can lead to negative effects on the body (US EPA 2016). There is an uncertainty of how PFAS can affect the

human body, but the European Environment Agency (EEA) has listed some effects of PFAS on human health and sorted them into two groups, high certainty and lower certainty. Some of the health issues are listed in Table 4:

Table 4 Health concerns regarding human exposure of PFAS (European Environment Agency 2020)

High certainty	Lower certainty
Increased cholesterol levels	Inflammatory bowel disease
Kidney and testicular cancer	Breast cancer
Liver damage	Increased miscarriage risk
Thyroid disease	Low sperm count and mobility

Regardless of whether PFAS affect the human body or not, there are still guidelines to follow. According to the European Food Safety Authority (EFSA) 4.4 μg per kg body weight and week is the highest amount of PFAS that the human body can ingest without any risks. The Swedish food agency (i.e Livsmedelsverket) has no juridical restrictions for the amount of PFAS in the drinking water but has highly recommended that the water should contain less than 90 ng L^{-1} for $\sum_{11}\text{PFAS}$ (Livsmedelsverket 2021b). The 11 PFAS included by the Swedish food agency are PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS and 6:2 FTS (NIRAS n.d). The European Commission has a limit of 100 ng L^{-1} (for $\sum_{20}\text{PFAS}$, the 20 by the European Commission chosen types of PFAS shown in Appendix Table 1 in Appendix 1)) in drinking water that will be legally binding before January 2023 (Livsmedelsverket 2021b). Gobelius et al. (2016) analysed 169 samples of drinking water in Sweden and found out that three percent of the samples exceeded the limit of 90 ng L^{-1} (Gobelius et al. 2016) and the average was 8.4 ng L^{-1} (Ahrens et al. 2016). The PFOS environmental quality standard value from the EU drinking water directive is 0.65 ng L^{-1} in inland surface water (European Environment Agency 2019).

2.2 PFAS treatment methods

There are many existing techniques and methods for treatment of PFAS contaminated water. Some of the techniques are based on sorption (US EPA 2018), some of destruction (US EPA 2020) and some on separation (Anazadehsayed et al. 2018). A selection of the techniques is described in the following chapters.

2.2.1 Sorption techniques

Sorption is a process where one particle or substance becomes attached to another particle. Adsorption is a both chemical and physical type of sorption where molecules or ions adsorb and accumulate to the surface of another phase. Activated carbon treatment is one physical adsorption technique and is the most studied treatment method for PFAS (US EPA 2018).

2.2.1.1 Granular Active Carbon

Granular activated carbon (GAC) is a material with a high porosity and therefore has a large surface area for PFAS to adsorb on, which makes it suitable for PFAS removal (US EPA 2018). GAC is proven to be more efficient for longer PFAS chains like PFOA and PFOS and does not have as high efficiency for shorter chains (US EPA 2018). This is due to the lower adsorption capacity of the shorter chained PFAS. Except for PFAS removal

GAC can also be used for removal of organic matters and taste and odour causing substances (Lindgren 2015).

2.2.1.1.1 Anion Exchange

Anion exchange (IX) treatment i.e. ion exchange (IX) treatment is another sorption technique that is used for PFAS removal (US EPA 2018). IX is a reversible reaction where ions are replaced with ions of the same electrical charge (Samco 2017). PFAS are a negatively charged contaminant and therefore a positively charged anion exchange resin is effective (US EPA 2018). The affinity of the resin to an ion will tell at what point they will adsorb.

The breakthrough curves that can be created (one for each ion) due to the variation in affinity can tell which resin that is suitable for which contaminant. IX can also be used to demineralize and soften water (Lindgren 2015).

2.2.2 Destructive techniques

2.2.2.1 Incineration

Using heat for destruction (mineralization) of chemicals is called incineration (Interstate Technology Regulatory Council 2020). Incineration as a destructive technique is a well-developed and used technique for the breakdown of other halogenated organic chemicals like PCBs (but for PFAS removal it is an ongoing area of research) (US EPA 2020). The heat is applied directly on the PFAS contaminated water (Interstate Technology Regulatory Council 2020). To break the carbon-halogen bond high temperature and long RT is needed. When the bond is broken the halogen is scrubbed from the flue gas. Since the stability of fluorinated compounds, the electronegativity of fluorine and the strong bonds there is a risk of an incomplete breakdown (when larger PFAS chains becomes smaller chains). Tetrafluoromethane (C^F_4) is the most difficult compound to break down and requires a temperature above 1 400 °C (US EPA 2020).

2.2.2.2 Combined ozonation

Ozonation can be combined with other methods to improve the degradation. Using a catalyst together with a high concentration of ozone is called catalysed ozonation and is a method to break down PFAS (Sjöblom 2020). A catalyst is a substance that increases the rate of a reaction without being consumed (Hammers 2017). Since ozone itself is unable to break down PFAS the catalysis assists the ozone and the PFAS turns into substances that are less hazardous (Sjöblom 2020). Franke et al. (2019) found out that this method is effective for PFAS at a length of C^F_7 - C^F_{11} with a removal of >98 % but less effective for PFAS that have longer or shorter chains. Therefore, this method can be used for specific PFAS but there is a need to be aware of eventual dangerous transformation products from ozonation (Franke et al. 2019).

Ultraviolet light (UV) combined with ozone (O_3) is another combination that is used for treatment of contaminated water. The irradiation of the UV creates a split of the ozone. The free oxygen is then reacting with the water to create H_2O_2 that decomposes into $\cdot OH$ (Dai et al. 2019).

The relationship between the concentration in the foam created by the ozonation and the treated water is called partition factor:

$$\lambda = \frac{C_{foam}}{C_{treated}} \quad (1)$$

where λ is the partition factor, C_{foam} is the PFAS concentration in foam and $C_{treated}$ is the PFAS concentration of the treated water. The partition factor has a nearly exponential relationship with the PFAS carbon number. This can be explained by the fact that air-water and octanol-water partition coefficients exponentially increases PFAS chain length. As mentioned by Franke et al. (2019) the same discovery was made by Dai et al (2019), the destruction of PFAS results in an increase of concentration of short chained PFCA (Dai et al. 2019).

2.2.3 Separation techniques

2.2.3.1 Foam fractionation

Foam fractionation is a process where hydrophobic molecules are removed from a liquid using aeration. This technique can be used with a rising column of foam (Anazadehsayed et al. 2018). Foam fractionation is an old technique, which also can appear if dissolved gas is released whether it is by purpose or not. The foam fractionation is used in two different ways, batchwise and continuous operations. The difference between the two ways of using foam fractionation is shown in Figure 2.

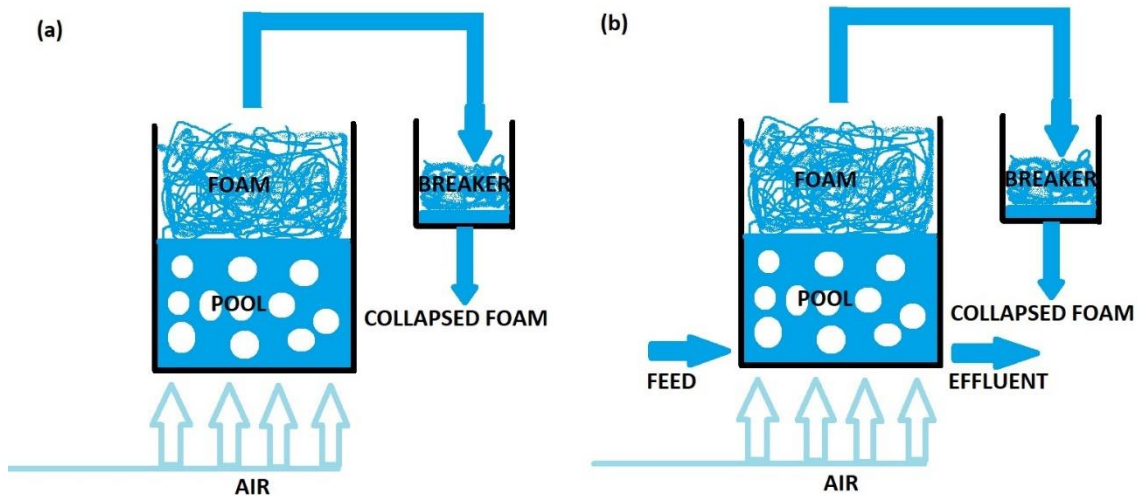


Figure 2 The two different ways of using foam fractionation, (a) batchwise and (b) continuous. Adapted from (Lemlich 1968).

As demonstrated in Figure 2, both methods get aerated from below, the bubbles adsorb substances that are removed when the foam is removed. The batchwise operation uses one batch, which gets aerated or gassed for a certain time and the whole batch gets exchanged by a new batch. The continuous operation has a constant flow through both the inlet and the outlet. There is also a possibility to reflux collapsed foam in to the pool again to concentrate it even more or to repeat the whole process but instead of using leachate as influent water, using collapse foam (Lemlich 1968).

There are many different parameters that can be adjusted to optimize the foam fractionation process regarding the removal efficiency, energy consumption etc. Some of the parameters are (Merz et al. 2011):

- pH value
- Temperature
- Feed flow rate
- Gas flow rate
- Bubble size

A variation of the feed inlet can also be made, where the inlet of the feed water can be placed in different heights and if needed some of the collapsed foam can be reintroduced to the pool (called reflux) to be processed a second time (Lemlich 1968).

Enriching mode is when the feed enters the column in the liquid pool, illustrated in Figure 3b, and stripping mode is when the feed inlet is in or above the foam and runs down to the opposite direction of the rising foam such as in Figure 3a.

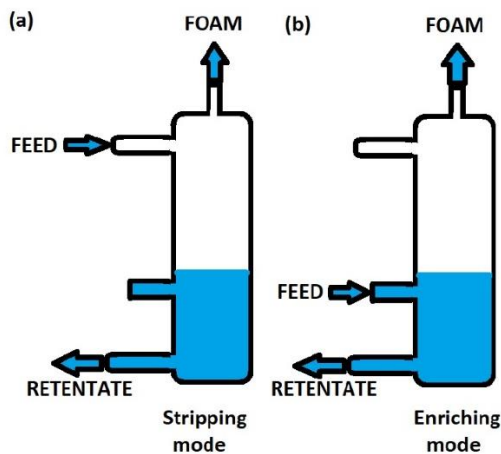


Figure 3 The difference between (a) stripping and (b) enriching mode. Adapted from Merz et al. (2011).

The stripping mode allows the inlet feed to fill free adsorption sites in the rising foam (Merz et al. 2011). The CT between the surface-active molecules and the gas bubbles is determined by the gas flow rate and affects the recovery. If the gas flow rate increases the CT decreases and therefore the time for adsorption decreases as well. Contrary, a high gas flow rate increases the volume of air bubbles and increases the hydrophobic area available for adsorption (Merz et al. 2011).

2.2.3.2 Nanofiltration

Membranes are another separation treatment technique that can be used for PFAS removal. A membrane can be pressure driven and the pore size can be chosen with respect to the purpose. Nanofiltration (NF) normally has a pore size of 1-10 nm (10^{-9} m) and operates at a pressure above 3 bar (Lundgren 2014). Micro and ultrafiltration (MF and UF) have pore sizes of around 10^{-6} m and $2 \cdot 10^{-9}$ to 10^{-7} , respectively (the pore size is not small enough for PFAS treatment) (Khulbe et al. 2008) and operates at a pressure of 0.8-3 bar (Lundgren 2014).

The United States Environmental Protection Agency (US EPA) states that high pressure membranes like NF are effective at removing PFAS and other unwanted substances like pesticides. NF can as well lower the hardness but still keep the wanted salts and minerals in the water (Lindgren 2015; US EPA 2018). The membranes can be made of for example highly cross-linked or organic polymers and ceramics. The removal efficiency of PFAS using NF are depending on the size of the molecule, intermolecular and acting forces between the molecules and the hydrophobicity of the particular molecule (Lindgren 2015).

2.3 Studies on foam fractionation as treatment for PFAS contaminated water

Foam fractionation as PFAS removal is a method that has been tested for water treatment.

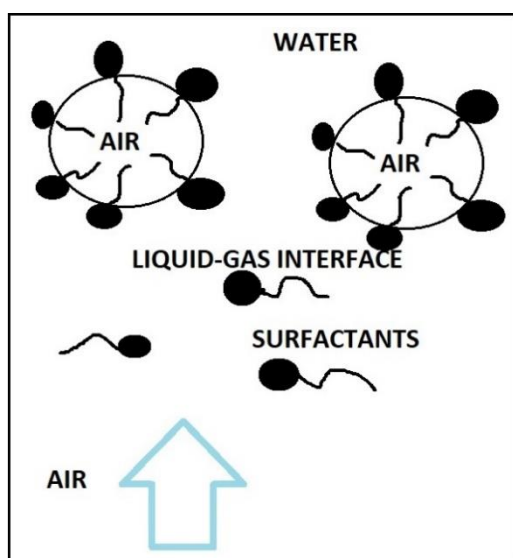


Figure 4 Surfactants attached to the air bubbles in the water. Adapted from Tharapiwattananon et al. (1996).

As shown in Figure 4 surfactants attach to the air bubbles due to their hydrophobic tails and hydrophilic heads (Tharapiwattananon et al. 1996).

2.3.1 Removal of PFOS & PFOA by foam floatation

A study of batchwise foam floatation as a technique of PFOX (PFOA and PFOS, X= A or S) removal was performed by Lee et al (2017). A measurement of PFOX removal was conducted in a column made of polyethylene at a length of 0.3 m and an inner diameter of 0.04 m.

The reaction time was set to five min and a gas diffuser with a pore size of 25-50 μm (air flow rate set to 7.5 L min^{-1}) was used varying only the cationic activators, all at a concentration of 11.5 mM, got the following PFOS removal percentage:

Table 5 Removal percentage (%) of PFOS using different cationic activators (Lee et al. 2017)

Cationic activator	PFOS removal percentage (%)
Fe ³⁺	99.5
La ³⁺	99
Al ³⁺	90
Ca ²⁺	29
Fe ²⁺	25
K ⁺	3

The numbers in Table 5 can be seen as an indication of which cation activators can be effective and which are less effective. The study also investigates how pH correlates to PFOX removal. It showed that PFOA-removal is more sensitive to pH than PFOS.

For both PFOS and PFOA the removal percentage was higher at low pH and had its maximum removal at pH 2.3 and for pH greater than or equal to 6 no removal was observed (regardless of a high ferric dose). The report explains the low removal at a high pH due to the competition between PFOX and OH⁻ where iron hydroxide (Fe(OH)_x) is produced (Lee et al. 2017).

2.3.2 Removal of PFAS by aeration foam collection

2.3.2.1 Aqueous film-forming foams

Another study by Meng et al. (2018) was performed on aqueous film-forming foams (AFFFs) used by fire fighters, which is a source of PFOS contamination in water. The treatment was done by removing the foam after aeration. This study was carried out with a column that had an inner diameter of 0.05 m and a height of 0.75 m. 0.6 L of contaminated water were added and a 10 µm in diameter air diffuser (air flow rate set to 75 mL min⁻¹ made of titanium was placed on the bottom of the column (Meng et al. 2018).

After a two-hour run a PFOS removal of 96 % was obtained. When adding already existing hydrocarbon surfactants such as alkyl polyglycosides (APGs) and using the same run time a PFOS removal of >99.9 % was achieved. Another discovery that was made in the study was that increasing the flow rate, the concentration of co-existing surfactants and the strength of the ions as well as decreasing initial PFOS concentration resulted in an increased foam value, which results in a higher PFOS removal. Due to the relatively low surface activity of the short-chain PFAS the foam collection was less effective for those PFAS but overall aeration-foam collection was found effective of high concentrations of PFOS from AFFF-contaminated wastewater (Meng et al. 2018).

2.3.2.2 Aeration foam collection

Another previous study was done by Kjellgren (2020) on aeration foam collection (both batch wise and continuous) as a treatment of PFAS contaminated water. A column of 1.0 m in height and an inner diameter of 0.057 m was used, which resulted in a volume of 2.4 L. The continuous experiments had a constant air flow rate at 2 L min⁻¹ and CT at approximately 5, 10 and 20 min (Kjellgren 2020).

The continuous experiments Σ PFAS removal varied between 72 and 94 % with an average of 86 % and the water with the highest CT, 20 min had the highest Σ PFAS

removal. The study showed that most of the PFAS were removed in the beginning of each experiment, but more time was needed to remove a larger amount and reach steady state (Kjellgren 2020).

3 Method and Materials

3.1 PFAS analysed

The following PFAS (Table 6) were detectable in the analysis and their number of carbons excluding their functional group and their component groups (ITRC 2020) are listed with their class (New Hampshire Department of Environmental Services 2019).

In total four short carboxylates, three short sulfonates, seven long carboxylates, four long sulfonates and eleven PFAS classified as rest were investigated.

Throughout this study the definition of long chained PFAS are with a carbon chain length of six or longer and for short chained PFAS a carbon chain length of five or shorter.

Table 6 Detectable PFAS in the HPLC MS/MS, its number of carbons, SC=short carboxylate, LC=long carboxylate, SS= short sulfonate, LS=long sulfonate & R=rest and its PFAS class. The identified PFAS in this study are marked cursive (New Hampshire Department of Environmental Services 2019; ITRC 2020)

Component	Full Name^b	PFAS class^a	Chain length (CF_n)^b	Component group^b
<i>PFBA</i>	<i>Perfluoro-n-butanoic acid</i>	PFCA	3	SC
<i>HFPO-DA</i>	<i>Hexafluoropropyleneoxide-dimer acid</i>	PFCA	3	R
<i>PFPeA</i>	<i>Perfluoro-n-pentanoic acid</i>	PFCA	4	SC
<i>PFHxA</i>	<i>Perfluoro-n-hexanoic acid</i>	PFCA	5	SC
<i>PFHxA</i>	<i>Perfluoro-n-hexanoic acid</i>	PFCA	5	SC
<i>PFHpA</i>	<i>Perfluoro-n-heptanoic acid</i>	PFCA	6	LC
<i>PFOA</i>	<i>Perfluorooctanoic acid</i>	PFCA	7	LC
<i>PFNA</i>	<i>Perfluorononanoic acid</i>	PFCA	8	LC
<i>PFDA</i>	<i>Perfluoro-n-decanoic acid</i>	PFCA	9	LC
<i>PFUnDA</i>	<i>Perfluoro-n-undecanoic acid</i>	PFCA	10	LC
<i>PFDoDA</i>	<i>Perfluoro-n-dodecanoic acid</i>	PFCA	11	R
<i>PFTriDA</i>	<i>Perfluoro-n-tridecanoic acid</i>	PFCA	12	LC
<i>PFTeDA</i>	<i>Perfluoro-n-tetradecanoic acid</i>	PFCA	12	LC
<i>PFBS</i>	<i>Perfluorobutanesulfonic acid</i>	PFSA	4	SS
<i>PFPeS</i>	<i>Perfluoropentane sulfonic acid</i>	PFSA	5	SS
<i>PFHxS</i>	<i>Perfluorohexanesulfonic acid</i>	PFSA	6	SS
<i>PFHpS</i>	<i>Perfluoroheptanesulfonic acid</i>	PFSA	7	LS
<i>PFOS</i>	<i>Perfluorooctanesulfonic acid</i>	PFSA	8	LS
<i>PFNS</i>	<i>Perfluorononane sulfonic acid</i>	PFSA	9	LS
<i>PFDS</i>	<i>Perfluorodecanesulfonic acid</i>	PFSA	10	LS
<i>4:2 FTSA</i>	<i>Fluorotelomer sulfonic acid</i>	Precursors	4	R
<i>6:2 FTSA</i>	<i>Fluorotelomer sulfonate</i>	Precursors	6	R
<i>8:2 FTSA</i>	<i>Fluorotelomer sulfonate</i>	Precursors	8	R

<i>Me-FOSAA</i>	<i>N-methylperfluorooctanesulfonamido acid</i>	Precursors	8	R
<i>Et-FOSAA</i>	<i>N-ethylperfluorooctanesulfonamido acid</i>	Precursors	8	R
<i>PFECHS</i>	<i>Perfluoro-4ethylcyclohexane-sulfonate</i>	Cyclic	8	R
9CI-PF3ONS	Potassium 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate	Additional	9	R
11CI-PF3OUds	11-chloroeicosafluoro-3-oxaundecane-1-sulfonic acid	Additional	11	R
NaDONA	Sodiumdodecafluoro-3H-4, 8-dioxananoate	PECA	7	R

3.2 Characterization of the landfill site at Hovgården

Hovgården is located twelve kilometres northeast of Uppsala. Hovgården is a waste facility that is owned and managed by Uppsala Vatten och Avfall and that is approximately 570 000 m² (Uppsala Vatten och Avfall AB 2018a). Hovgården has been an active facility since 1971 and that consists of an active and an old landfill, two sludge cells, a recycling centre, a treatment plant for leachate, six hard standing surfaces for intermediate storage and a section for compost (Uppsala Vatten och Avfall AB 2018a) as seen in Figure 5 (Bonnet 2017).

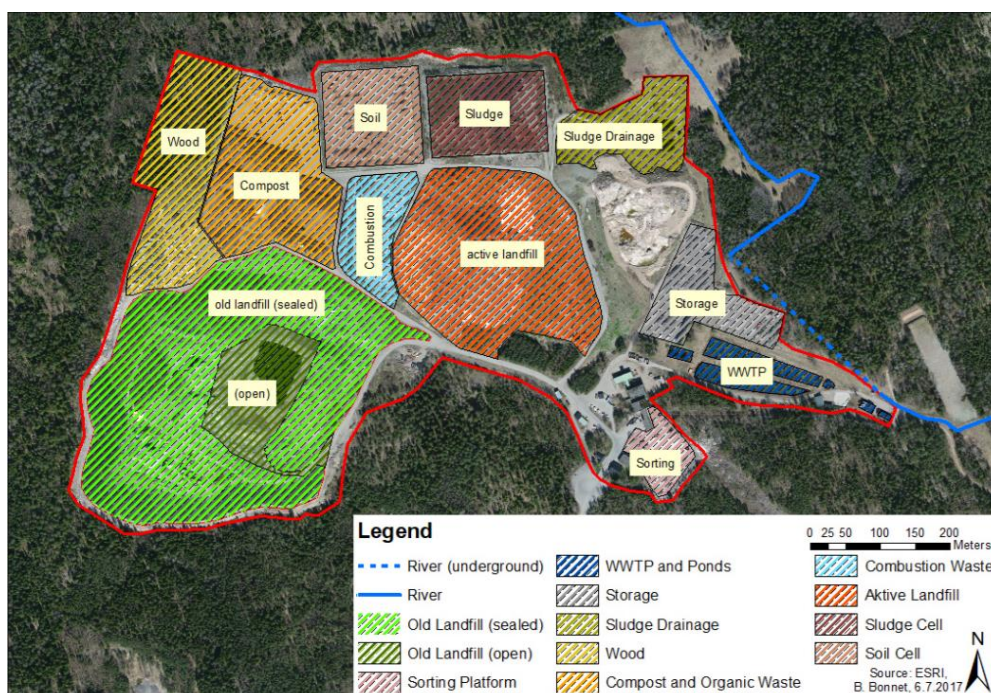


Figure 5 Map over Hovgården made by Bonnet (Bonnet 2017). Map published with approval of Bonnet.

To prevent the leachate to reach the groundwater are drainage pipes placed under the landfill and all other areas on the sight. This leads all the water to a treatment plant. The leachate and the surface water from the hard standing is treated with a mechanical step were mainly iron and manganese are oxidised and to some extent the organic matters

as well. After the aeration, particles in the water are sedimented using lamella sedimentation (when the leachate passes plenty of sloping lamellas before reaching overflow drain (Åtgärdsportalen 2017)). Later the water passes a biological step with a surface made for microorganisms to grow at. Therefore, the amount of organic matters decreases and there is also an oxidation of ammoniacal nitrogen to nitrate nitrogen. After the biological step there is a sedimentation pond, two parallel polishing ponds and an aerated pond before reaching the recipient Funbosjön through the small watercourses Tomtaån, Lissån and Hovgårdsbäcken (Uppsala Vatten och Avfall AB 2018a).

3.3 Experimental setup

The experimental setup was a continuous operation with a constant in and outflow. A plastic cylinder with a radius of 0.093 m was used and the water level was set to 1.7 m, which resulted in a water volume of 46 L. The leachate was obtained from the pump station at Hovgårdens landfill and led through a hose to a 300 L container to ensure that the influent concentration was the same through the whole experiment. The first leachate from the pump station was poured out to prevent unnecessary sediments. The leachate in the container was stirred using a Berner SP/ENC pump. A *Watson Marlow 630Bp* peristaltic pump was attached through hoses to the container of leachate and to the cylinder. The leachate in the container was stirred using a Berner SP/ENC pump. A *Watson Marlow 630Bp* peristaltic pump was attached through hoses to the container of leachate and to the cylinder.

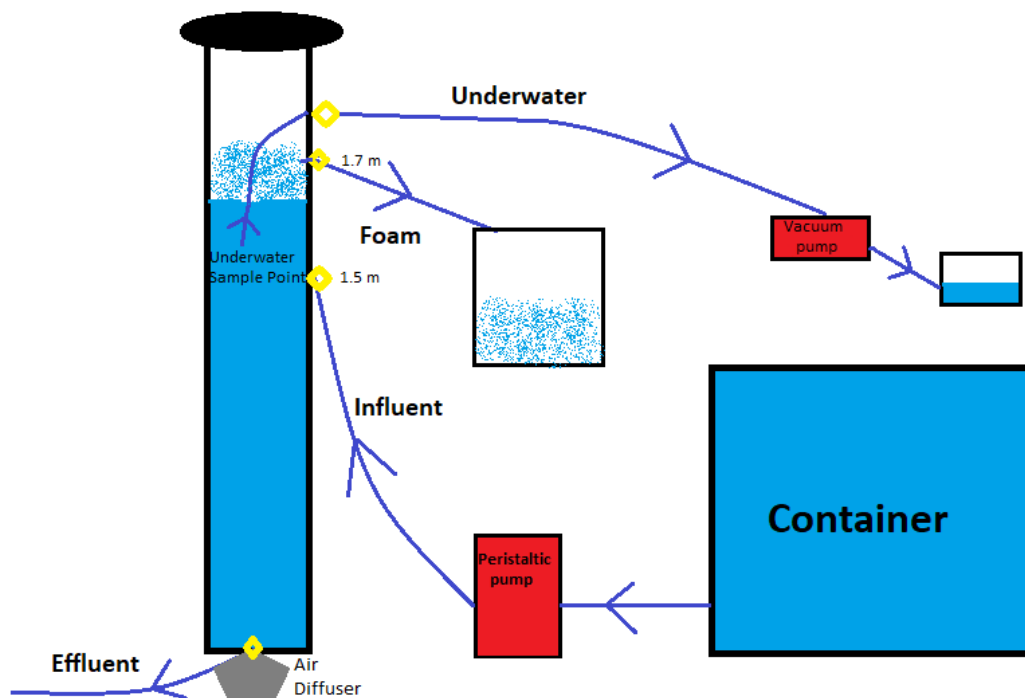


Figure 6 Schematic figure of the experimental set-up.

The cylinder had nine valves and the valve used for the inlet water was at a height of 1.5 m and the outlet valve used for the foam collection was at a height of 1.7 m. One valve at the bottom of the cylinder was open and the height of that outlet hose was adjusted to regulate the height of the water level and therefore get the foam creation at a desired height. At the bottom of the cylinder an air diffuser of the brand *Wika* was installed and a constant airflow of 10 L min^{-1} was used to create foam. The effluent water and the foam

were sampled in 250 mL sample flasks from the outlet hoses at given timesteps. In between each experiment the column and hoses were rinsed with tap water and before changing the CT the column was filled with tap water and strong dekalcol (an acidic agent for general cleaning and lime removal) was added as a cleaning step.

For the CT 10 min FF 10, 20 and 30 %, samples of the water below the foam were taken as well. This was done using a sampling container that was connected to a *Gast* vacuum pump and the cylinder. The outlet hose was placed approximately five cm under the foam level.

For each experiment (except for the CT 10 min FF 5 %), samples of the influent and effluent water were sent to ALS Scandinavia for a general chemistry evaluation. The results from those evaluations are listed in Appendix 3.

3.4 Solid Phase Extraction and instrumental analysis

3.4.1 Materials used in the SPE and instrumental analysis

In the SPE method a sonication bath was used as well as ordinary lab equipment like funnels, beakers, E-flasks, pipettes, lab clamps, tongs, scales and vacuums. Specific equipment like glass fibre filters (pore size 0.45 μm), wax extraction cartridges of the brand *Oasis*, SPE reservoirs, centrifuge tubes, 15 mL PP-tubes and 2 mL PP-vial were utilized.

The machines that were used were a centrifuge of the model *Eppendorf 5810r*, a nitrogen evaporator of the brand *Organomation Associates inc* and a high-pressure liquid chromatograph.

The following chemicals were required: *MeOH* (Methanol), *EtOH* (Ethanol), *NH₄CH₃CO₂* (Ammonium acetate), *CH₃COOH* (Acetic acid), *NH₄OH* (Ammonium hydroxide), Milli-Q water, Internal Standard (concentration of 0.05 $\mu\text{g mL}^{-1}$) and Standard (concentration of 250 ng mL^{-1}).

3.4.2 Course of action

A solid phase extraction (SPE) is a method to single out and adsorb a specific compound from a liquid before analysing it by making a complete phase separation. The SPE was used as an aid to determine the PFAS concentrations in the samples and the procedure is described below (Gobelius et al. 2018).

Before the procedure, all the lab equipment was rinsed three times with methanol to remove any earlier contamination. The sample was placed in a sonication bath for five min to remove any particles that had adsorbed to the walls at the inside of the flask. Then the sample was filtered through a glass fibre filter, and the filtration was rinsed three times with methanol to include the last compounds of the sample. In between each sample, all the equipment was rinsed three times with methanol. The glass filter removes all unwanted suspended solids (the adsorbed fraction of PFAS are thus not included in the analysis). This step is a way to make the SPE in upcoming steps go faster because it prevents clogging in later steps.

When all the samples were filtered, a negative blank sample (125 mL Milli-Q water) was added and all samples are spiked with 100 μL of an internal standard mixture, a mass label compound with different isotopes of PFAS, mostly using carbon 13 instead of

carbon 12, which is found in the calibration curve, so a comparison between the native peak and the internal peak can be made. The positive blank was spiked with 100 μL standard (native compounds) as well. After the spiking, the samples were placed in a sonication bath for five min for earlier given reasons.

Then *SPE WAX extraction* cartridges were placed on a glass rack and each cartridge was rinsed with 4 mL 0.1 % ammonium hydroxide in methanol, then 4 mL methanol and 4 mL of Milli-Q water to drop through subsequently (at a rate of approximately 1 drop s^{-1}).

SPE reservoirs were rinsed three times with methanol and then placed over the cartridges. Each sample was run through a cartridge. Important notification, the cartridges should never get fully dry, and the sample should be dripping and not pouring, otherwise the PFAS have no time to absorb to the cartridge.

The SPE reservoirs were removed, and the cartridge was rinsed with 4 mL of 25 mM ammonium acetate buffer. The slightly acidic buffer removes the salts and other unwanted compounds in the cartridge. The samples were then placed in a centrifuge tube and centrifuged in a centrifuge of the model *Eppendorf 5810r* at 3000 rpm for two min to get all the water out of the samples. After that, the samples were placed above the 15 mL PP-tubes, which are labelled with the sample ID and then were eluted, first with 4 mL methanol and then 4 mL of 0.1 % ammonium hydroxide in methanol to get the PFAS out. The samples were then vacuumed to get completely dry.

The samples were placed in a N_2 evaporator of the brand *Organomation Associates inc.* to evaporate the samples and concentrate them from 8 mL to 1 mL. The PP-tubes were rinsed once with methanol and then concentrated down to 1 mL again, before transferring the samples to 2 mL PP-vials. Then the PP-tubes were rinsed with methanol 3 times and the methanol was poured into the PP-vials to get the last PFAS out and then evaporated down to 1 mL again. At last, the vials were vortexed to completely mix them and to get the adsorbed components of the walls. The samples were run in HPLC MS/MS, which is a high-pressure liquid chromatography. The peaks were assigned to different compounds and based on the integration of the peaks the concentration could be determined using an analytic program called Sciex OS.

3.5 Experiments Performed Based on Contact Time (CT) and Foam Fraction (FF)

There were a total of six different experiments conducted containing 17 runs. Duplicates and triplicates were done to ensure the significance of the results. All experiments had an air flow of 10 L min⁻¹. The CT and FF for the different experiments are listed in Table 7.

Table 7 Number of runs and collected samples for the different experiments

Experiment	Number of runs	Total number of collected samples
CT 10 min FF 5 %	2	18
CT 20 min FF 5 %	3	18
CT 30 min FF 5 %	3	18
CT 10 min FF 10 %	3	18
CT 10 min FF 20 %	3	18
CT 10 min FF 30 %	3	18

In addition to Table 7 all the samples are listed in Appendix Table 2 in Appendix 2. As shown in Table 8 the air ratio three times greater for CT 30 min than for CT 10 min since the water flow is three times greater for the CT 10 min than for the CT 30 min.

Table 8 The air ratio of the different contact times (air flow divided by the water flow)

CT [min]	Water flow [L min ⁻¹]	Air flow [L min ⁻¹]	Air ratio $\left[\frac{\text{Air flow}}{\text{Water flow}}\right]$
10	4.6	10	2.2
20	2.3	10	4.3
30	1.5	10	6.5

3.5.1 CT 10 min FF 5 %

To reach a CT of 10 min the inlet flow was set to 4.6 L min⁻¹ and the FF to 0.23 L min⁻¹. Two samples were taken from the inlet, and from both the effluent water and the foam at the timesteps 5, 10, 15 and 20 min. Since no samples from this experiment were sent to the ALS Scandinavia, this experiment is excluded from the results based on those samples.

3.5.2 CT 20 min FF 5 %

To reach a CT of 20 min the inlet flow was set to 2.3 L min⁻¹ and the FF to 0.11 L min⁻¹. Three samples were taken from the inlet and both the effluent water and the foam at the timesteps 15 and 45 min.

3.5.3 CT 30 min FF 5 %

To reach a CT of 30 min the inlet flow was set to 1.5 L min⁻¹ and the FF to 0.077 L min⁻¹. Three samples were taken from the inlet and both the effluent water and the foam at the timesteps 20 and 60 min.

3.5.4 CT 10 min FF 10 %

To reach a CT of 10 min the inlet flow was set to 4.6 L min⁻¹ and the FF to 0.46 L min⁻¹. One sample was taken from the inlet, and from both the effluent water and foam at

timesteps 15 and 30 min. One sample from the water approximately 5 cm under the foam was taken at the timestep 30 min.

3.5.5 CT 10 min FF 20 %

To reach a CT of 10 min the inlet flow was set to 4.6 L min⁻¹ and the foam flow to 0.92 L min⁻¹. One sample was taken from the inlet, and from both the effluent water and foam at timesteps 15 and 30 min. One sample from the water approximately 5 cm under the foam was taken at the timestep 30 min.

3.5.6 CT 10 min FF 30 %

To reach a CT of 10 min the inlet flow was set to 4.6 L min⁻¹ and the foam flow to 1.4 L min⁻¹. One sample was taken from the inlet, and from both the effluent water and foam at timesteps 15 and 30 min. One sample from the water approximately 5 cm under the foam was taken at the timestep 30 min.

3.6 Formulas used

3.6.1 Experimental setup

The volume [m³] of the leachate in the cylinder was calculated using the following equation:

$$V = \pi * r^2 * h \quad (2)$$

where r is the radius [m] of the cylinder and h [m] is the height of the water level.

Converting the volume in m³ to L is a multiplying factor of 1000.

Using the volume and the time, the volumetric flow rate [L min⁻¹] was calculated by the following equation:

$$Q = \frac{V}{T} \quad (3)$$

where Q is the volumetric flow rate and T [min] is the time.

The CT [min] was calculated using the equation:

$$CT = \frac{V}{Q} \quad (4)$$

The actual foam fraction [%] was calculated as the measured foam flow divided by the inlet flow.

$$\frac{Q_F}{Q_{in}} * 100 \quad (5)$$

where Q_F is the foam flow and Q_{in} is the influent flow.

3.6.2 Analytical part

The mass of the samples [g] was calculated using the weight different between the empty flask and the flask after being filled:

$$Sample\ weight = Filled\ flask - Empty\ flask \quad (6)$$

The removal efficiency was calculated using Equation (7)

$$Re = \left(1 - \frac{C_{ef}}{C_{in}}\right) * 100 \% \Leftrightarrow Re = \frac{C_{in} - C_{ef}}{C_{in}} * 100 \% \quad (7)$$

where C_{in} is the PFAS concentration in the feed water (influent) and C_{ef} is the PFAS concentration in the treated water (effluent). The error bars in the result section shows standard deviation (STDAV), i.e., how much the samples differ from the average. The standard deviation was calculated with the following equation

$$\sigma = \sqrt{\frac{\sum (x - \bar{x})^2}{n}} \quad (8)$$

where x is the sample value, \bar{x} is the mean value and n is the sample size.

The mass flow, \dot{m} [ng min^{-1}] was calculated taking the flow times the concentration at a certain time step.

$$\dot{m} = Q * C \quad (9)$$

The mass balance as a fraction was calculated in two different ways. The mass balance for different timesteps it was calculated as a fraction of the effluent and foam at that single timestep. The mass balance for the whole run was calculated as an estimation of the total (a) effluent and (b) foam mass flow divided by the total influent mass flow. This was calculated using the average foam and effluent flow for the whole run.

$$a) \text{ massbalance} = \frac{\dot{m}_{ef}}{\dot{m}_{in}} \quad b) \text{ massbalance} = \frac{\dot{m}_f}{\dot{m}_{in}} \quad (10a \ \& \ 10b)$$

where \dot{m}_{ef} is the effluent mass flow, \dot{m}_f is the foam mass flow and \dot{m}_{in} is the influent mass flow (all for both total mass flow and mass flow at a timestep).

The enrichment factors, (a) E_u (underwater) and (b) E_f (foam) was calculated as fractions of foam and underwater concentrations divided by the inlet concentration.

$$a) E_u = \frac{C_u}{C_{in}} \quad b) E_f = \frac{C_f}{C_{in}} \quad (11a \ \& \ 11b)$$

where C_u is the PFAS concentration in the water under the foam,

The relationship between the removal efficiency and both the concentration of iron and conductivity are plotted as well to see if there were any clear connections between the parameters and the removal efficiency.

4 Results

Graphs of the RE for all detected compounds and their standard deviation can be found in Appendix 3 together with the results from the ALS evaluation.

4.1 CT 10 min FF 5 %

As shown in Figure 7 and Table 9, the removal efficiency was 44 percentage points higher for Σ long chained PFAS than for Σ short chained PFAS. The removal efficiency increased through the whole experiment for all categories except for the Σ short chained PFAS that at two sampling points had a higher concentration (about 100 ng L^{-1}) in the effluent water than in the influent water, which resulted in a negative removal efficiency.

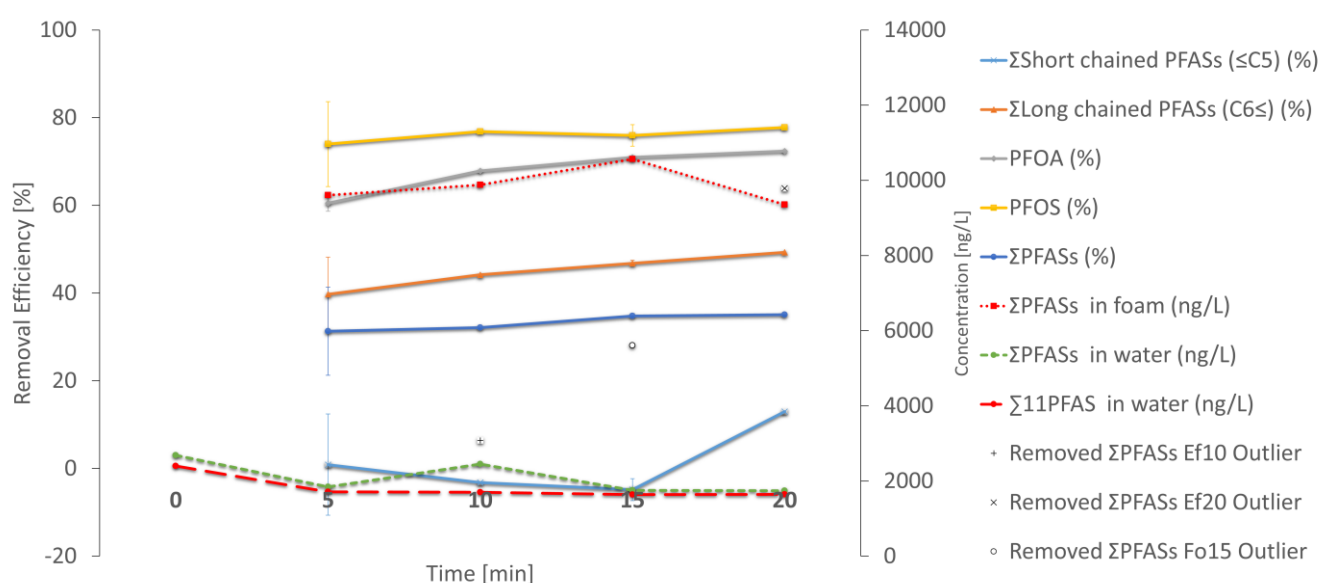


Figure 7 Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L^{-1}) of PFAS in foam and water from the 10 min CT 5 % FF experiment, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

Table 9 Removal efficiency [%] from the CT 10 min FF 5 % experiment

Category\Time [min]	5	10	15	20
Σ Short chained PFAS ($\leq C_5$)	0.8	-3.2	-4.8	13
Σ long chained PFAS ($\geq C_6$)	40	44	47	49
PFOA	61	68	70	72
PFOS	73	77	76	78
Σ PFAS	31	32	33	35

The results for the three outliers, the 10 min effluent sample from run 2 (sample ID 10:5-2Ef10), the 20 min effluent sample from run 2 (sample ID 10:5-2Ef20) and the five min foam sample from run 1 (sample ID 10:5-1F5) were removed. This means that the effluent

10 min, effluent 20 min and foam 15 min are only based on one run instead of two runs as for the other points.

The foam fraction was in between 1.8 and 2.6 %, and an average of 2.3 % (Table 10), so less than half of the aimed foam flow. The individual graphs for RE and standard deviation each compound or found in Appendix Figure 1 in Appendix 3.

Table 10 The average calculated foam fraction (%) for the CT 10 min FF 5 % experiment

Time [min]	5	10	15	20
Actual Foam Fraction [%]	2.6	2.5	2.5	1.8

Since there was a high uncertainty of the results from this experiment, the results are excluded of the comprehensive discussions and figures and are not reliable.

4.2 CT 20 min FF 5 %

As shown in the Figure 8b and Table 11, the removal efficiency was 68 percentage points higher for the Σ long chained PFAS than for the Σ short chained PFAS. The removal efficiency increased from min 15 to 45 for the Σ short chained PFAS and Σ PFAS but decreased for the other categories including concentration of Σ PFAS in the foam. The total Σ PFAS average removal efficiency was 56 % for all runs. The individual graphs for RE and standard deviation for each compound are found in Appendix Figure 2 in Appendix 3.

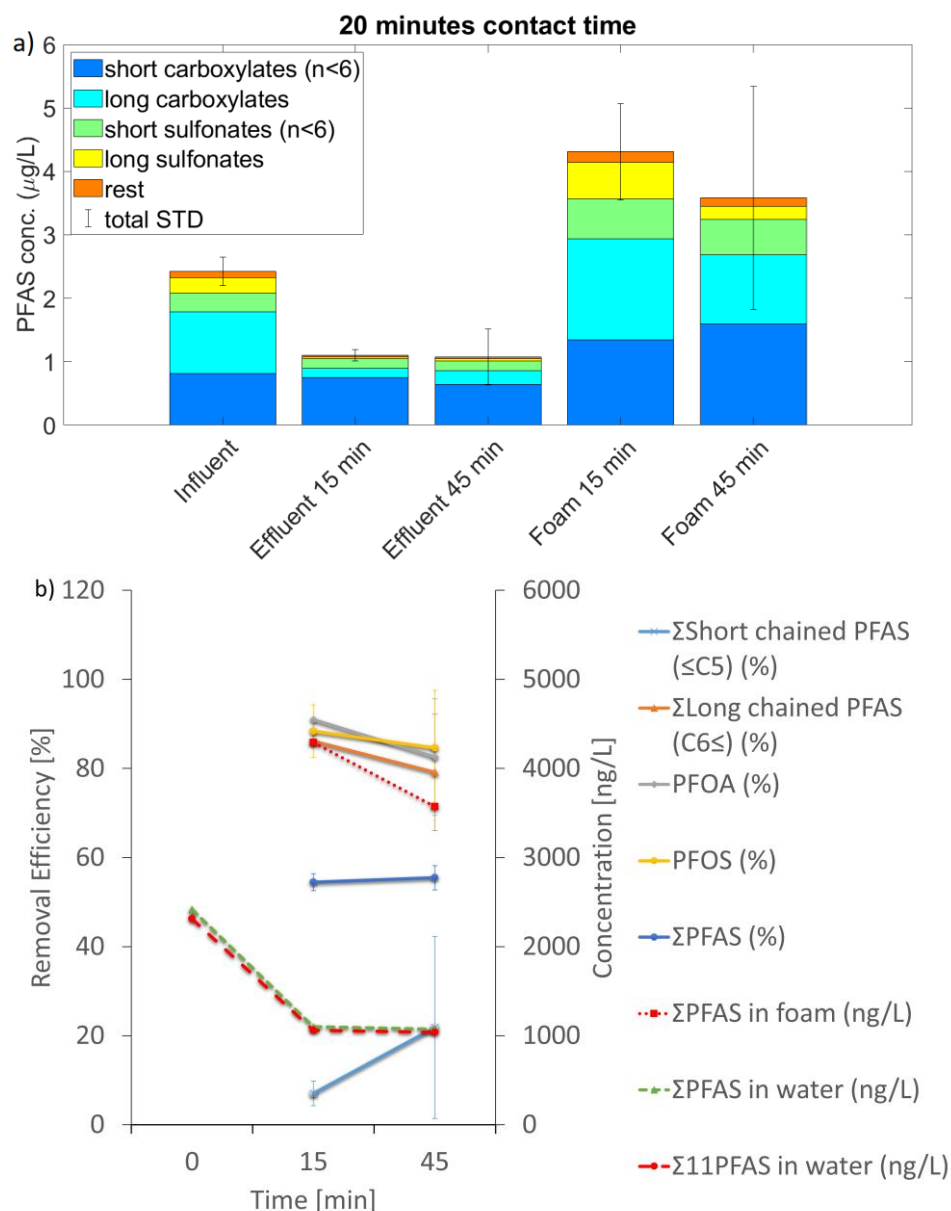


Figure 8 (a) PFAS composition ($\mu\text{g L}^{-1}$) for the influent, effluent and foam for the different time steps for the CT 20 min FF 5 % experiment, colour coding as per insert legend. (b) Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L^{-1}) of PFAS in foam and water from the CT 20 min FF 5 % experiment, colour and symbol coding as per insert legend. For both figures, the error bars are showing the standard deviation.

Table 11 Removal efficiency [%] from the CT 20 min FF 5 % experiment

Category\Time [min]	15	45
ΣShort chained PFAS ($\leq C_5$)	7.0	22
Σlong chained PFAS ($\geq C_6$)	86	79
PFOA	91	83
PFOS	88	85
ΣPFAS	55	56

The average actual foam flow was 6.8 % (Table 12), a higher flow than the aimed flow.

Table 12 The average calculated foam fraction (%), the concentration of iron (mg L^{-1}) and the conductivity of the water (mS m^{-1}) for the CT 20 min FF 5 % experiment. The – represents a lack of value to be inserted

Category\Time [min]	0	15	45
Actual Foam Fraction [%]	-	6.9	6.7
C_{Fe} [mg L^{-1}]	5.5	-	4.3
Conductivity [mS m^{-1}]	500	-	12

In addition to the conductivity and concentration of iron (Table 12), more results from the ALS-Scandinavia evaluation for the CT 20 min FF 5 % are found in Appendix Table 3 in Appendix 3. Samples were only sent to the ALS-Scandinavia lab for the influent water and the effluent water at the last timestep and therefore there are no values of the 15 min sample and there was no foam sampling at the very beginning.

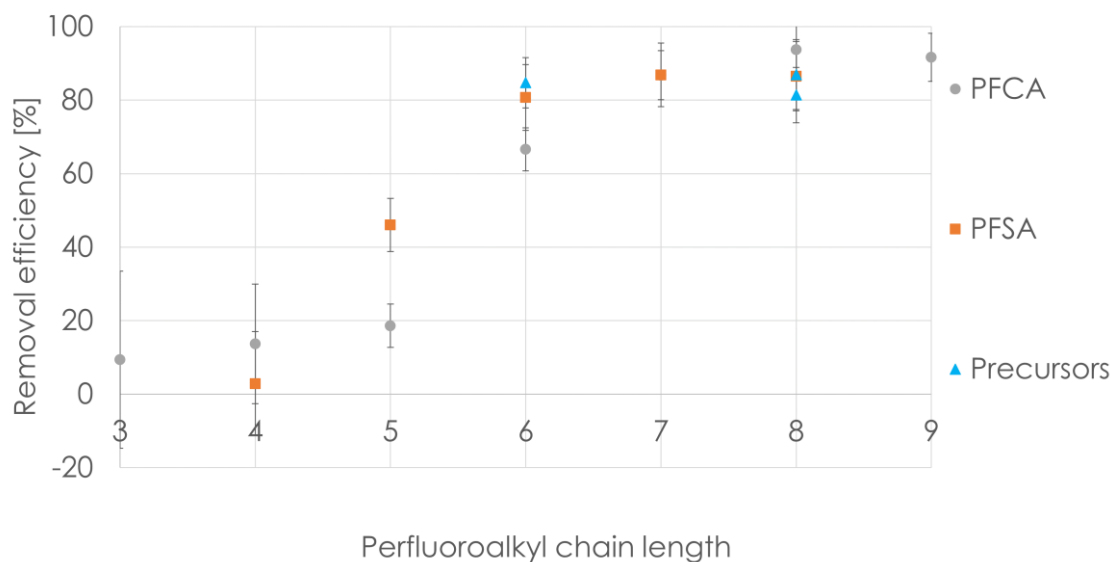


Figure 9 Removal efficiencies (%) for different chain lengths and different classes for the CT 20 min FF 5 %, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

The average RE for PFCA are 54 %, for PFSA 61 % and for precursors 84 % (Figure 9). The average STDAV for PFCA are 10 %, for PFSA 9.7 % and for precursors 7.9 %.

4.3 CT 30 min FF 5 %

As shown in the Figure 10b and Table 13, the removal efficiency was 63 percentage points higher for the Σ long chained PFAS than for the Σ short chained PFAS. The removal efficiency was fairly stable between the sampling points, but the concentration of Σ PFAS in the foam increased with 32 percentage points from 5900 ng L⁻¹ to 7800 ng L⁻¹. The total Σ PFAS average removal efficiency was 59 % for all runs. The individual graphs for RE and standard deviation for each compound are found in Appendix Figure 3 in Appendix 3.

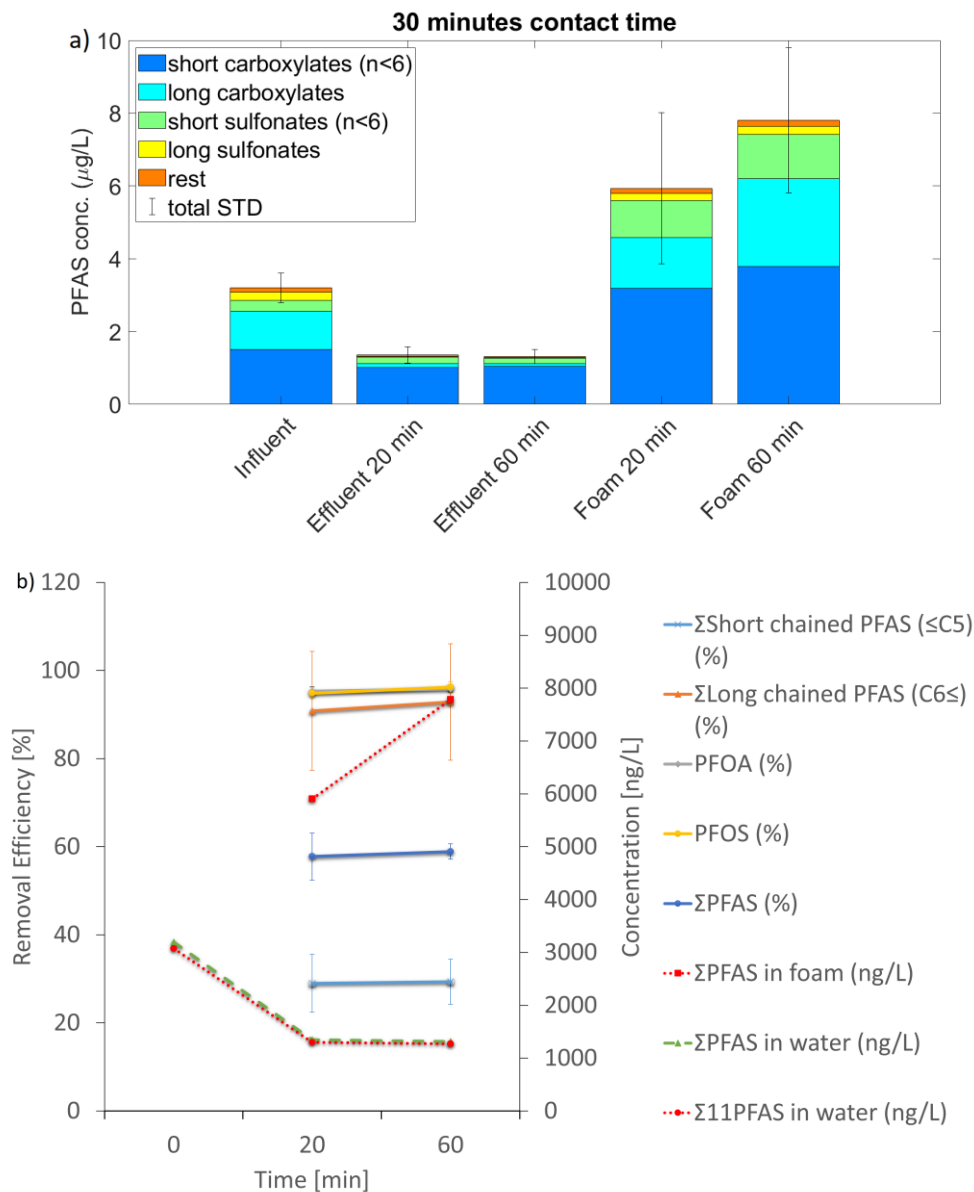


Figure 10 (a) PFAS composition ($\mu\text{g L}^{-1}$) for the influent, effluent and foam for the different time steps for the CT 30 min FF 5 % experiment, colour coding as per insert legend. (b) Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L^{-1}) of PFAS in foam and water from the CT 30 min FF 5 % experiment, colour and symbol coding as per insert legend. For both figures, the error bars are showing the standard deviation.

Table 13 Removal efficiency [%] from the CT 30 min FF 5 % experiment

Category\Time [min]	20	60
ΣShort chained PFAS ($\leq C_5$)	29	29
ΣLong chained PFAS ($\geq C_6$)	91	93
PFOA	95	96
PFOS	95	96
ΣPFAS	58	59

The average foam flow was 6.4 % (Table 14), a little higher than the aimed flow.

Table 14 The average calculated foam fraction (%), the concentration of iron (mg L^{-1}) and the conductivity of the water (mS m^{-1}) for the CT 30 min FF 5 % experiment. The - represents a lack of value to be inserted

Category\Time [min]	0	20	60
Actual Foam Fraction [%]	-	5.7	7.0
C_{Fe} [mg L^{-1}]	4.5	-	3.7
Conductivity [mS m^{-1}]	520	-	550

In addition to the conductivity and concentration of iron, more results from the ALS-Scandinavia evaluation for the CT 30 min FF 5 % are found in Appendix Table 4 in Appendix 3. Samples were only sent to the ALS-Scandinavia lab for the influent water and the effluent water at the last timestep and therefore there are no values of the 20 min sample and there was no foam sampling at the very beginning.

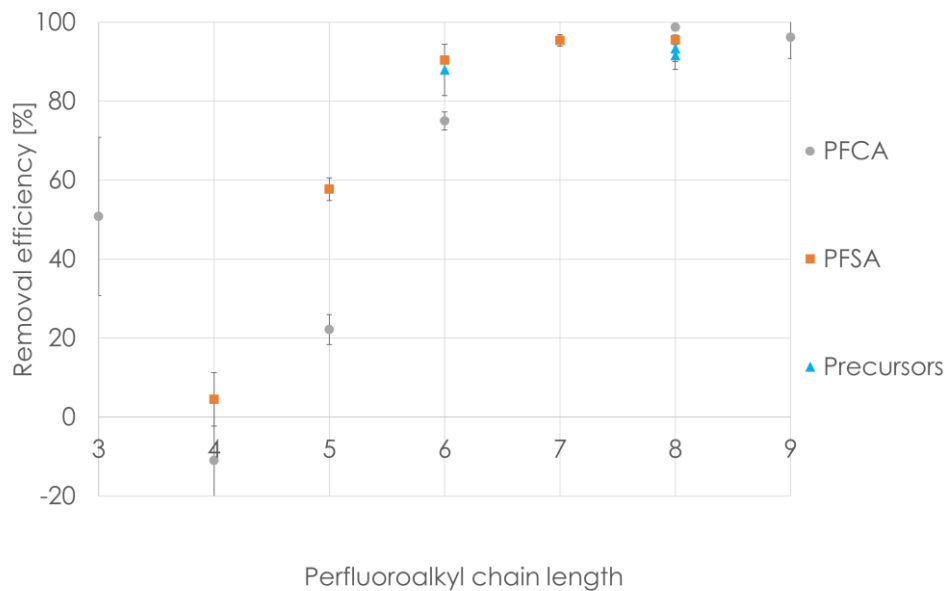


Figure 11 Removal efficiencies (%) for different chain lengths and different classes for the CT 30 min FF 5 %, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

The average RE for PFCA are 61 %, for PFSA 69 % and for precursors 91 % (Figure 11). The average STDAV for PFCA are 6.8 %, for PFSA 3.0 % and for precursors 4.0 %.

4.4 CT 10 min FF 10 %

As shown in Figure 12b and Table 15, the removal efficiency was 65 percentage points higher for the Σ long chained PFAS than for the Σ short chained PFAS. The concentration of Σ PFAS in the foam increased with 10 percentage points from 2600 ng L⁻¹ to 2800 ng L⁻¹ with time while the other categories were fairly stable through the experiment. The total Σ PFAS average removal efficiency was 53 % for all runs. The individual graphs for RE and standard deviation for each compound are found in Appendix Figure 4 in Appendix 3.

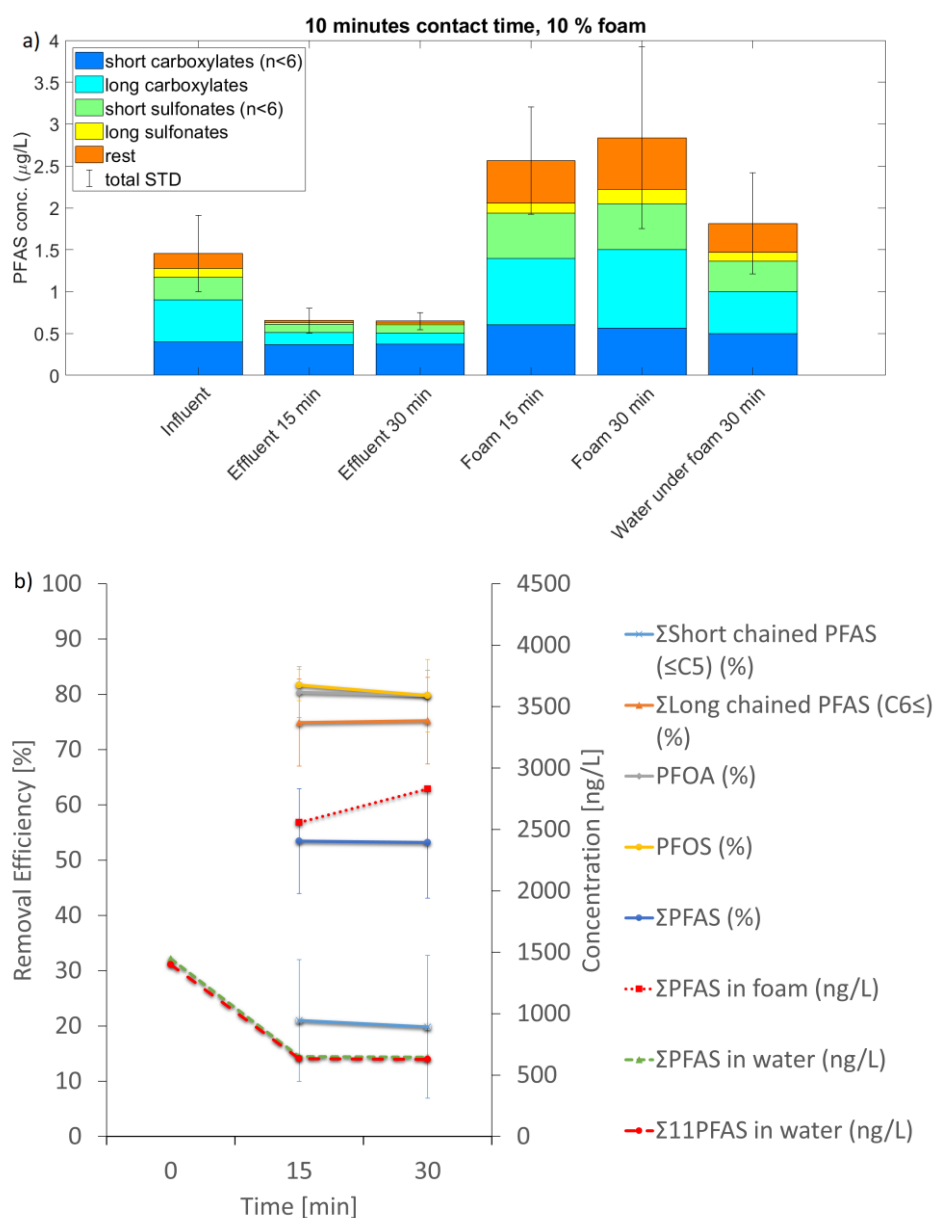


Figure 12 (a) PFAS composition ($\mu\text{g L}^{-1}$) for the influent, effluent and foam for the different time steps for the CT 10 min FF 10 % experiment, colour coding as per insert legend. (b) Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L⁻¹) of PFAS in foam and water from the CT 10 min FF 10 % experiment, colour and symbol coding as per insert legend. For both figures, the error bars are showing the standard deviation.

Table 9 Removal efficiency [%] from the CT 10 min FF 10 % experiment

Category\Time [min]	15	30
ΣShort chained PFAS ($\leq C_5$)	21	20
Σlong chained PFAS ($\geq C_6$)	75	75
PFOA	80	80
PFOS	82	80
ΣPFAS	53	53

In addition to the conductivity and concentration of iron (Table 16), more results from the ALS-Scandinavia evaluation for the CT 10 min FF 10 % are found in Appendix Table 5 in Appendix 3.

Table 16 The concentration of iron (mg L^{-1}) and the conductivity (mS m^{-1}) of the water for the CT 10 min FF 10 % experiment

Category\Time [min]	0	30
C_{Fe} [mg L^{-1}]	4.0	3.9
Conductivity [mS m^{-1}]	320	330

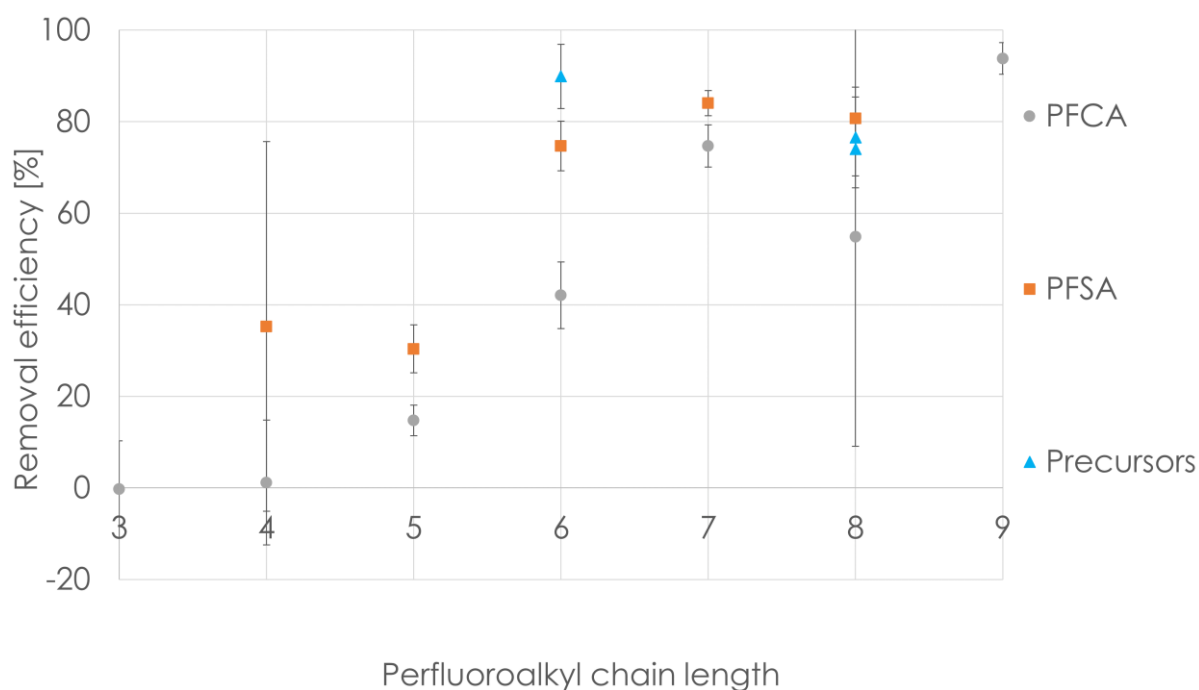


Figure 13 Removal efficiencies (%) for different chain lengths and different classes for the CT 10 min FF 10 %, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

The average RE for PFCA are 40 %, for PFSA 61 % and for precursors 80 % (Figure 13). The average STDAV for PFCA are 13 %, for PFSA 12 % and for precursors 8.0 %.

4.5 CT 10 min FF 20 %

As shown in Figure 14b and Table 17 the removal efficiency was 72 percentage points higher for the Σ long chained PFAS than for the Σ short chained PFAS. The removal efficiency of Σ short chained PFAS were negative, but all other categories were quite stable throughout the experiment. The total Σ PFAS average removal efficiency was 41 % for all runs. The individual graphs for RE and standard deviation for each compound are found in Appendix Figure 5 in Appendix 3.

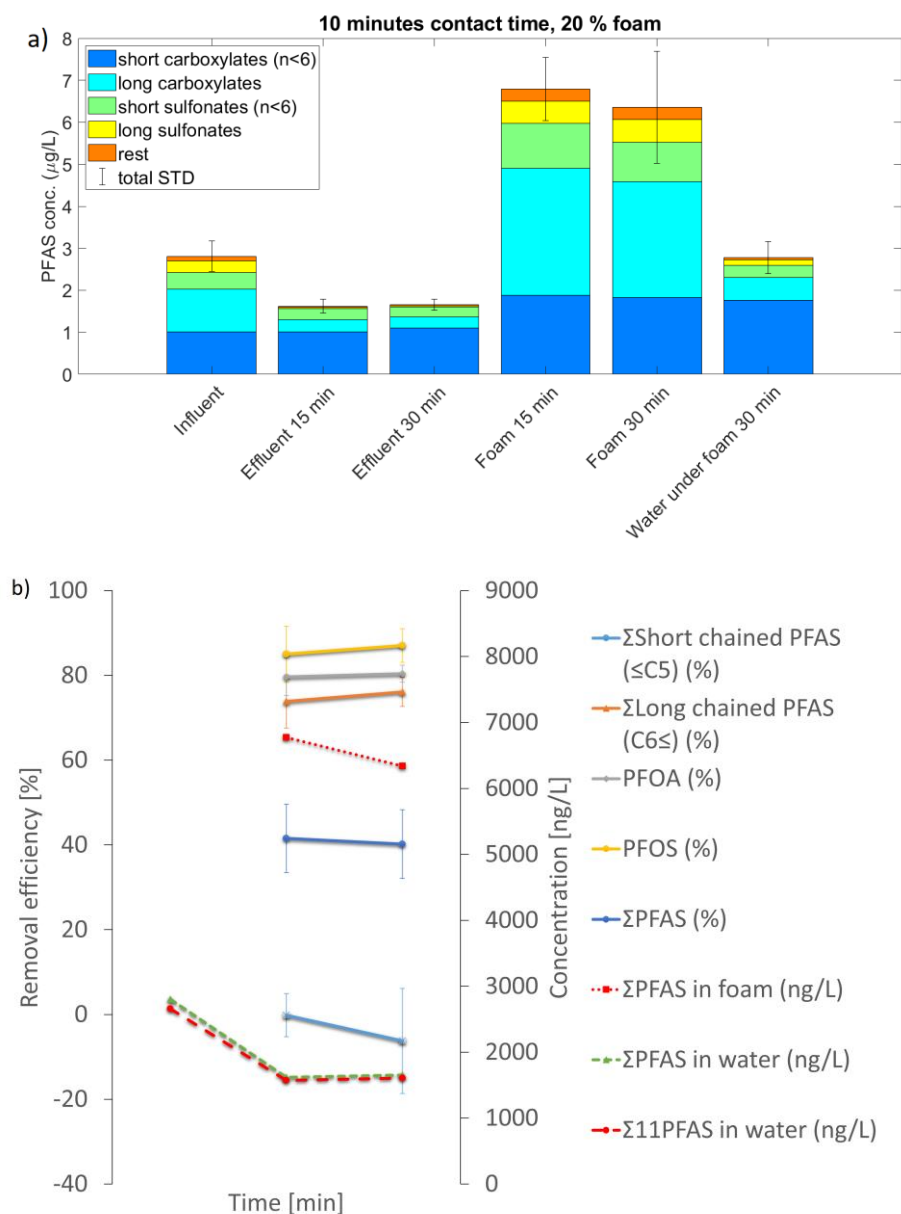


Figure 14 (a) PFAS composition ($\mu\text{g L}^{-1}$) for the influent, effluent and foam for the different time steps for the CT 10 min FF 20 % experiment, colour coding as per insert legend. (b) Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L^{-1}) of PFAS in foam and water from the CT 10 min FF 20 % experiment, colour and symbol coding as per insert legend. For both figures, the error bars are showing the standard deviation.

Table 17 Removal efficiency [%] from the CT 10 min FF 20 % experiment

Category\Time [min]	15	30
ΣShort chained PFAS ($\leq C_5$)	-0.16	-6.3
ΣLong chained PFAS ($\geq C_6$)	74	76
PFOA	80	80
PFOS	85	87
ΣPFAS	42	40

In addition to the concentration of iron and conductivity (Table 18), more results from the ALS-Scandinavia evaluation for the CT 10 min FF 10 % are found in Appendix Table 6 in Appendix 3.

Table 18 The concentration of iron (mg L^{-1}) and the conductivity (mS m^{-1}) of the water for the CT 10 min FF 20 % experiment

Category\Time [min]	0	30
C_{Fe} [mg L^{-1}]	5.0	5.1
Conductivity [mS m^{-1}]	520	510

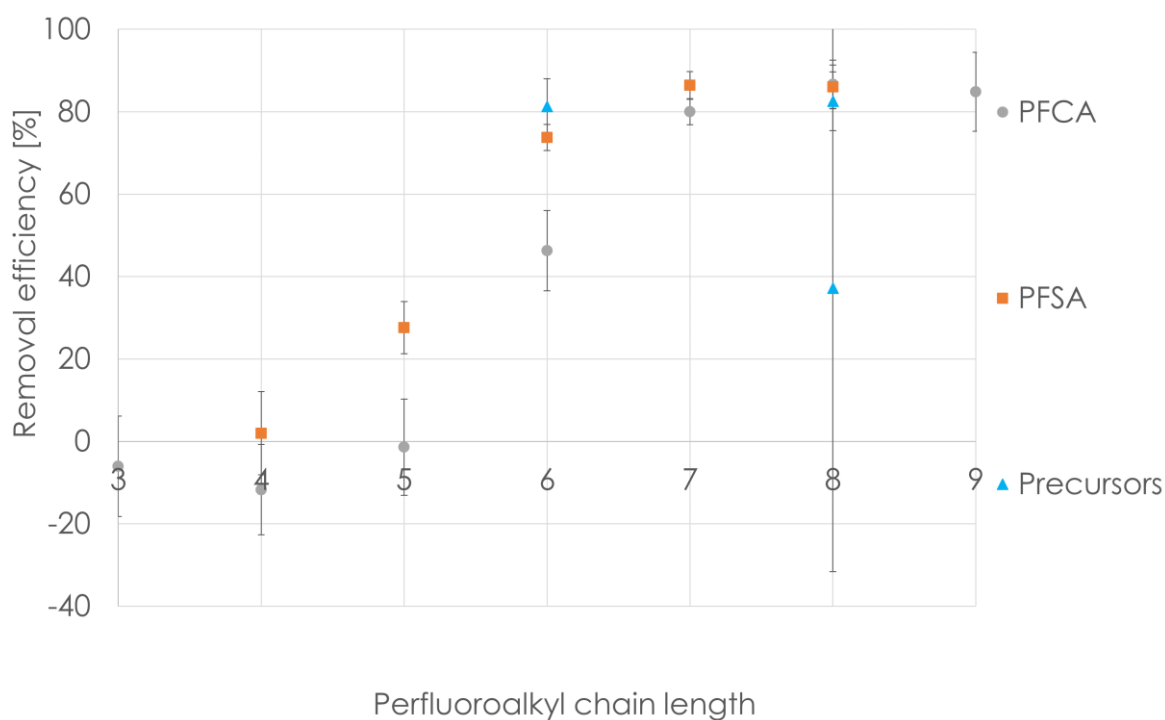


Figure 15 Removal efficiencies (%) for different chain lengths and different classes for the CT 10 min FF 20 %, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

The average RE for PFCA are 40 %, for PFSA 55 % and for precursors 67 % (Figure 15). The average STDAV for PFCA are 9.0 %, for PFSA 6.0 % and for precursors 28 %.

4.6 CT 10 min FF 30 %

As shown in Figure 16b and Table 19 the removal efficiency was 58 percentage points higher for Σ long chained PFAS than it was for Σ short chained PFAS. The concentrations were stable between the timesteps for all categories except for the Σ long chained PFAS that had 7 percentage points higher removal efficiency for the second timestep. The total Σ PFAS average removal efficiency was 46 % for all runs. The individual graphs for RE and standard deviation for each compound are found in Appendix Figure 6 in Appendix 3.

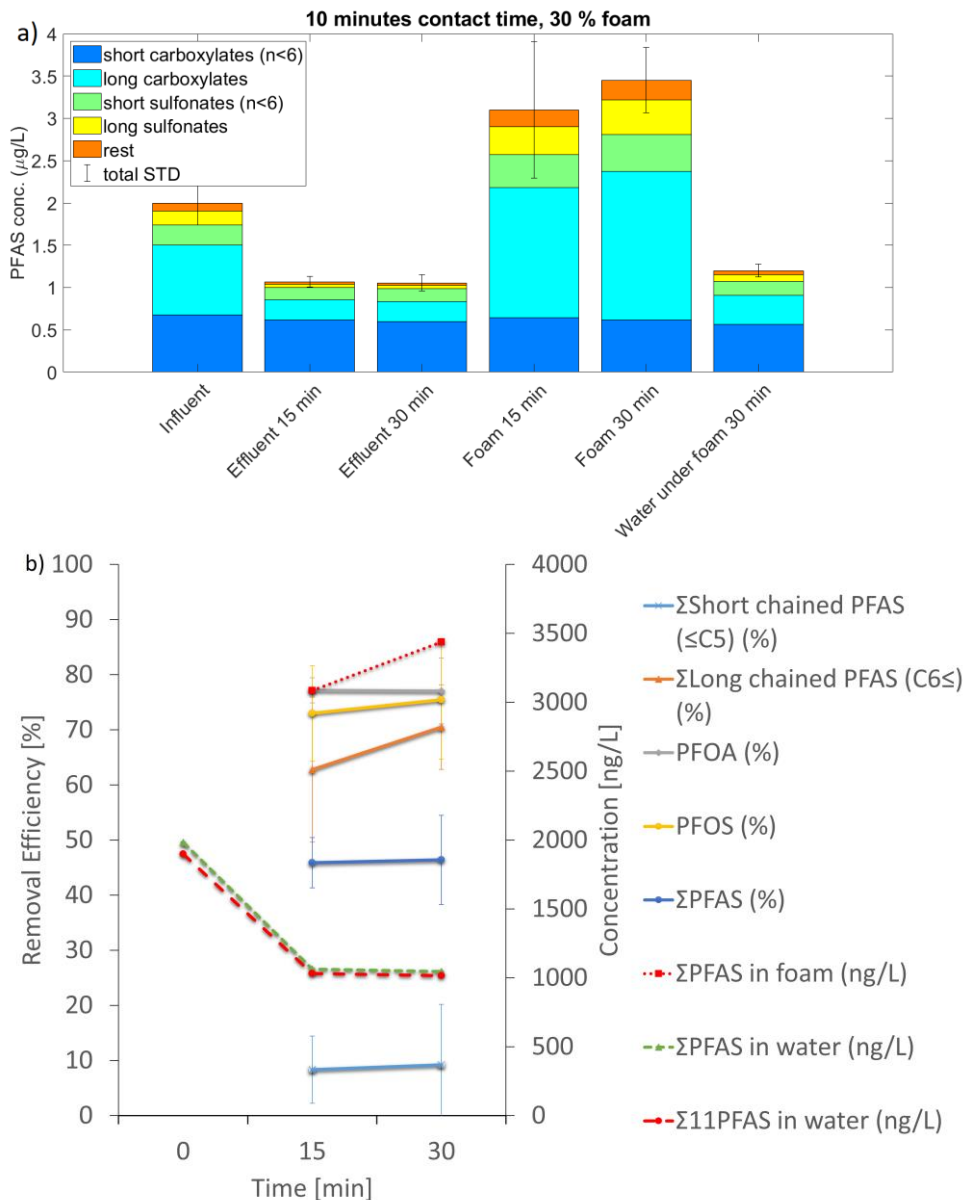


Figure 16 (a) PFAS composition ($\mu\text{g L}^{-1}$) for the influent, effluent and foam for the different time steps for the CT 10 min FF 30 % experiment, colour coding as per insert legend. (b) Average removal efficiency (%) as a function of time (min) for Σ PFAS, Σ short chained PFAS ($\leq C_5$), Σ long chained PFAS ($\geq C_6$), PFOA, PFOS, and average concentrations (ng L^{-1}) of PFAS in foam and water from the CT 10 min FF 30 % experiment, colour and symbol coding as per insert legend. For both figures, the error bars are showing the standard deviation.

Table 19 Removal efficiency [%] from the CT 10 min 30 % FF experiment

Category\Time [min]	15	30
ΣShort chained PFAS ($\leq C_5$)	8.3	9.2
ΣLong chained PFAS ($\geq C_6$)	63	70
PFOA	77	77
PFOS	73	75
ΣPFAS	46	46

In addition to the concentration of iron and conductivity (Table 20), more results from the ALS-Scandinavia evaluation for the CT 10 min FF 30 % are found in Appendix Table 7 in Appendix 3.

Table 20 The concentration of iron (mg L^{-1}) and the conductivity (mS m^{-1}) of the water for the CT 10 min FF 30 % experiment

Category\Time [min]	0	30
C_{Fe} [mg L^{-1}]	5.1	4.5
Conductivity [mS m^{-1}]	400	430

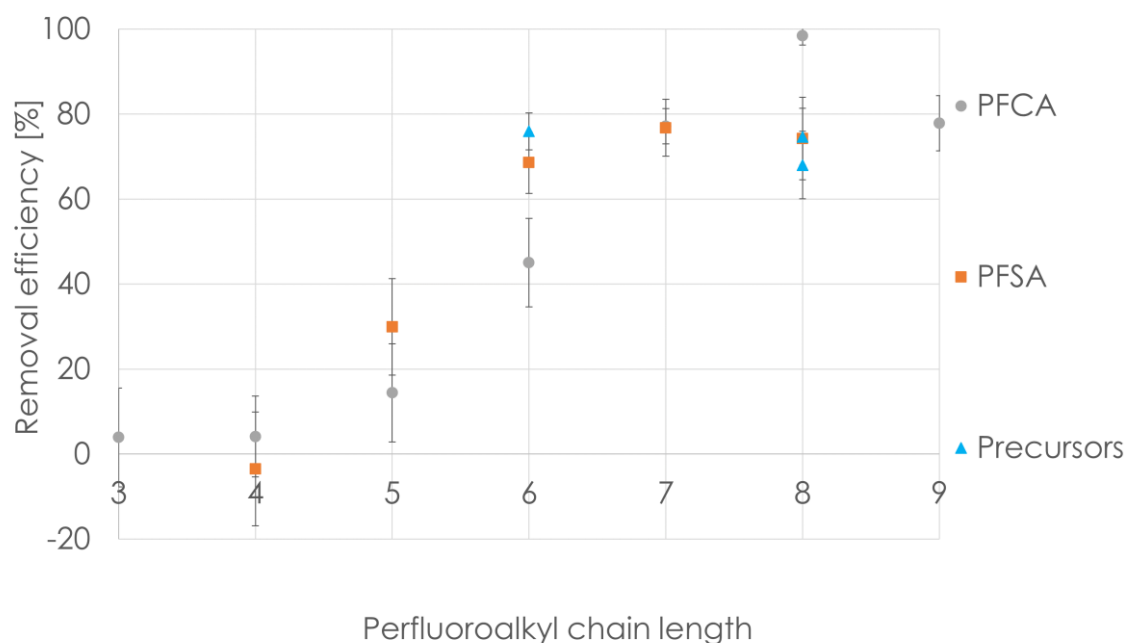


Figure 17 Removal efficiencies (%) for different chain lengths and different classes for the CT 10 min FF 30 %, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

The average RE for PFCA are 46 %, for PFSA 49 % and for precursors 68 % (Figure 17). The average STDAV for PFCA are 8.0 %, for PFSA 10 % and for precursors 6.0 %.

4.7 Comprehensive and comparative results

4.7.1 Removal efficiencies and concentrations

Figure 18 demonstrates a comparison of the average PFAS removal efficiency for all experiments. As illustrated the removal efficiency for Σ PFAS increase with an average of 3 % when changing from CT 20 min to CT 30 min. Comparing the CT 10 min removal efficiencies the removal efficiency is highest for the CT 10 min FF 10 % (53 %) and lowest for the CT 10 min FF 20 % (41 %).

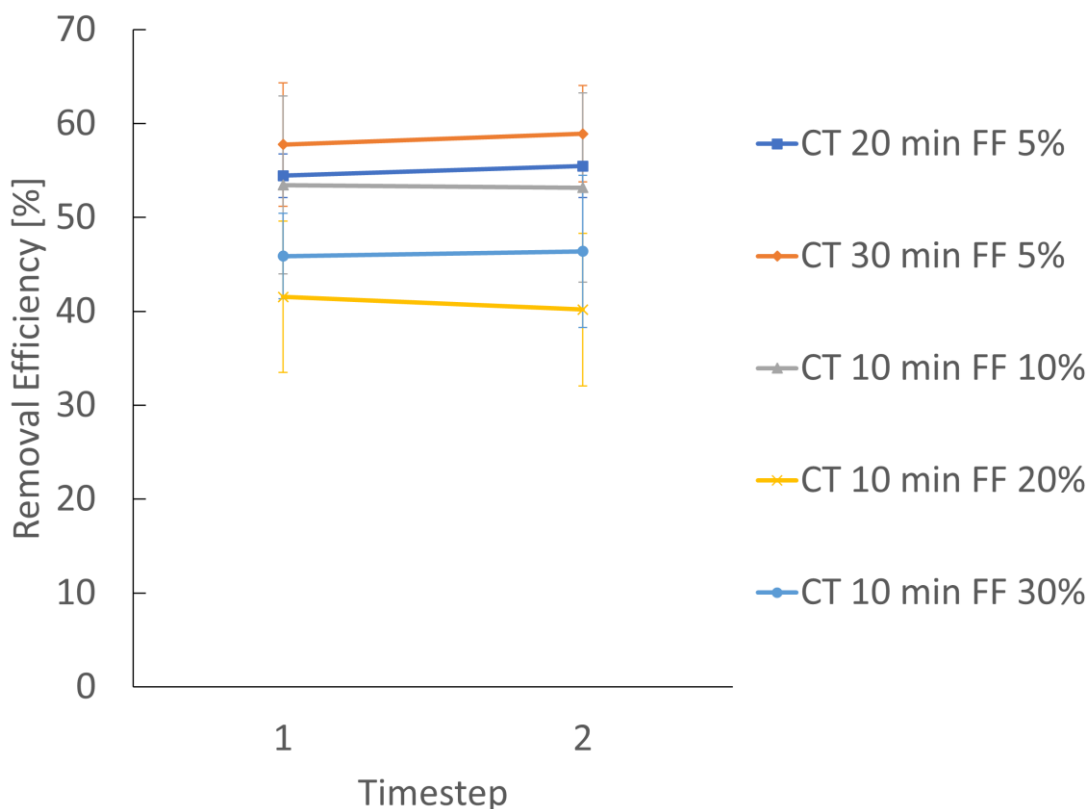


Figure 18 Average Σ PFAS removal efficiency (%) for all experiments at each timestep described in method section 3.5, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

Comparing Figure 8a and Figure 10a a higher contact time led to an increase of short compounds in the foam. The 30 min contact time had the highest removal of all experiments and mostly short carboxylates and short sulfonates in the effluent water. As seen in each experiment individual result graphs above, the difference in concentration of Σ PFAS in the water and Σ_{11} PFAS in the water is very small, since the majority of the detected PFAS are listed as Σ_{11} PFAS.

As shown in Figures 8a, 10a, 12a, 14a and 16a there are barely any long sulfonates, or the components categorised as *rest* in the effluent water in any experiment. For all the CT 10 min there is an enrichment of short sulfonates in the foam and a smaller enrichment of short carboxylates in the foam. For all experiments, the concentration of carboxylates were higher than the concentration of sulfonates.

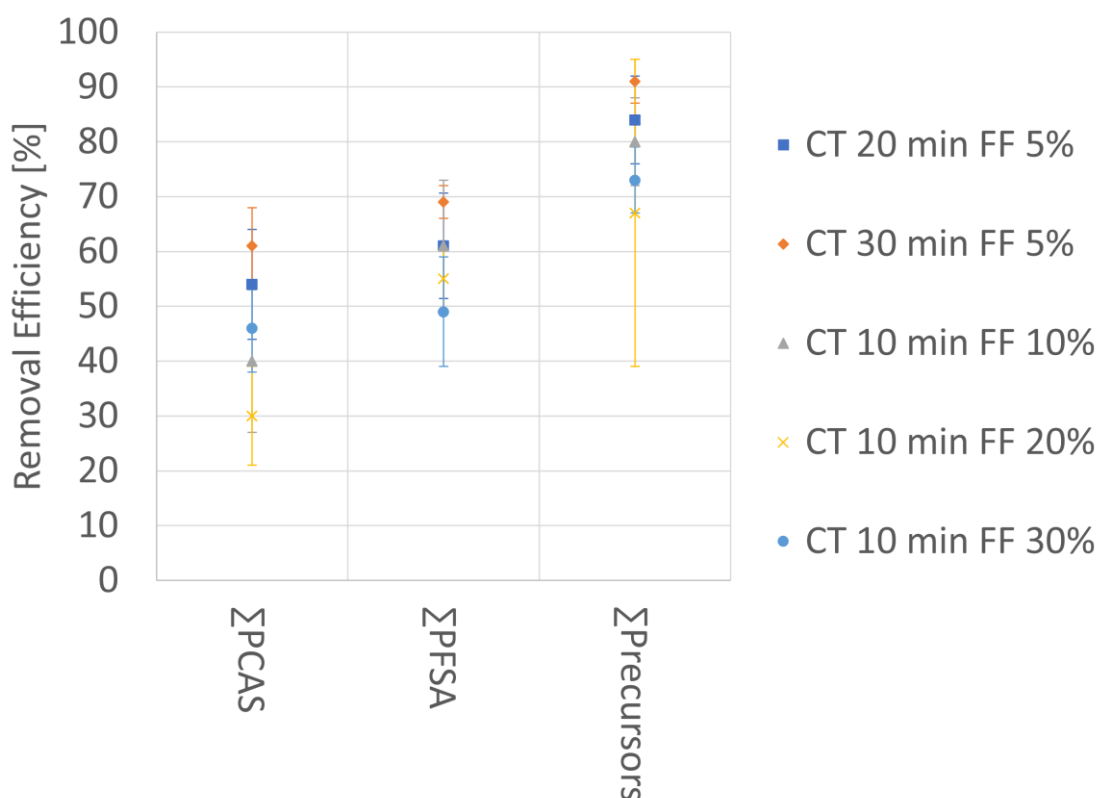


Figure 19 Removal efficiencies (%) for the different classes within the different experiments, colour and symbol coding as per insert legend. The error bars are showing the standard deviation.

From Figure 19, each experiment had the highest RE for precursors (total average 78 %), second highest for PFSA (total average 59 %) and lowest for PFCA (total average 48 %).

Table 10 Comparison between different inlet concentrations (ng L^{-1}), average removal efficiencies (%) for different runs and percentage of long chained PFAS (%). The highest and lowest numbers for each experiment are coloured in orange (highest) and red (lowest)

Experiment	CT 20 min FF 5 %			CT 30 min FF 5 %					
Run	1	2	3	1	2	3			
C_{in} [ng L^{-1}]	2200	2500	2500	3200	3200	3200			
RE_{avg} [%]	52	57	55	57	64	54			
PFAS $\geq C_6$ [%]	55	62	62	46	42	46			
Experiment	CT 10 min FF 10 %			CT 10 min FF 20 %			CT 10 min FF 30 %		
Run	1	2	3	1	2	3	1	2	3
C_{in} [ng L^{-1}]	950	1700	1700	2400	2900	3100	1800	1800	2300
RE_{avg} [%]	43	51	66	31	42	49	43	40	55
PFAS $\geq C_6$ [%]	60	50	67	53	55	61	61	57	60

As shown in Table 21 four of the five experiments had the lowest removal efficiency for the run with the lowest inlet concentration and four of the five experiments had the highest removal efficiency for the run with the highest inlet concentration. Two of the experiments had the highest removal efficiency for the runs with highest percentage of long chained PFAS, but both runs also had the highest total inlet concentration.

Two of the experiments had the lowest removal efficiency for the runs with the lowest percentage of long chained PFAS, but they also had the lowest inlet concentration.

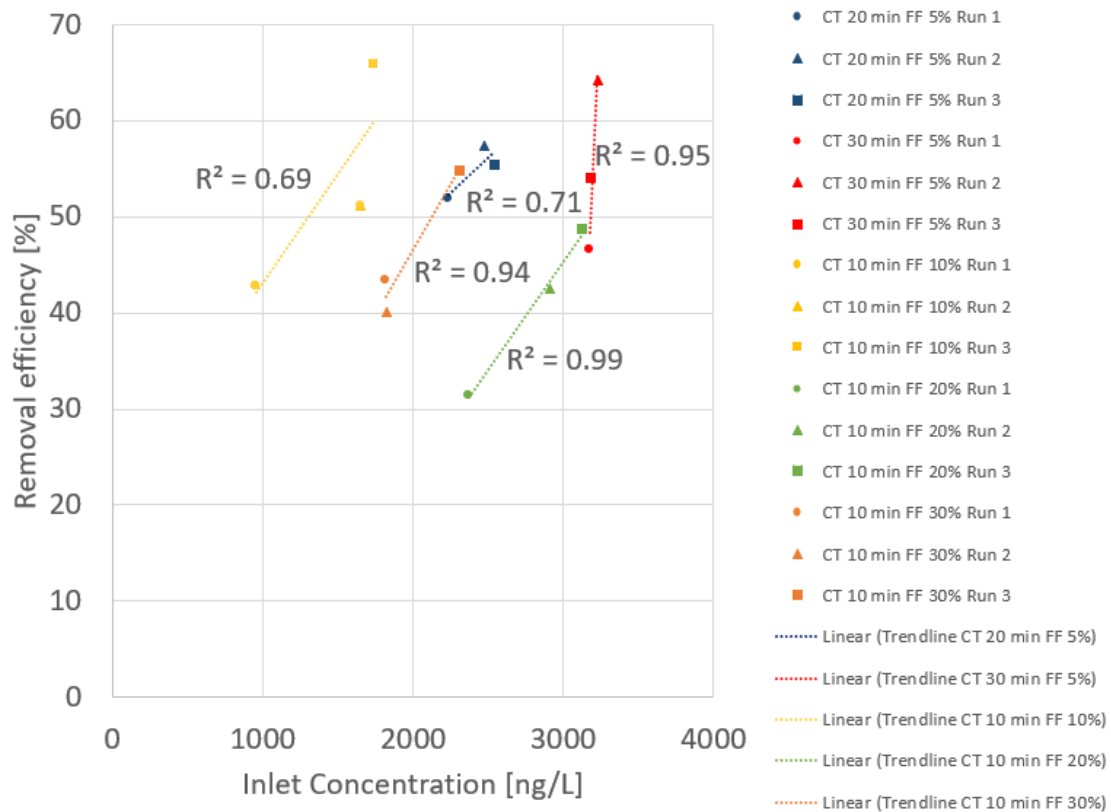


Figure 20 Relations between inlet PFAS concentrations (ng L^{-1}) and removal efficiency (%) for the different runs with trendlines and R^2 -values, colour and symbol coding as per insert legend.

Figure 20 is a scatter plot of the relations between inlet concentration and removal efficiency. The R^2 -value is above 0.9 for CT 30 min FF 5 % and CT 10 min 20 and 30 % and close to 0.7 for CT 20 min FF 5 % and CT 10 min FF 10 %.

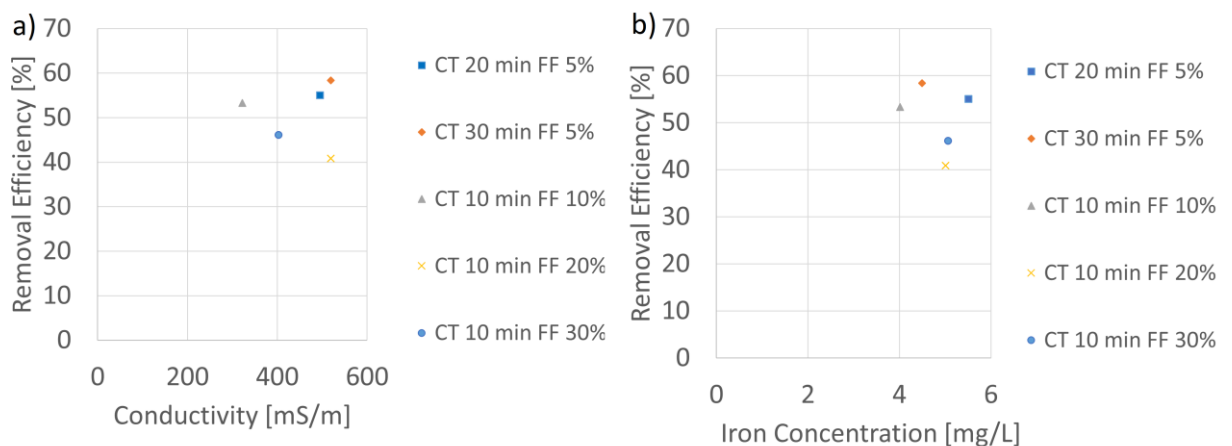


Figure 21 The removal efficiency (%) in relation to (a) the inlet conductivity (mS m^{-1}) and (b) inlet concentration of iron (mg L^{-1}) for the different experiments, colour and symbol coding as per insert legend.

The relationship between the average removal efficiency and conductivity is plotted to the left (Figure 21a) and the average removal efficiency and iron concentration is plotted to the right (Figure 21b) for each experiment. No clear connections between iron concentration/conductivity and removal efficiency are shown Figure 21.

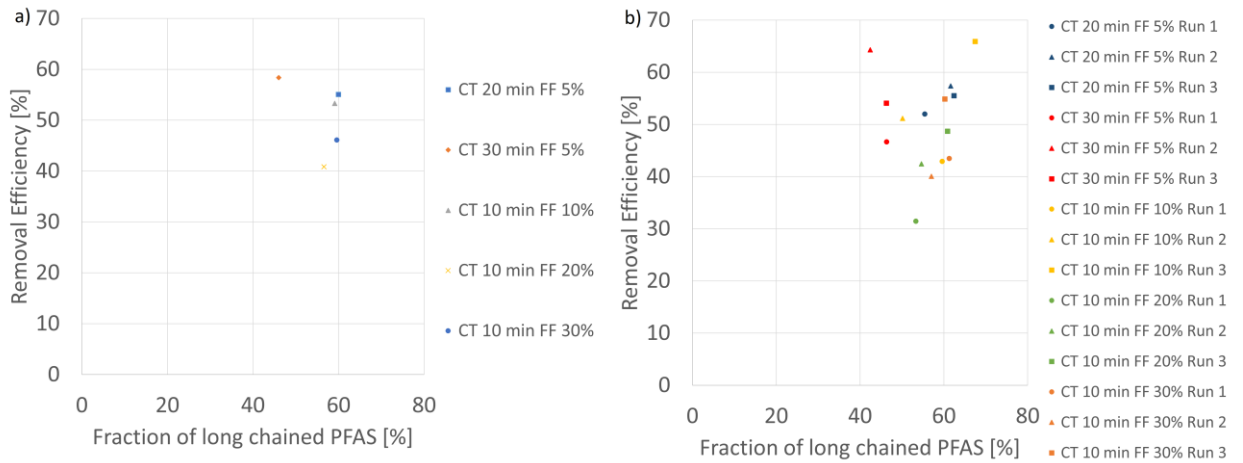


Figure 22 The relationship between the fraction of long chained PFAS and removal efficiency (%) for (a) the different experiment and (b) the different runs, colour and symbol coding as per insert legend.

The relation between the RE and fraction of long chained PFAS for the different experiments are shown in Figure 22a. The fraction of long chained PFAS were similar for all experiments except for the CT 30 min FF 5 % where the fraction was lower, but the experiment had the highest removal efficiency. Within the experiments, the different runs (Figure 22b) had no clear connections between fraction of long chained PFAS and removal efficiency.

4.7.2 Mass Balance and Enrichment

The enrichment in the underwater and foam is shown as a fraction of the influent water in Figure 23. The enrichment in the underwater is higher for short chained PFAS than for short chained PFAS. The enrichment decreases with an increasing foam fraction for long chained PFAS but has its maximum (1.6) in for the short chained PFAS in the experiment with 20 % foam fraction. The enrichment in the foam is higher for long chained PFAS and the highest enrichment (2.6) is in the experiment with 20 % foam fraction as well.

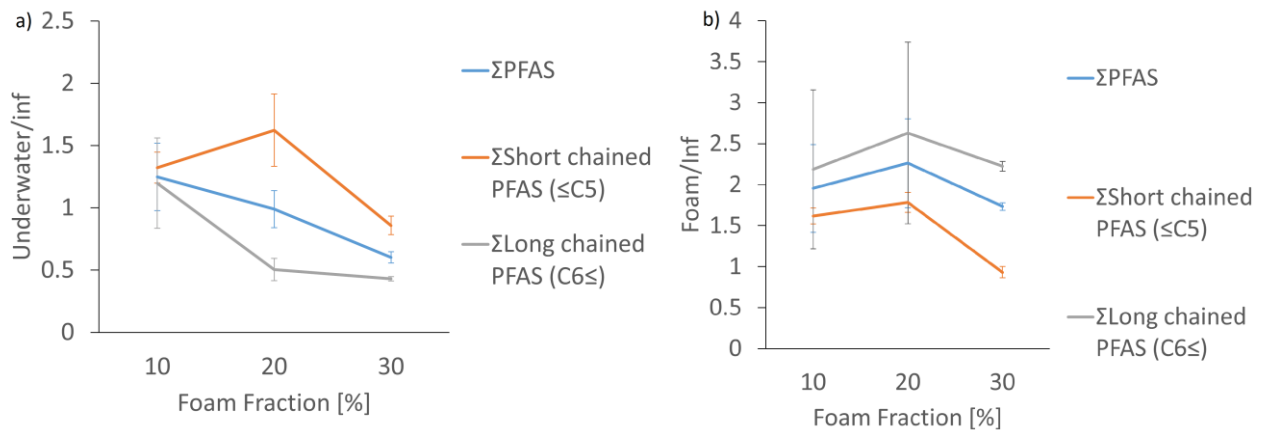


Figure 23 Average enrichment in the (a) underwater and (b) foam in relation to the influent water for CT 10 min 10, 20 and 30 % runs, colour coding as per insert legend. The error bars are showing the standard deviation.

As shown in Figure 24 a higher foam fraction results in a higher mass flow in the foam. The percentage of the effluent mass flow is higher than for the foam flow for all categories except for the CT 10 min FF 30 %.

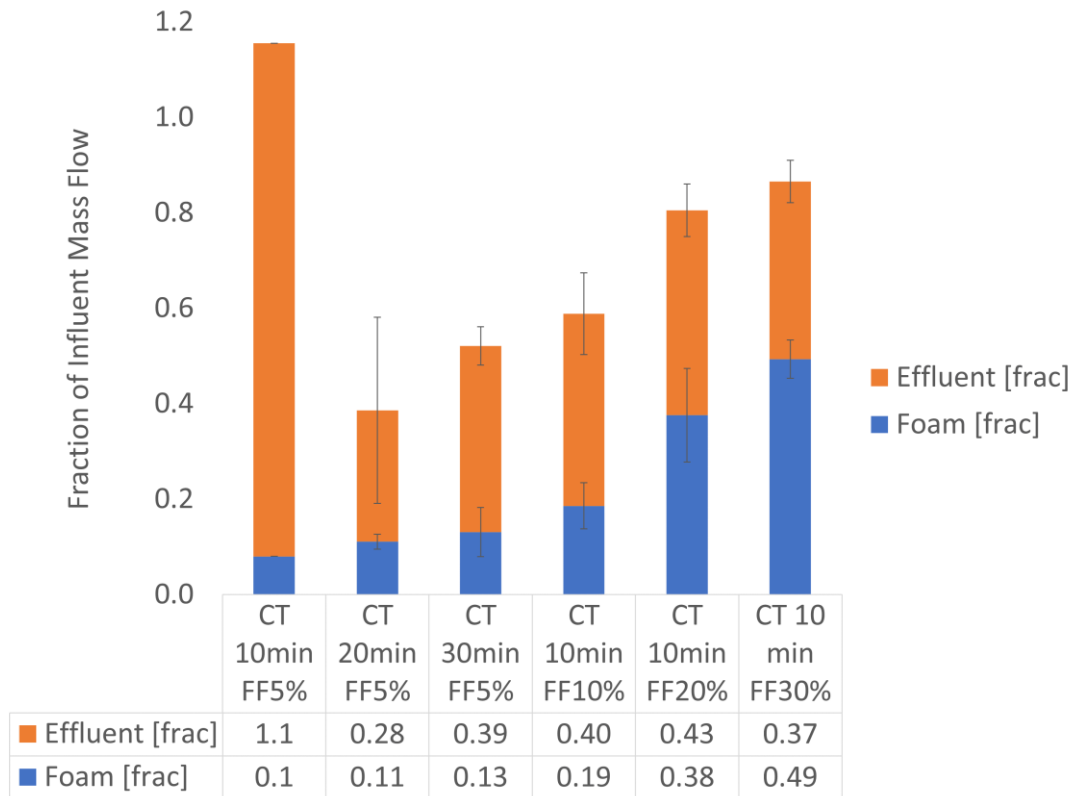


Figure 4 The average mass balance as a fraction of the total mass flow of the effluent and foam divided by the total influent mass flow. The orange bars are the effluent mass flow, the blue bars are the foam mass flow and the error bars are showing the standard deviation. The numbers under the bar chart are the fraction of effluent flow and foam mass flow.

The mass balances at single timesteps are shown in the bar charts in Figure 25 instead as of for the whole experiments as in Figure 24. The graphs confirms that a higher foam fraction results in a higher mass balance fraction. The CT 10 min FF 5 % has big fluctuations in its mass balance and varies between 0.7 and 2.

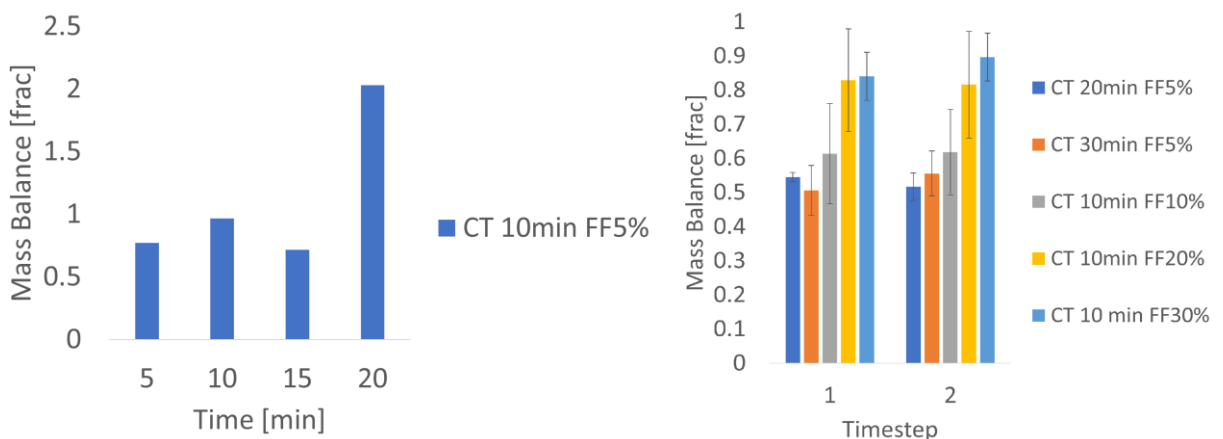


Figure 5 Mass balance for (a) the CT 10 min FF5 % as a function of time and (b) the other experiments for different time steps. The mass balance is shown as the fraction of foam and effluent flow compared to the influent flow. The error bars are showing the standard deviation

5 Discussion

5.1 Removal Efficiency and Concentrations

The removal efficiency of short chained PFAS ($\leq C_5$) was less effective than for long chained PFAS ($\geq C_6$) in all experiments. The average removal efficiency for short chained PFAS were 10 % while it was 67 % for long chained PFAS. The difference of 57 percentage points can confirm Meng et al. (2018) theory that the lower surface activity of the short-chain PFAS led to a lower removal efficiency. This was as predicted and the most throughout trend in this study.

Since each experiment had the highest removal efficiency for precursors (total average 78 %), second highest for PFSA (total average 59 %) and lowest for PFCA (total average 48 %), the results from this study are unambiguous. One thing to keep in mind before drawing any conclusions is that the PFCA and PFSA has an average chain length of 6 while the precursors have an average chain length of 7.3, which could be an explanation to the higher removal efficiency.

The removal efficiency did not increase with elapsed time within the experiment. If each experiment was divided into equally big parts, the removal efficiency for \sum PFAS were 1 % higher for the second half (45 % for the first and 46 % for the second part). For \sum long chained PFAS the average removal efficiency was 78 % for the first half and 81 percent for the second part of the experiment. Kjellgren (2020) had an increasing removal efficiency with time but found steady state within the first 20 min of each continuous experiment. Meng (2018) also had an increased removal efficiency with an increasing time and obtained steady state between 90 and 120 min. To detect when steady state was obtained in this study sampling at more timesteps would have been necessary in the earlier parts of the experiments.

The total average \sum PFAS removal efficiency was 46 %, which was in general lower than for Kjellgren (2020) continuous experiments that was 86 %. The air flow rate of 10 L min^{-1} for a column of 46 L (this experiment) has a lower fraction than Kjellgren (2020) with 2 L min^{-1} and a column of 2.4 L. That could be one explanation since a higher air flow rate leads to a higher removal efficiency (Tharapiwattananon et al. 1996). When there are less air bubbles per volume leachate for the PFAS to get exposed to and therefore interact with and get removed by, the fact that a lower air flow rate results in a lower removal efficiency is a safe assumption.

For the CT 10 min, the increase of FF from 10 to 20 and 30 % did not result in a noticeable higher removal of \sum short chained, \sum long chained or \sum PFAS. For the three different FF (in the order 10, 20 and 30 %) the removal efficiency for short chained PFAS were 21, -3 and 9 % and for long chained PFAS 75, 75 and 67 %. This shows that the proportion between foam and effluent flow does not necessarily affect the removal efficiency. A higher percentage of foam was obtained by decreasing the effluent flow and therefore increasing the foam flow, but with a wetter foam/higher foam flow, the foam creation could have been affected. There was barely any visible foam creation for the high foam flows, contrarily to the lower foam flows where high foam creation was observed.

When keeping the FF at 5 % and increasing the CT from 10 to 20 and 30 min, average \sum PFAS removal efficiency increased from 33 % (CT 10 min), to 55 % (CT 20 min) and

59 % (CT 30 min). Like Kjellgren (2020) continuous experiments, a higher CT resulted in a higher RE. However, in this case, the airflow rate is three times bigger for the CT 30 min than for CT 10 min, so that could have affected the results. From the three first experiments CT 10-, 20- and 30-min FF 5 % it can be concluded that an increasing contact time results in a higher removal efficiency as long as the airflow is constant. Further investigations of the effect of the air flow rate should be done.

In general, to determine the optimal contact time, could a consideration of many parameters be done. Depending on what is preferable for the specific case, the optimal contact time could be seen as the lowest contact time before the removal efficiency stops to increase. The optimal contact time could also be seen as the lowest contact time that fulfils the requirements of the effluent concentrations or a contact time that fits well into the rest of the wastewater treatment chain. If for example the residence time of the other processes in the wastewater treatment chain is 30 min, it could complicate the process if another contact time would be used for this part of the chain.

The \sum_{11} PFAS had an effluent concentration between 630 and 1600 ng L⁻¹, which all is far from the upcoming requirement of 90 ng L⁻¹ for drinking water (Livsmedelsverket 2021b). Regarding the PFOS European Quality Standard limit of 0.65 ng L⁻¹ for inland surface water (European Environment Agency 2019) none of the effluent concentrations fulfilled that requirement. The effluent concentrations of PFOS varied between 6 and 108 ng L⁻¹ and with an average concentration of 30 ng L⁻¹. Using this method only for leachate to remove PFAS before making drinking water is therefore not an option and that could be for many more reasons than just the \sum_{11} PFAS concentrations. Regarding the concentration in the surface water, it will be a matter of dilution, the quantities and concentration of the effluent leachate in comparison to the recipient.

Four of the five experiments had the highest removal efficiency for the run with the highest inlet concentration and the lowest removal efficiency for the run with the lowest inlet concentration (Table 21 and Figure 20). This stands in contrast to Meng (2018), who found that a decreased PFOS inlet concentration resulted in a higher removal efficiency but is confirmed by the observation of Tharapiwattanon et al. (1996) that the removal efficiency increases with an increased concentration of surfactants. Keeping this in mind, a higher inlet concentration may result in a higher removal efficiency, but the R²-values in Figure 20 are not unambiguous. One explanation for a possible connection could be that the air bubbles will encounter more PFAS on their way up to the surface, and therefore more PFAS will adsorb to the bubbles and therefore get removed. If that theory is correct a higher inlet concentration may result in a higher removal efficiency until the bubbles get saturated, which might have been the case for Meng (2018). However further investigations need to be done before these speculations could be proven or not.

Since foam fractionation is proven to be more efficient for long chained PFAS the fraction of long chained PFAS were compared with the removal efficiency and the concentration (Table 21). This was done to ensure that it was not just because the runs with the highest removal efficiency of PFAS also had the highest fraction of long chained PFAS.

This was not the case since two experiments with the highest inlet concentration and removal efficiency did not have the highest fraction of long chained PFAS. A separate scatter plot (Figure 22) was made to see the relationship between the fraction of long

chained PFAS and removal efficiency both between the experiments and the different runs. The scatter plot did not show any relations but there are other parameters that affect the removal efficiency, so it does not prove that removal efficiency and fraction of long chained PFAS are uncorrelated.

In this study there are no clear connections between the iron inlet concentrations and the removal efficiency as shown in Figure 21b. The inlet concentration of iron was in between 4 and 5.5 mg L⁻¹ and the inlet concentrations or the difference might be too small to affect any noticeable difference in removal efficiency. The lack of connection stands in contrast to Lee (2017) who found that iron was a successful cationic activator. Since the difference in concentration between the experiments was small, other parameters (CT, FF, C_{PFAS} etc.) might have had a bigger impact. But if these parameters all would have been the same between the experiments there might have been a clearer connection between removal efficiency and iron inlet concentration.

There is no connection between conductivity and removal efficiency either (Figure 21a) since the two experiments with the highest conductivity have the highest and lowest removal efficiency.

If this method is proven to be effective only for long chained PFAS and not for short chained PFAS, an additional step for PFAS removal of short chained PFAS in the water treatment chain would be required.

5.2 Mass balance and Enrichment

Excluding the CT 10 min FF 5 % the fraction of the foam and effluent compared to the influent mass flow increased with a higher foam percentage from 53 (average for all FF 5 %) and 87 (FF 30 %) as can be seen in Figure 25. The biggest difference between different timesteps was in the CT 10 min FF 30 % where it differs six percentage points. The mass balance fraction should be approximately the same for different timesteps (Kjellgren 2020). The CT 10 min 5 % had at one timepoint mass balance fraction of more than two, i.e., twice as much PFAS going out of the column as going in. If there is an enrichment of PFAS in the column, the mass flows out of the column can be greater than influent mass flow at one timepoint (Meng 2018). But an increase from 0.71 at 15 min to 2.0 at 20 min is hard to explain in any other way than that it is a measurement error. And shown in Figure 24, the total mass flow out of the column for the whole experiment is higher than the inlet flow which is impossible and probably the results of the same measurement error as mentioned above. From Figure 24 the proportion of foam mass flow increase with a higher foam percentage as the effluent mass flow proportion decreases.

The enrichment in the underwater decreases with higher foam percentage (except for the short chained PFAS at 20 % FF) (Figure 24a) which is confirmed by Figure 24 of the mass balance that less PFAS are left in the column.

A higher enrichment for a dryer foam is also confirmed by (Tharapiwattananon et al. 1996). At all sample points there is a higher enrichment of short chained PFAS compared to the long chained PFAS in the underwater. But for some samples the enrichment factor is below one i.e., the influent concentration is higher and there is no enrichment at all. This is in contrariety to the enrichment in the foam where the long chained PFAS are enriched more than the short chained PFAS. This difference explains the higher removal

efficiency for long chained PFAS. The higher surface activity makes the long chained PFAS enrich in the foam (in the air bubbles) while the short chained PFAS enrich in the water as earlier described by Meng (2018). The CT 10 min FF 20 % has the highest enrichment in the foam but the lowest average removal efficiency.

5.3 Sources of Errors

As earlier described, the SPE is a method with many steps, and one possible reason for the first run (CT 10 min FF 5 %) to have outliers is man-made errors in the SPE method. Within the SPE there are multiple things that could have been done wrong, for example if two samples were mixed up with each other, if the spiking or mixing between chemicals was done incorrectly, if there was an earlier contamination in the equipment or if the integration was done incorrectly.

The foam flow was fluctuating, and the smaller foam fraction the more fluctuant foam flows due to a more unstable water level. The lowest measured foam flow for the 5 % experiments at one timestep was 1.7 % (sample ID 10:5-2F20), and the highest measured foam flow, 9.9 % (sample ID 30:5-1F60). When the aimed foam fraction differs between different runs the results will vary which will result in higher standard deviations. It can be hard to compare two experiments that were designed to have only one differing variable when the variable foam flow will result in at least two differing variables. To prevent this source of uncertainty a vacuum pump could be installed to control the foam flow. Using a higher foam fraction resulted in a more stable water level and therefore more stable foam flows.



Figure 26 Variation of raw water qualities from two different experiments.

Another source of error is the varying quality of inlet water. Figure 26 shows two different experiments from two different days where the raw water has different colours. To the left is the usual water colour (CT 10 min FF 5 %) and to the right is the CT 10 min FF 10 % experiment where the water was green. The various feed water could affect the experiment in many ways due to the different compositions. However, the composition of the leachate will vary depending on for example what processes on the landfills are ongoing while harvesting the water. One parameter that varies in the inlet water is the average Σ PFAS concentration, that was more than twice as high for the 30-min CT (3200 ng L⁻¹) as for the 10 min CT 10 % FF experiment (1400 ng L⁻¹).

The inlet concentration could vary for different runs within an experiment and that is the explanation for why the error bars are high for the influent and foam values. When the inlet water has a different inlet composition with regards to e.g. organic matter a fair

comparison between the removal efficiencies can be hard to make. Therefore, knowing the exact content of the water and how it affects the removal efficiency could be of importance. To avoid this problem all experiments could use the same inlet water. Now a 300 L plastic container was used for each run. To achieve that all 17 runs were ran with the same inlet water a container of approximately 5000 L would have to be used. And if the water were stored for weeks in that container the composition could possibly change within time.

If this method is to be upscaled and used, it could be problematic if it does not work for all raw water qualities or if the efficiency varies a lot depending on the raw water. If the relationship between the raw water quality and certain processes at the landfill could be stated, then at least the water treatment could be customized to fulfil the specific requirements for that water. The fact that there is a relation between removal efficiency and inlet concentration can make it problematic to compare different experiments if the inlet concentration varied a lot. The experiment with highest removal efficiency was the experiment with highest inlet concentration and if the inlet concentration would have been lower, there is no guarantee that the removal efficiency still would have been the highest.

The individual removal efficiency for each compound shown in the graphs with removal efficiency for different chain lengths and groups and graphs in Appendix 3 could be a little misleading for some compounds. If the compounds were detected at very low concentrations the removal efficiency between the runs could vary more and since a measurement error of a few ng L^{-1} is fatal if the inlet concentration has a magnitude of 10 ng L^{-1} but makes only a small difference if the concentration is of a magnitude of 200 ng L^{-1} .

6 Conclusions and Further Studies

Considering the overall results with an average removal efficiency of 46 % it is safe to state that aeration foam collection overall reduces PFAS levels in the leachate from Hovgården. There was an increasing concentration of PFAS or a very small reduction for some compounds and categories so there is no guarantee that aeration foam collection will reduce all the different PFAS levels or work as a single method for PFAS removal. Depending on the requirements of the water quality and PFAS levels an additional method could be necessary.

Considering the three first experiments, CT 10-, 20- and 30-min FF5 %, the study indicates that an increased contact time results in an increasing removal efficiency. But since there are other parameters that vary i.e., air flow rate further investigations are needed to state it beyond reasonable doubt. To decide the optimal contact time an evaluation of the different parameters and specific requirements could be advantageous. With only the results from this study it can be hard to answer how the collected amount of foam affects the reduction of PFAS. Comparing the CT 10 min FF 10, 20 and 30 %, the average removal was highest for the FF 10 % (53 %) but lowest for the FF 20 % (41 %) so no clear connections between foam fraction (i.e., collected amount of foam) and removal efficiency can be made from this.

This study can state that removal efficiency depends on the perfluoroalkyl carbon chain lengths where the long chained PFAS are removed to a higher extent than short chained

PFAS. Earlier conclusions that aeration foam collection is more effective for long chained PFAS than for short chained can therefore be confirmed. Removal efficiency do also depend on its functional group, were PFCA (all carboxylates) had a 11 % lower removal efficiency than PFSA (all sulfonates).

Whether the hypotheses were proven or not varies between the different hypotheses. A higher contact time resulted in an increased reduction and it worked better for long chained PFAS. There was a small enrichment of short chained PFAS in the underwater, but only for the CT 10 min FF 10 and 20 % (1.3 and 1.6). A higher foam fraction did not result in a higher removal efficiency.

What was supposed to be a study with only two different parameters, contact time and foam fraction ended up with eight different parameters, contact time, foam fraction, conductivity, inlet PFAS concentration, inlet concentration of iron, fraction of long chained PFAS and air flow ratio. This complicated the possibility to draw any conclusions from the results even if there are some clear indications of different connections.

Some further studies to be done could be to repeat this but with the same influent water and compare the results. A comparison with the different contact times, but the same air flow rate to see if the contact time or air flow rate affects the removal efficiency the most. A second foam fractionation i.e., a reflux or using collapsed foam as influent water to see what foam concentrations one can achieve would also be of interest.

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8 Appendix

8.1 Appendix 1: Σ_{20} PFAS

Σ_{20} PFAS from the Swedish food agency (Livsmedelsverket 2021b)

Appendix Table 1 Σ_{20} PFAS that is 20 PFAS chosen by the The European Commission to be kept under 100 ng L⁻¹

1	Perfluorbutansyra (PFBA)	11	Perfluorbutansulfonsyra (PFBS)
2	Perfluorpentansyra (PFPA)	12	Perfluorpentansulfonsyra (PFPS)
3	Perfluorhexansyra (PFHxA)	13	Perfluorhexansulfonsyra (PFHxS)
4	Perfluorheptansyra (PFHpA)	14	Perfluorheptansulfonsyra (PFHpS)
5	Perfluoroktansyra (PFOA)	15	Perfluoroktansulfonsyra (PFOS)
6	Perfluornonansyra (PFNA)	16	Perfluornonansulfonsyra (PFNS)
7	Perfluordekansyra (PFDA)	17	Perfluordekansulfonsyra (PFDS)
8	Perfluorundekansyra (PFUnDA)	18	Perfluorundekansulfonsyra
9	Perfluordodekansyra (PFDoDA)	19	Perfluordodekansulfonsyra
10	Perfluortridekansyra (PFTrDA)	20	Perfluortridekansulfonsyra

8.2 Appendix 2: Sample ID

Appendix 2 contains a table of all the sample IDs, what experiments they belong to, what date they were obtained, how long the experiments had been running before sampling and what their sample type.

Appendix Table 2 All the sample IDs and associated information

Sample ID	Experiment	Date	Time	Sample type
10:5-1i	CT 10 min FF 5 %	2021-02-22	0	Influent
10:5-2i	CT 10 min FF 5 %	2021-02-22	0	Influent
10:5-1Ef5	CT 10 min FF 5 %	2021-02-22	5	Effluent
10:5-2Ef5	CT 10 min FF 5 %	2021-02-22	5	Effluent
10:5-1Ef10	CT 10 min FF 5 %	2021-02-22	10	Effluent
10:5-2Ef10	CT 10 min FF 5 %	2021-02-22	10	Effluent
10:5-1Ef15	CT 10 min FF 5 %	2021-02-22	15	Effluent
10:5-2Ef15	CT 10 min FF 5 %	2021-02-22	15	Effluent
10:5-1Ef20	CT 10 min FF 5 %	2021-02-22	20	Effluent
10:5-2Ef20	CT 10 min FF 5 %	2021-02-22	20	Effluent
10:5-1F5	CT 10 min FF 5 %	2021-02-22	5	Foam
10:5-2F5	CT 10 min FF 5 %	2021-02-22	5	Foam
10:5-1F10	CT 10 min FF 5 %	2021-02-22	10	Foam
10:5-2F10	CT 10 min FF 5 %	2021-02-22	10	Foam
10:5-1F15	CT 10 min FF 5 %	2021-02-22	15	Foam
10:5-2F15	CT 10 min FF 5 %	2021-02-22	15	Foam
10:5-1F20	CT 10 min FF 5 %	2021-02-22	20	Foam
10:5-2F20	CT 10 min FF 5 %	2021-02-22	20	Foam
20:5-1ia	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-1ib	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-2ia	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-2ib	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-3ia	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-3ib	CT 20 min FF 5 %	2021-03-01	0	Influent
20:5-1Ef15	CT 20 min FF 5 %	2021-03-01	15	Effluent
20:5-2Ef15	CT 20 min FF 5 %	2021-03-01	15	Effluent
20:5-3Ef15	CT 20 min FF 5 %	2021-03-01	15	Effluent
20:5-1Ef45	CT 20 min FF 5 %	2021-03-01	45	Effluent
20:5-2Ef45	CT 20 min FF 5 %	2021-03-01	45	Effluent
20:5-3Ef45	CT 20 min FF 5 %	2021-03-01	45	Effluent
20:5-1F15	CT 20 min FF 5 %	2021-03-01	15	Foam
20:5-2F15	CT 20 min FF 5 %	2021-03-01	15	Foam
20:5-3F15	CT 20 min FF 5 %	2021-03-01	15	Foam
20:5-1F45	CT 20 min FF 5 %	2021-03-01	45	Foam
20:5-2F45	CT 20 min FF 5 %	2021-03-01	45	Foam
20:5-3F45	CT 20 min FF 5 %	2021-03-01	45	Foam
30:5-1ia	CT 30 min FF 5 %	2021-03-01	0	Influent
30:5-1ib	CT 30 min FF 5 %	2021-03-08	0	Influent
30:5-2ia	CT 30 min FF 5 %	2021-03-08	0	Influent
30:5-2ib	CT 30 min FF 5 %	2021-03-08	0	Influent
30:5-3ia	CT 30 min FF 5 %	2021-03-08	0	Influent

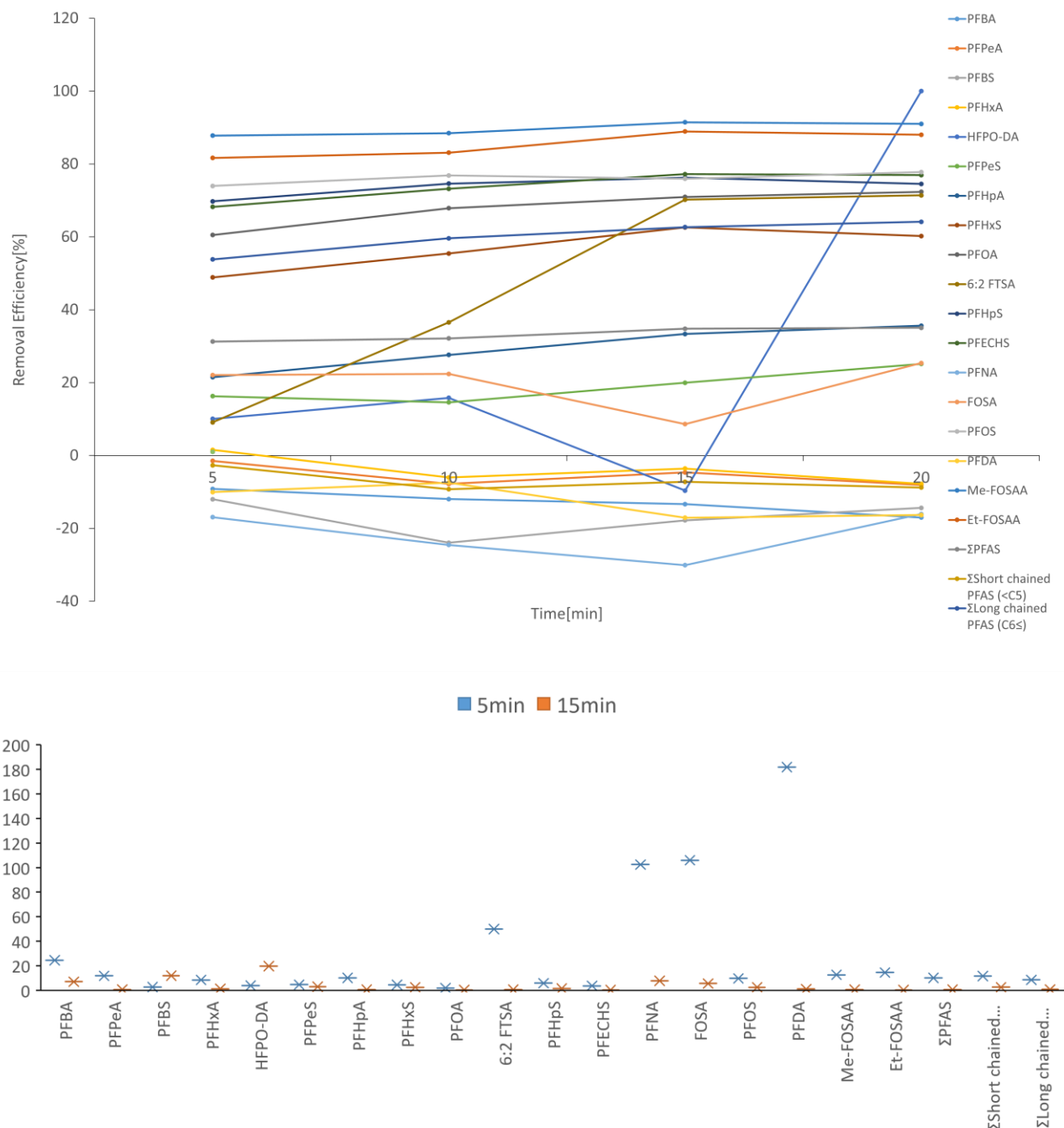
30:5-3ib	CT 30 min FF 5 %	2021-03-08	0	Influent
30:5-1Ef20	CT 30 min FF 5 %	2021-03-08	20	Effluent
30:5-2Ef20	CT 30 min FF 5 %	2021-03-08	20	Effluent
30:5-3Ef20	CT 30 min FF 5 %	2021-03-08	20	Effluent
30:5-1Ef60	CT 30 min FF 5 %	2021-03-08	60	Effluent
30:5-2Ef60	CT 30 min FF 5 %	2021-03-08	60	Effluent
30:5-3Ef60	CT 30 min FF 5 %	2021-03-08	60	Effluent
30:5-1F20	CT 30 min FF 5 %	2021-03-08	20	Foam
30:5-2F20	CT 30 min FF 5 %	2021-03-08	20	Foam
30:5-3F20	CT 30 min FF 5 %	2021-03-08	20	Foam
30:5-1F60	CT 30 min FF 5 %	2021-03-08	60	Foam
30:5-2F60	CT 30 min FF 5 %	2021-03-08	60	Foam
30:5-3F60	CT 30 min FF 5 %	2021-03-08	60	Foam
10:10-1i	CT 10 min FF 10 %	2021-04-12	0	Influent
10:10-2i	CT 10 min FF 10 %	2021-04-12	0	Influent
10:10-3i	CT 10 min FF 10 %	2021-04-12	0	Influent
10:10-1u	CT 10 min FF 10 %	2021-04-12	30	Underwater
10:10-2u	CT 10 min FF 10 %	2021-04-12	30	Underwater
10:10-3u	CT 10 min FF 10 %	2021-04-12	30	Underwater
10:10-1EF15	CT 10 min FF 10 %	2021-04-12	15	Effluent
10:10-2EF15	CT 10 min FF 10 %	2021-04-12	15	Effluent
10:10-3EF15	CT 10 min FF 10 %	2021-04-12	15	Effluent
10:10-1EF30	CT 10 min FF 10 %	2021-04-12	30	Effluent
10:10-2EF30	CT 10 min FF 10 %	2021-04-12	30	Effluent
10:10-3EF30	CT 10 min FF 10 %	2021-04-12	30	Effluent
10:10-1F15	CT 10 min FF 10 %	2021-04-12	15	Foam
10:10-2F15	CT 10 min FF 10 %	2021-04-12	15	Foam
10:10-3F15	CT 10 min FF 10 %	2021-04-12	15	Foam
10:10-1F30	CT 10 min FF 10 %	2021-04-12	30	Foam
10:10-2F30	CT 10 min FF 10 %	2021-04-12	30	Foam
10:10-3F30	CT 10 min FF 10 %	2021-04-12	30	Foam
10:20-1i	CT 10 min FF 20 %	2021-04-19	0	Influent
10:20-2i	CT 10 min FF 20 %	2021-04-19	0	Influent
10:20-3i	CT 10 min FF 20 %	2021-04-19	0	Influent
10:20-1u	CT 10 min FF 20 %	2021-04-19	30	Underwater
10:20-2u	CT 10 min FF 20 %	2021-04-19	30	Underwater
10:20-3u	CT 10 min FF 20 %	2021-04-19	30	Underwater
10:20-1EF15	CT 10 min FF 20 %	2021-04-19	15	Effluent
10:20-2EF15	CT 10 min FF 20 %	2021-04-19	15	Effluent
10:20-3EF15	CT 10 min FF 20 %	2021-04-19	15	Effluent
10:20-1EF30	CT 10 min FF 20 %	2021-04-19	30	Effluent
10:20-2EF30	CT 10 min FF 20 %	2021-04-19	30	Effluent
10:20-3EF30	CT 10 min FF 20 %	2021-04-19	30	Effluent
10:20-1F15	CT 10 min FF 20 %	2021-04-19	15	Foam
10:20-2F15	CT 10 min FF 20 %	2021-04-19	15	Foam
10:20-3F15	CT 10 min FF 20 %	2021-04-19	15	Foam
10:20-1F30	CT 10 min FF 20 %	2021-04-19	30	Foam
10:20-2F30	CT 10 min FF 20 %	2021-04-19	30	Foam

10:20-3F30	CT 10 min FF 20 %	2021-04-19	30	Foam
10:30-1i	CT 10 min FF 30 %	2021-04-19	0	Influent
10:30-2i	CT 10 min FF 30 %	2021-04-12	0	Influent
10:30-3i	CT 10 min FF 30 %	2021-04-12	0	Influent
10:30-1u	CT 10 min FF 30 %	2021-04-12	30	Underwater
10:30-2u	CT 10 min FF 30 %	2021-04-12	30	Underwater
10:30-3u	CT 10 min FF 30 %	2021-04-12	30	Underwater
10:30-1EF15	CT 10 min FF 30 %	2021-04-12	15	Effluent
10:30-2EF15	CT 10 min FF 30 %	2021-04-12	15	Effluent
10:30-3EF15	CT 10 min FF 30 %	2021-04-12	15	Effluent
10:30-1EF30	CT 10 min FF 30 %	2021-04-12	30	Effluent
10:30-2EF30	CT 10 min FF 30 %	2021-04-12	30	Effluent
10:30-3EF30	CT 10 min FF 30 %	2021-04-12	30	Effluent
10:30-1F15	CT 10 min FF 30 %	2021-04-12	15	Foam
10:30-2F15	CT 10 min FF 30 %	2021-04-12	15	Foam
10:30-3F15	CT 10 min FF 30 %	2021-04-12	15	Foam
10:30-1F30	CT 10 min FF 30 %	2021-04-12	30	Foam
10:30-2F30	CT 10 min FF 30 %	2021-04-12	30	Foam
10:30-3F30	CT 10 min FF 30 %	2021-04-12	30	Foam

8.3 Appendix 3: Results

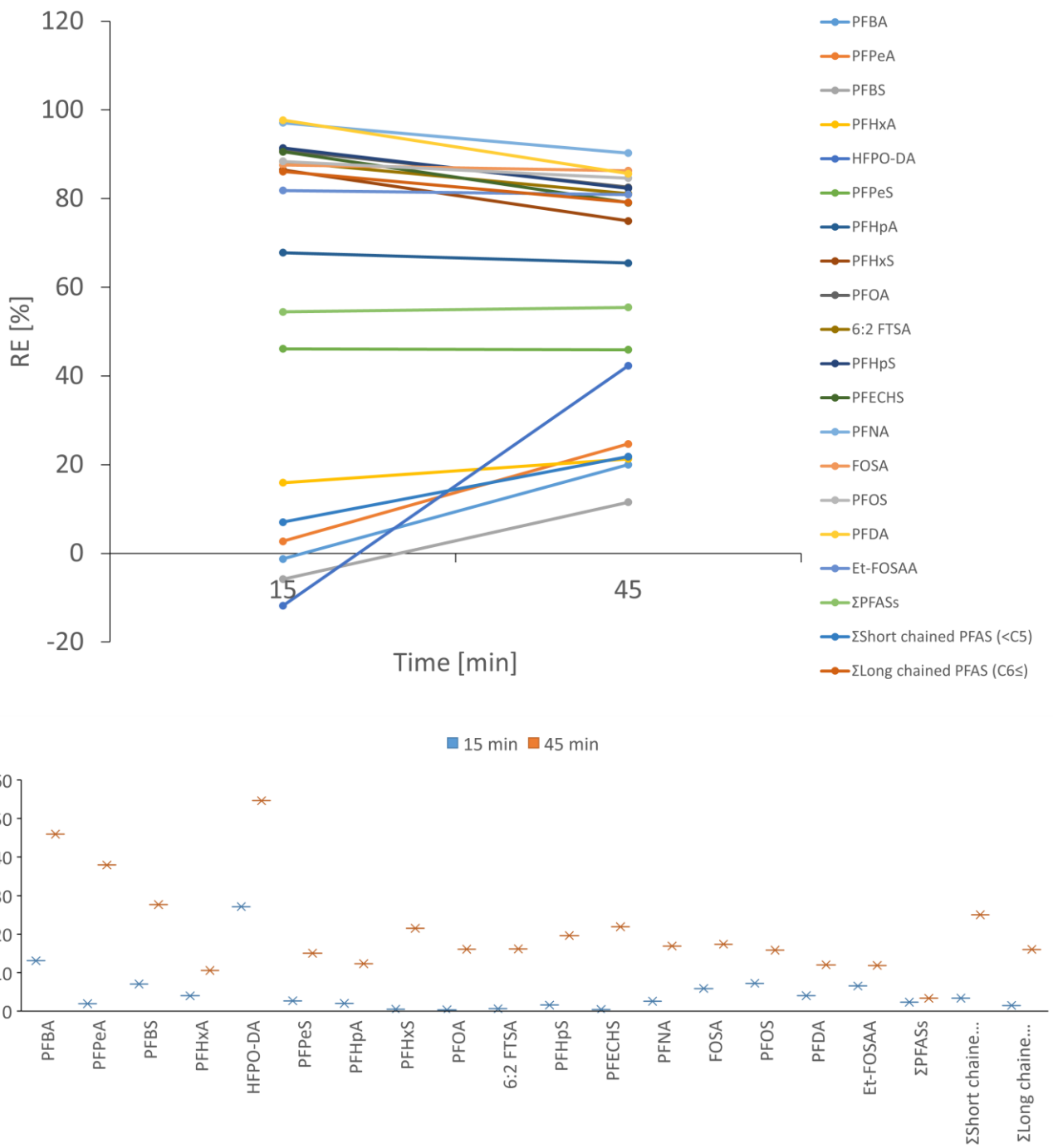
In Appendix 3 figures and tables of removal efficiency and standard deviation for each detected compound in each experiment is shown. For all experiments except for CT 10 min FF 5 % ALS results from general chemistry evaluation is found. In the ALS table percentage differences greater than 50 % are written in orange and percentage differences greater than -50 are written in red.

8.3.1 CT 10 min Foam Fraction 5 %



Appendix Figure 1 RE and STDAV of the RE for all compounds with CT 10 min & FF 5 %, colour and symbol coding as per insert legend. STDAV only plotted at two timesteps due to outliers at timestep 10 and 20 min.

8.3.2 CT 20 min Foam Fraction 5 %

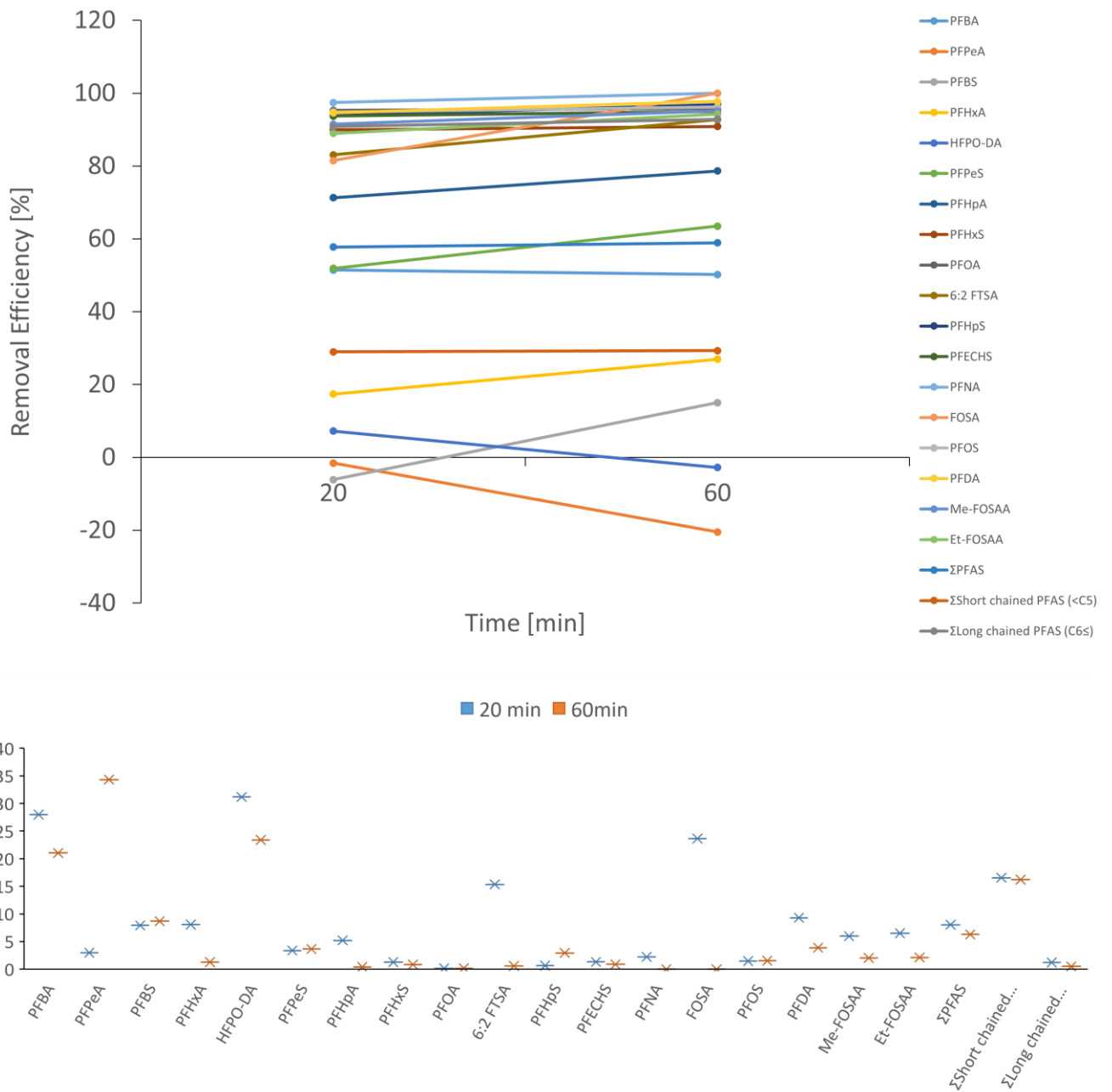


Appendix Figure 2 RE and STDAV of the RE for all compounds with CT 20 min & FF 5 %, colour and symbol coding as per insert legend.

Appendix Table 3 Results from the ALS-Scandinavia for the CT 20 min FF 5% experiment. The - represents a lack of value to be inserted

ELEMENT	SAMPLE	Influent	Effluent	Δ %
Sampling Date	2021-03-01			
U. uran	$\mu\text{g L}^{-1}$	43	42	-1.1
Ca. kalcium	mg L^{-1}	170	170	3.6
Mn. mangan	$\mu\text{g L}^{-1}$	490	520	5.0
Na. natrium	mg L^{-1}	730	740	1.2
K. kalium	mg L^{-1}	224	250	9.4
Fe. järn	mg L^{-1}	5.5	4.3	-22
Al. aluminium	$\mu\text{g L}^{-1}$	55	41	-24
Cu. koppar	$\mu\text{g L}^{-1}$	28	41	45
Mg. magnesium	mg L^{-1}	58	60	3.1
hårdhet	$^{\circ}\text{dH}$	37	38	3.6
DOC. löst organiskt kol	mg L^{-1}	39	39	-1.3
totalt fosfor som P	mg L^{-1}	0,098	0.095	-3.1
totalt kväve som N	mg L^{-1}	52	47	-10
nitrit	mg L^{-1}	0,72	0.74	3.5
nitrit som N	mg L^{-1}	0,22	0.23	3.2
COD-Mn	mg L^{-1}	19	20	1.5
NH4. ammonium	mg L^{-1}	63	62	-0.48
ammoniak- + ammoniumkväve	mg L^{-1}	49	48	-0.41
PO4. fosfat	mg L^{-1}	0,12	0.12	5.1
PO4-P. fosfat som P	mg L^{-1}	0,038	0.041	7.9
NO3. nitrat	mg L^{-1}	35	35	-0.29
NO3-N. nitrat som N	mg L^{-1}	7,9	7.9	-0.13
fluorid	mg L^{-1}	<0,50	<0.50	0
klorid	mg L^{-1}	870	870	0
SO4. sulfat	mg L^{-1}	210	210	-0.47
mättemperatur				
pH	$^{\circ}\text{C}$	21	22	5.2
turbiditet	FNU	49	46	-6.9
konduktivitet	mS m^{-1}	500	12	-98
pH		7,6	7,7	1.3
alkalinitet	$\text{mg HCO}_3^- \text{L}^{-1}$	1400	1200	-8.1
totalt organiskt kol (TOC)	mg L^{-1}	41	40	-3.7

8.3.3 CT 30 min Foam Fraction 5 %

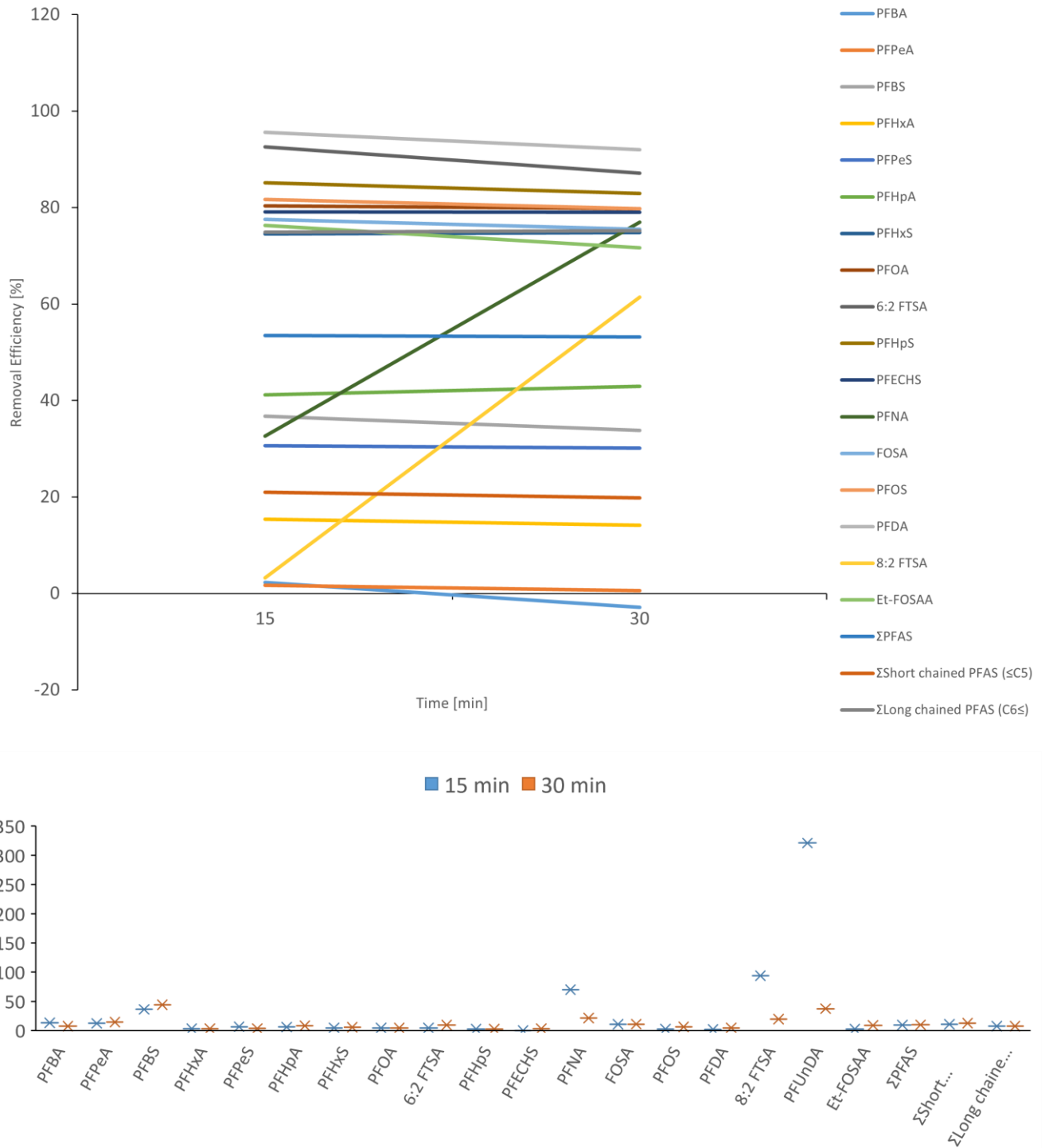


Appendix Figure 3 RE and STDAV of the RE for all compounds with CT 20 min & FF 5 %, colour and symbol coding as per insert legend.

Appendix Table 4 Results from the ALS-Scandinavia for the CT 30 min FF 5% experiment. The - represents a lack of value to be inserted

ELEMENT	SAMPLE	Influent	Effluent	Δ %
Sampling Date	2021-03-08			
U. uran	$\mu\text{g L}^{-1}$	34	34	-0.89
Ca. kalcium	mg L^{-1}	160	170	3.1
Mn. mangan	$\mu\text{g L}^{-1}$	480	450	-6.3
Na. natrium	mg L^{-1}	790	800	2.4
K. kalium	mg L^{-1}	260	270	2.3
Fe. järn	mg L^{-1}	4.5	3.7	-17
Al. aluminium	$\mu\text{g L}^{-1}$	35	23	-35
Cu. koppar	$\mu\text{g L}^{-1}$	26	27	0.76
Mg. magnesium	mg L^{-1}	63	65	2.2
hårdhet	$^{\circ}\text{dH}$	37	38	3.0
DOC. löst organiskt kol	mg L^{-1}	46	46	-0.43
totalt fosfor som P	mg L^{-1}	0.09	0.077	-14
totalt kväve som N	mg L^{-1}	53	46	-13
nitrit	mg L^{-1}	0.82	1.2	41
nitrit som N	mg L^{-1}	0.30	0.35	41
COD-Mn	mg L^{-1}	34	20	-42
NH ₄ . ammonium	mg L^{-1}	73	72	-1.1
ammoniak- + ammoniumkväve	mg L^{-1}	57	56	-1.1
PO ₄ . fosfat	mg L^{-1}	0.062	0.078	26
PO ₄ -P. fosfat som P	mg L^{-1}	0.020	0.025	25
NO ₃ . nitrat	mg L^{-1}	22	22	-0.45
NO ₃ -N. nitrat som N	mg L^{-1}	5.0	5.0	0
fluorid	mg L^{-1}	0.52	<0.50	-
klorid	mg L^{-1}	1100	1000	0
SO ₄ . sulfat	mg L^{-1}	220	210	-0.47
mättemperatur				
pH	$^{\circ}\text{C}$	19	19	-1.6
turbiditet	FNU	53	56	5.1
konduktivitet	mS m^{-1}	520	550	5.2
pH		7.7	7.8	1.3
alkalinitet	$\text{mg HCO}_3^- \text{L}^{-1}$	1500	1300	-11
totalt organiskt kol (TOC)	mg L^{-1}	46	46	0.65

8.3.4 CT 10 min Foam Fraction 10 %

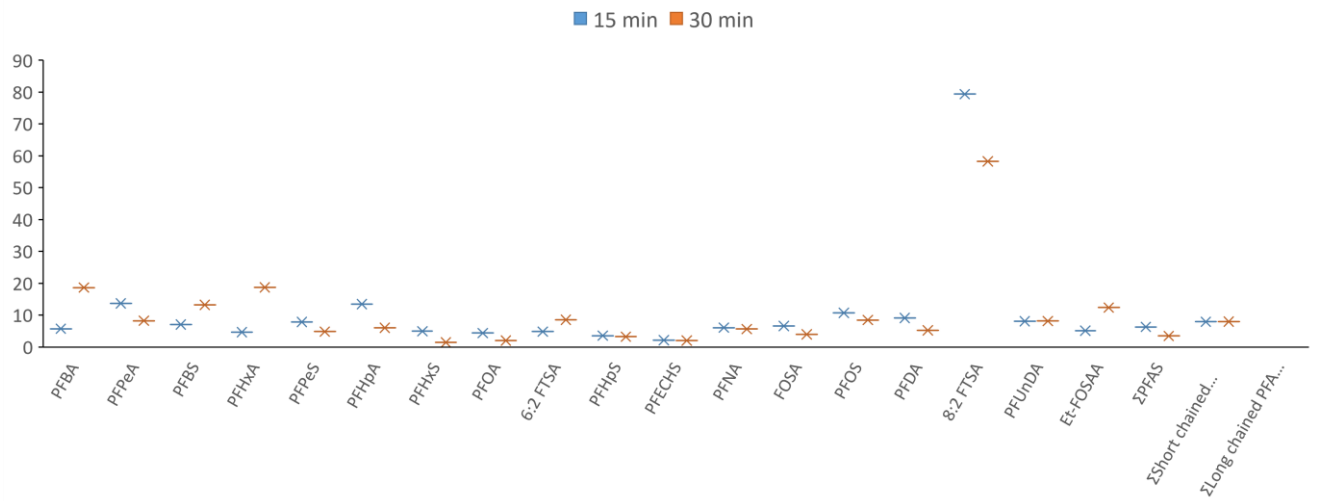
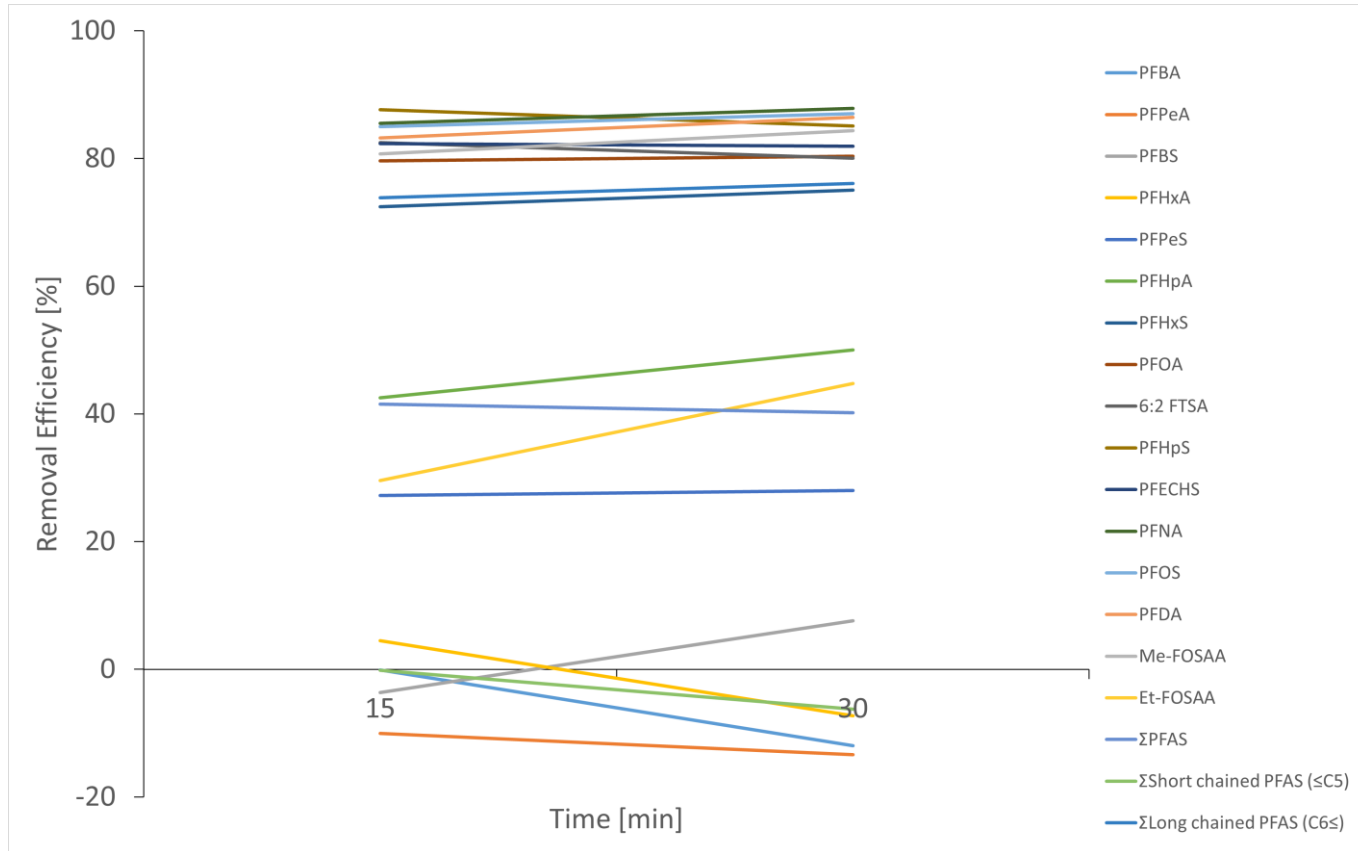


Appendix Figure 4 RE and STDAV of the RE for all compounds with CT 10 min & FF 10 %, colour and symbol coding as per insert legend.

Appendix Table 5 Results from the ALS-Scandinavia for the CT 10 min FF 10% experiment. The - represents a lack of value to be inserted

ELEMENT	SAMPLE	Influent	Effluent	Δ %
Sampling Date	2021-04-12			
U. uran	$\mu\text{g L}^{-1}$	36	36	0.28
Ca. kalcium	mg L^{-1}	130	130	0
Mn. mangan	$\mu\text{g L}^{-1}$	400	410	3.8
Na. natrium	mg L^{-1}	470	490	4.7
K. kalium	mg L^{-1}	160	170	3.0
Fe. järn	mg L^{-1}	4.0	3.9	-2.7
Al. aluminium	$\mu\text{g L}^{-1}$	260	190	-28
Cu. Koppar	$\mu\text{g L}^{-1}$	29	33	13
Mg. magnesium	mg L^{-1}	39	40	2.1
hårdhet	$^{\circ}\text{dH}$	27	28	1.1
DOC. löst organiskt kol	mg L^{-1}	49	43	-14
totalt fosfor som P	mg L^{-1}	1.1	1.1	-3.7
totalt kväve som N	mg L^{-1}	58	52	-11
nitrit	mg L^{-1}	0.16	0.81	420
nitrit som N	mg L^{-1}	0.047	0.25	430
COD-Mn	mg L^{-1}	45	39	-14
NH ₄ . ammonium	mg L^{-1}	83	80	-3.6
ammoniak- + ammoniumkväve	mg L^{-1}	65	62	-3.6
PO ₄ . fosfat	mg L^{-1}	1.1	1.2	17
PO ₄ -P. fosfat som P	mg L^{-1}	0.35	0.40	17
NO ₃ . nitrat	mg L^{-1}	0.58	13	2100
NO ₃ -N. nitrat som N	mg L^{-1}	0.13	2.9	2100
fluorid	mg L^{-1}	<0.50	<0.50	-
klorid	mg L^{-1}	610	640	3.9
SO ₄ . sulfat	mg L^{-1}	200	190	-0.51
mättemperatur				
pH	$^{\circ}\text{C}$	21	21	-0.48
turbiditet	FNU	68	66	-3.1
konduktivitet	mS m^{-1}	320	330	0.93
pH		7.7	7.8	1.3
alkalinitet	$\text{mg HCO}_3^- \text{L}^{-1}$	980	1100	6.9
totalt organiskt kol (TOC)	mg L^{-1}	51	47	-6.1

8.3.5 CT 10 min Foam Fraction 20 %

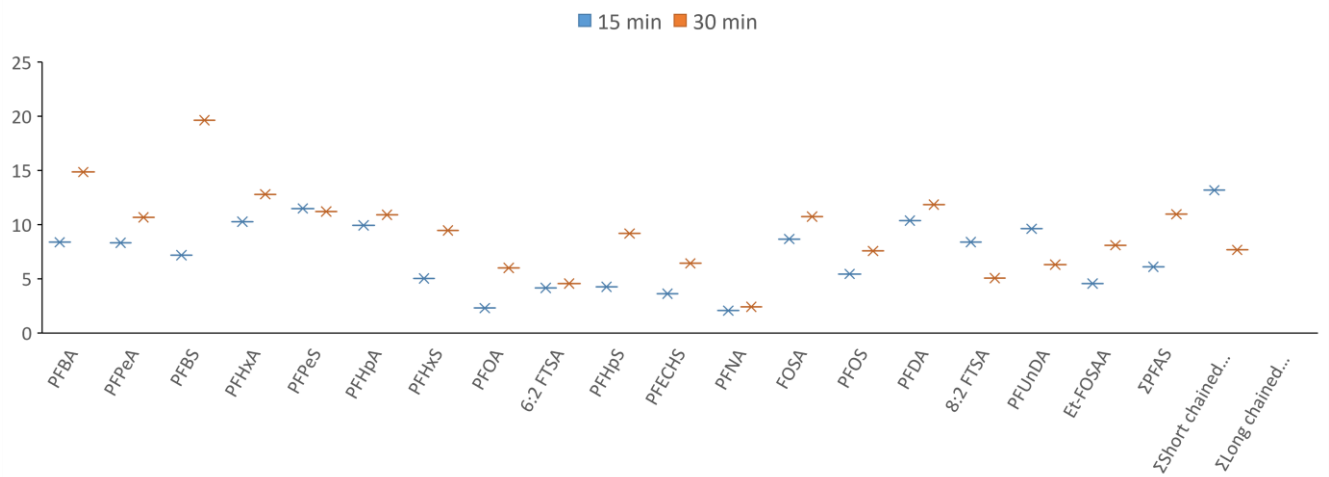
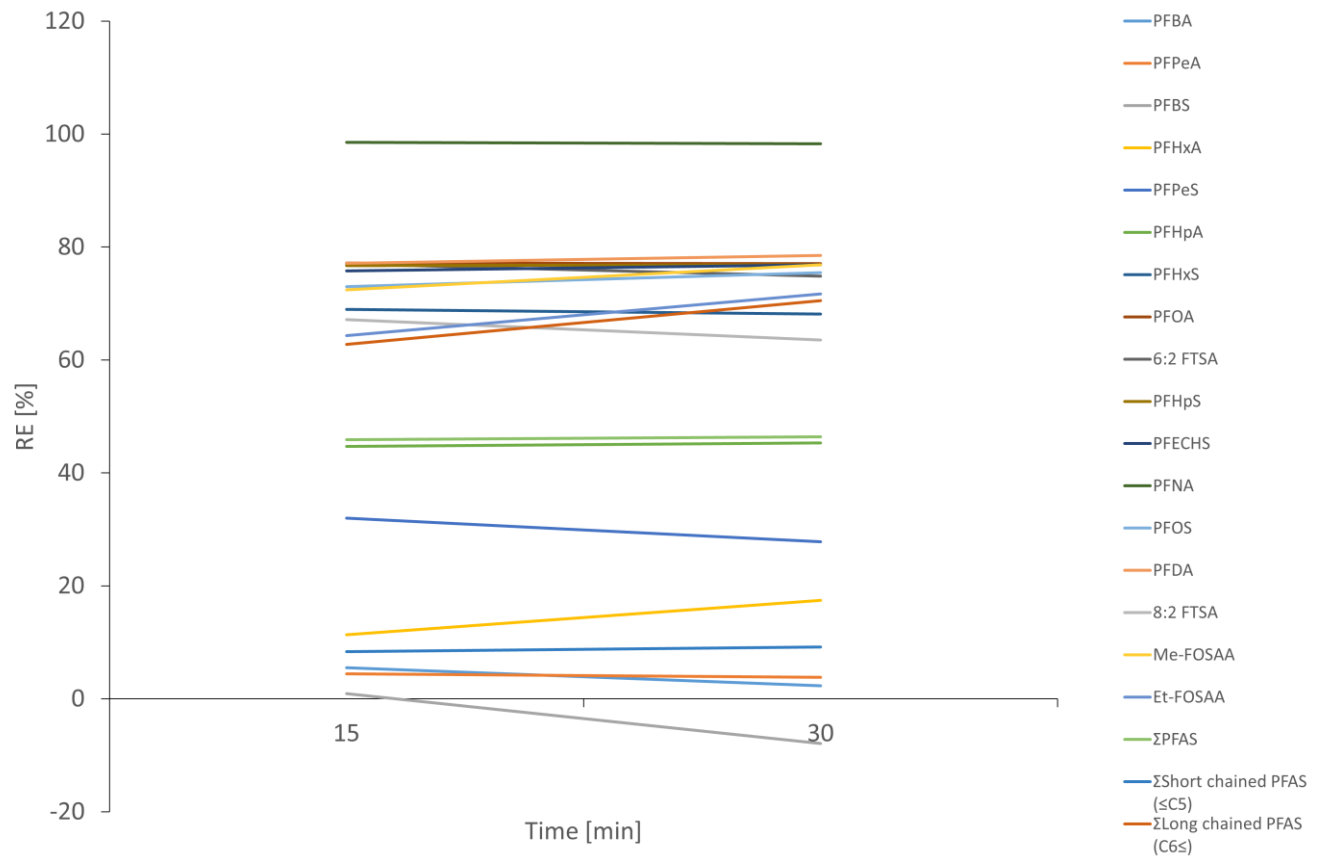


Appendix Figure 5 RE and STDAV of the RE for all compounds with CT 20 min & FF 20 %, colour and symbol coding as per insert legend.

Appendix Table 6 Results from the ALS-Scandinavia for the CT 10 min FF 20% experiment. The - represents a lack of value to be inserted

ELEMENT	SAMPLE	Influent	Effluent	Δ %
Sampling Date	2021-04-19			
U. uran	$\mu\text{g L}^{-1}$	31	33	4.5
Ca. kalcium	mg L^{-1}	150	160	2.0
Mn. mangan	$\mu\text{g L}^{-1}$	490	500	0.61
Na. natrium	mg L^{-1}	770	790	2.2
K. kalium	mg L^{-1}	270	270	1.9
Fe. järn	mg L^{-1}	5.0	5.1	0.80
Al. aluminium	$\mu\text{g L}^{-1}$	11	7.4	-30
Cu. koppar	$\mu\text{g L}^{-1}$	28	30	8.4
Mg. magnesium	mg L^{-1}	59	60	2.2
hårdhet	$^{\circ}\text{dH}$	35	36	2.0
DOC. löst organiskt kol	mg L^{-1}	43	44	1.4
totalt fosfor som P	mg L^{-1}	0.095	0.079	-17
totalt kväve som N	mg L^{-1}	50	47	-5.8
nitrit	mg L^{-1}	0.69	0.67	-3.6
nitrit som N	mg L^{-1}	0.21	0.20	-3.3
COD-Mn	mg L^{-1}	22	35	56
NH ₄ . ammonium	mg L^{-1}	72	72	0.56
ammoniak- + ammoniumkväve	mg L^{-1}	56	56	0.36
PO ₄ . fosfat	mg L^{-1}	0.055	<0.040	-
PO ₄ -P. fosfat som P	mg L^{-1}	0.018	<0.013	-
NO ₃ . nitrat	mg L^{-1}	14	15	5.7
NO ₃ -N. nitrat som N	mg L^{-1}	3.2	3.4	5.3
fluorid	mg L^{-1}	<0.50	<0.50	-
klorid	mg L^{-1}	990	990	0.61
SO ₄ . sulfat	mg L^{-1}	160	160	-1.2
mättemperatur				
pH	$^{\circ}\text{C}$	21	20	-1.9
turbiditet	FNU	48	53	11
konduktivitet	mS m^{-1}	520	510	-1.2
pH		7.9	8	1.3
alkalinitet	$\text{mg HCO}_3^- \text{L}^{-1}$	1400	1400	0
totalt organiskt kol (TOC)	mg L^{-1}	45	45	0

8.3.6 CT 10 min Foam Fraction 30 %



Appendix Figure 6 RE and STDAV of the RE for all compounds with CT 10 min & FF 30 %, colour and symbol coding as per insert legend.

Appendix Table 7 Results from the ALS-Scandinavia for the CT 10 min FF 30% experiment. The - represents a lack of value to be inserted

ELEMENT	SAMPLE	Influent	Effluent	Δ %
Sampling Date	2021-04-12			
U. uran	$\mu\text{g L}^{-1}$	26	26	-1.2
Ca. kalcium	mg L^{-1}	160	160	0.64
Mn. mangan	$\mu\text{g L}^{-1}$	440	430	-2.3
Na. natrium	mg L^{-1}	640	650	1.1
K. kalium	mg L^{-1}	220	220	0.9
Fe. järn	mg L^{-1}	5.1	4.5	-11
Al. aluminium	$\mu\text{g L}^{-1}$	63	59	-7.3
Cu. koppar	$\mu\text{g L}^{-1}$	26	28	8.5
Mg. magnesium	mg L^{-1}	50	50	0.80
hårdhet	$^{\circ}\text{dH}$	33	34	0.90
DOC. löst organiskt kol	mg L^{-1}	36	36	0.56
totalt fosfor som P	mg L^{-1}	0.15	0.25	62
totalt kväve som N	mg L^{-1}	40	40	-1.0
nitrit	mg L^{-1}	0.65	0.76	18
nitrit som N	mg L^{-1}	0.20	0.23	18
COD-Mn	mg L^{-1}	38	46	21
NH4. ammonium	mg L^{-1}	62	62	-0.5
ammoniak- + ammoniumkväve	mg L^{-1}	48	48	-0.41
PO4. fosfat	mg L^{-1}	0.15	0.28	81
PO4-P. fosfat som P	mg L^{-1}	0.049	0.090	84
NO3. nitrat	mg L^{-1}	16	17	1.2
NO3-N. nitrat som N	mg L^{-1}	3.7	3.7	1.1
fluorid	mg L^{-1}	<0.50	<0.50	-
klorid	mg L^{-1}	820	830	2.0
SO4. sulfat	mg L^{-1}	200	200	-1.5
mättemperatur				
pH	$^{\circ}\text{C}$	21	21	0.50
turbiditet	FNU	56	41	-26
konduktivitet	mS m^{-1}	400	430	5.5
pH		7.7	8	3.9
alkalinitet	$\text{mg HCO}_3^- \text{L}^{-1}$	1300	1300	-1.6
totalt organiskt kol (TOC)	mg L^{-1}	37	36	-1.1