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An investigation into the use of spent activated carbon in the filtration of PFAS contaminated leachate

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are synthetic organic chemicals that have hazardous effects on human health and the environment. Several pollution hotspots have resulted from the use of PFAS-containing fire-fighting foams. A significant concern is that the contamination can spread from contaminated sites to nearby water bodies and drinking water. Activated carbon (AC) is one of the most common binding materials (sorbents) for PFAS-contaminated soil and water.

This study investigated the hypothesis that many spent AC filters are sent to landfills while potentially having unused sorption sites for organic contaminants such as PFAS compounds. The AC types to be tested included pellets from air- and biogas purification, granulated AC, and anthracite from wastewater treatment. All samples were first analysed for their contaminant content, obtained from their previous applications, and then batch leaching tests were conducted to determine the leachability of these contaminants into the water phase. Then the material´s ability to sorb PFAS was investigated, also trough batch adsorption tests, with PFAS-contaminated leachate containing varying concentrations of the sorbents.

The PFAS-contaminated leachate was dominated by the short-chained PFAS perfluoropentanoic acid (PFPeA) and perfluorobutanoic acid (PFBA), among the 33 quantified PFAS compounds. From the various sorbents, the spent AC sample from the air purification company CLAIRS provided the highest potential for reuse as a PFAS sorbent with an overall 97% efficiency using 4 g of AC. Biogas purification and wastewater treatment sorbents showed less efficiency, due to natural organic matter such as humus and BTEX on the sorption sites. In conclusion, the air purification sorbent showed a greater adsorption affinity to PFAS, however, there is still uncertainties around the environmental impacts of the substantial amount of sulphur leakage from these spent AC types. Therefore, further testing needs to be performed on other air filtration samples, not only on the sulphur leaching. It must also be tested in a large-scale field test.

Sammendrag

Per- og polyfluoralkylstoffer (PFAS) er syntetiske organiske kjemikalier som har skadelige effekter på menneskers helse og miljø. Flere forurensa plasser kommer fra tidligere bruk av PFAS-holdig brannslukkningsskum. En betydelig bekymring er, at forurensningen kan spre seg fra forurensede områder til nærliggende vannforekomster og drikkevann. Aktivt karbon (AC) er et av de vanligste bindematerialene (sorbentene) for PFAS-forurenset jord og vann.

Dette studiet undersøker potensialet for brukt AC fra tre forskjellige kilder, om de kan gjenbrukes som en PFAS-sorbent. AC-sorbentene som skal testes er pellets fra luft- og biogassrensing, granulert AC og antrasitt fra avløpsvannbehandling.
Alle AC-prøvene blei først analysert for å finne ut av hvilke forurensinger de hadde fra tidligere bruk. Siden blei en batch-lekking-test uført, for å se hvor lett de hadde for å slippe forurensingene sine ut i vann. Så ble prøvene testet for deres evne til å sorbere PFAS, også ved batch-adsorpsjons test, med varierende mengder av sorbenter.

Det PFAS-kontaminerte sigevannet var dominert av de kortkjedete PFAS komponentene perfluorpentansyre (PFPeA) og perfluorbutansyre (PFBA), blant de 33 kvantifiserte PFAS-ene i sigevannet. Av de ulike sorbentene ga den brukte AC fra luftrenseselskapet CLAIRS det høyeste potensialet for gjenbruk som en PFAS-sorbent, med en samlet effektivitet på 97 % ved bruk av 4 g AC. Absorbenter for biogassrensing og avløpsvannsbehandling viste mindre effektivitet på grunn av DOC og BTEX i sorpsjonsstedene. Avslutningsvis viste luftrensnings sorbenten en større adsorpsjonsevne for PFAS. Imidlertid er det en betydelig mengde svovellekkasje fra AC'en. Ytterligere testing må utføres på andre luftfiltreringsprøver, med søkelys på PFAS-adsorpsjon og svovelutlekking. Det må også testes ytterligere i felt.

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Terminology

AC	Activated carbon
AFFF	Aqueous film-forming foam
DOC:	Dissolved organic carbon
DM:	Dry matter
FTS	6:2 Fluorotelomer sulfonate
GAC	Granulated active carbon
HDPE	High density polyethylene
K_d	The soil-water partition coefficient
K_{OC}	The organic carbon-water partition coefficient
KKV:	Kristiansand Kommune vannverk
L/S	Liquid / solid
LOQ:	Limit of quantification
MINA:	Faculty of Environmental Science and Natural Resource Management
NMBU:	The Norwegian University of Life Science
NOM:	Natural organic material
NRV:	Nedre Romerike vannverk
PAH:	Polycyclic aromatic hydrocarbons
PBT:	Persistent, bioaccumulative and toxic
PCB:	Polychlorinated biphenyls
PFAS	Per- and polyfluoroalkyl substances
PFBS	Perfluorobutane sulfonate
PFBA	Perfluorobutanoic acid
PFHpA	Perfluoroheptanoic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctyl sulfonate
PFPeA	Perfluoropentanoic acid
PFPeS	Perfluoropentane sulfonate
Reach	Registration, Evaluation, Authorisation, and Restriction of Chemicals
SAFF	Surface Active Foam Fraction

TOC: Total organic carbon

TMF: The magical factory

1   Introduction

Per- and polyfluoroalkyl substances (PFAS) are a sizeable group of artificial carbon-fluorinated substances with unique physicochemical properties such as water, stain, fire, and grease repellent properties. PFAS has since the 1950s been used in different products, such as fire-fighting foams, food contact materials, Teflon products, cleaning products, silicon, and textiles (Haug et al., 2020; Jian et al., 2018; Knutsen et al., 2019; Wang et al., 2017).

Even though PFAS has been known since the 50s, there hasn't been given any considerable attention to its environmental problems before the early 2000s. Since then, PFAS have been discovered ubiquitously in the environment, from the oceans to the biota and even in human blood (Sinclair et al., 2020). Figure 1 shows a timeline of PFAS history.

Anthropogenic activities have resulted in a large scale PFAS contaminations, with over 6,330 compounds in the environment (Miljødirektoratet, 2021b) and these can potentially cause adverse effects, both to the biota and to human health (Haug et al., 2020; Knutsen et al., 2019; Krafft & Riess, 2015; Masoner et al., 2020). Thus, remediation of pollution hot spots, like landfills, is urgent to protect the surrounding environments from contaminations. While the remediation of PFAS-contaminated water and soil is highly established (Bolan et al., 2021; Hale et al., 2017), the treatment of PFAS in landfill leachate is still limited.

Hence, alternative methods, that are both cost-effective and sustainable for site remediation, must be established.
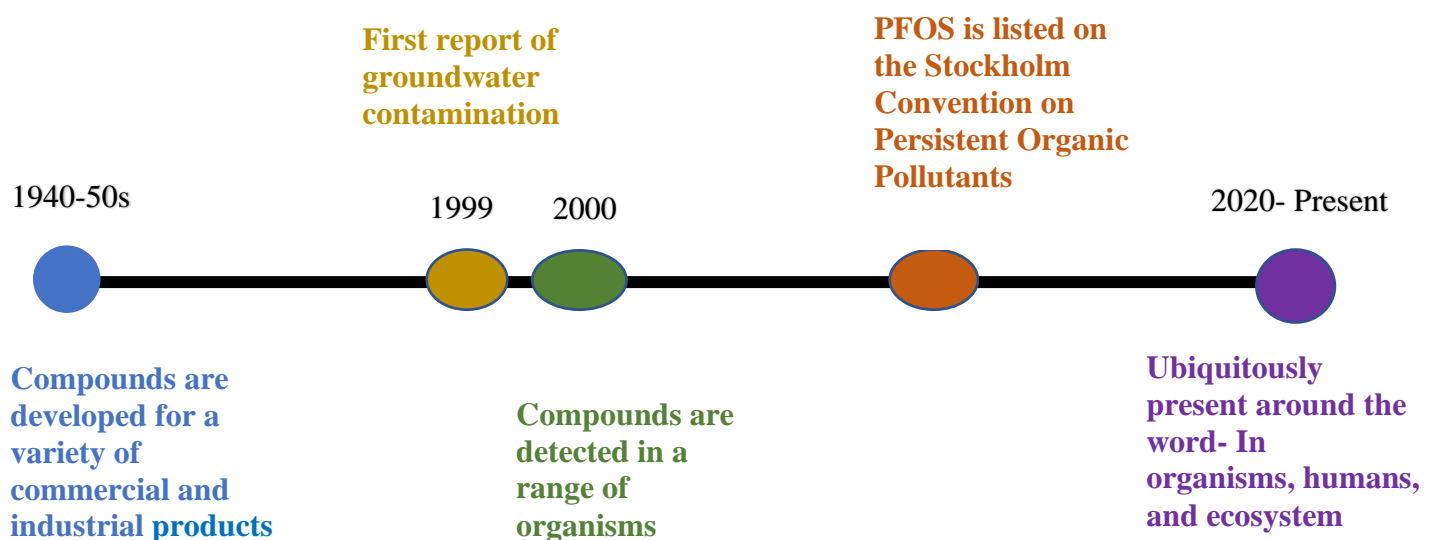
Figure 1: Overview over the PFAS timeline, Inspired by (Sinclair et al., 2020)

1.1   PFAS

PFAS is defined as a substance that contains alkyl parts consisting of fully (per-) or partly (poly-) fluorinated carbon chains, where fluorine atoms have replaced the hydrogen atoms (Miljødirektoratet, 2021b; Sinclair et al., 2020; Wang et al., 2017).

The chains are attached to functional groups, typically carboxylic or sulfonic acids (see Table1) (American Water Works Association, 2019; Buck et al., 2011).

PFAS contains both a hydrophilic (attracted to water)- and hydrophobic (water-repellent) parts. The hydrophilic part is located in the functional "head" groups, while the hydrophobic is located in the fluorinated "tail" (figure 2) (Hale, n.d; Rahman et al., 2014).

Fluorinated tail
(**Hydrophobic**)

Functional group
(**Hydrophilic**)
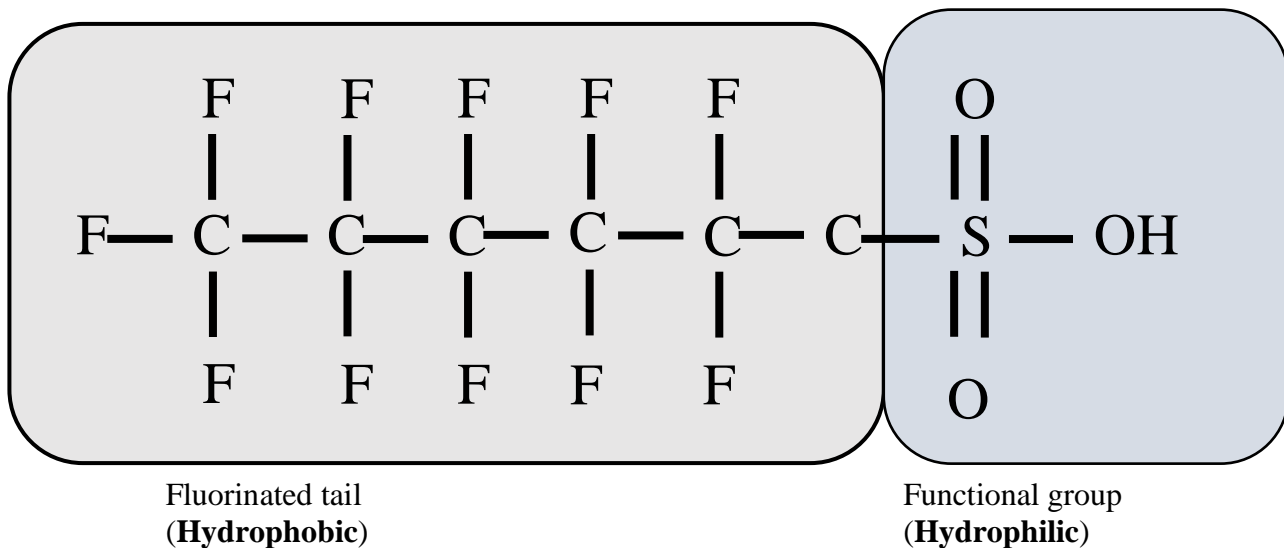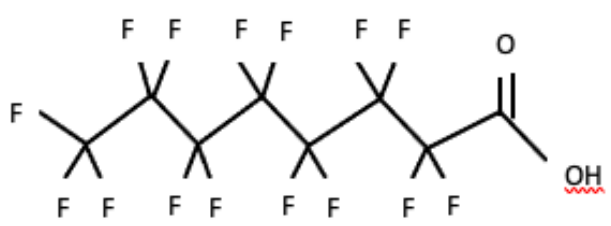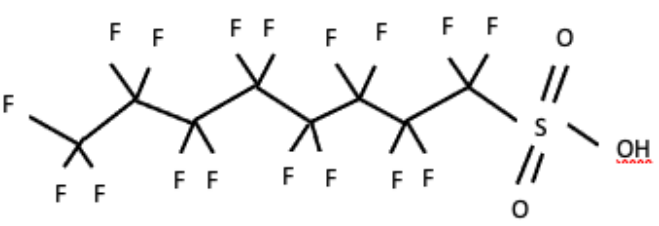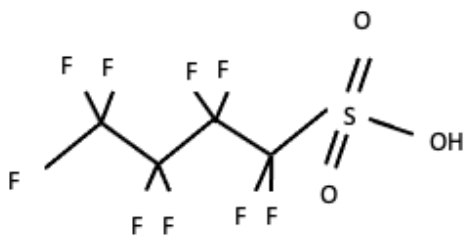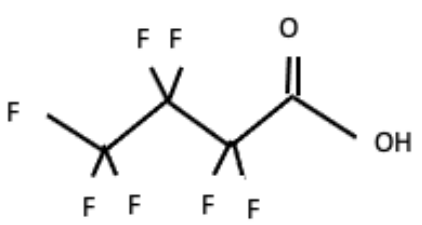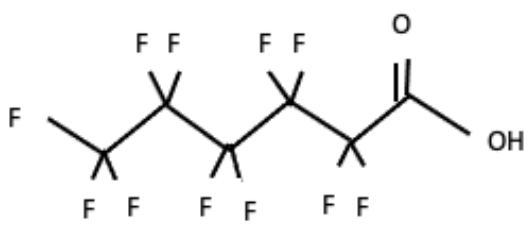
Figure 2 Illustration of the PFAS compound PFOS, with its hydrophobic "tail" and hydrophilic "head"

Table 1: Names, abbreviations, numbers of carbon and structure for five of the most common PFAS compounds. Perfluorooctanoic acid (PFOA), perfluorooctane sulfonate (PFOS), perfluorobutane sulfonate (PFBS), perfluorobutanoic acid (PFBA) and perfluorohexanoic acid (PFHxA) Inspired from (Zhang et al., 2019).

PFAS name	Abbreviation	Structure
Perfluorooctanoic acid	PFOA (C8)	
Perfluorooctane sulfonate	PFOS (C8)	
Perfluorobutane sulfonate	PFBS (C4)	
Perfluorobutanoic acid	PFBA (C4)	
Perfluorohexanoic acid	PFHxA (C6)	

The carbon-fluorine (C-F) bonds are among the strongest bonds in organic chemistry, particularly due to fluorine's strong electron drawing effect, making PFAS a very stable and persistent substance (Chohan et al., 2021; Giesy & Kannan, 2001; Zhao et al., 2016). The strong C-F bond makes PFAS largely resistant to biotic transformation (i.e., it is broken down very slowly in nature) and is often called a "Forever Chemical." PFAS is very mobile and can move through the environment via several routes, such as landfill leachates, oceans, streams, and groundwater. In addition to being very mobile, several PFASs are classified as persistent, bioaccumulative and toxic (PBT) (European Comission, 2020; Sinclair et al., 2020).
The physicochemical properties, such as the length of the carbon chain, degree of fluorination and the functional groups of PFAS, decide its fate in the environment, whether it is mobile or bioaccumulative (Knutsen et al., 2019; Sörengård et al., 2020).

Some, of the adverse effects PFAS has on humans, are documented, there is however a need for further research. Earlier studies of human exposure to two particular PFAS compounds , (PFOA and PFOS) showed that it may lead to increased cholesterol levels, immunotoxicity, kidney and testicular cancer, thyroid disease, inhibiting the development of antibodies after vaccination, increase the blood pressure and preeclampsia in pregnant woman (ATSDR, 2020; European Comission, 2020; Haug et al., 2020).

High exposures to PFAS can have other developmental disorders such as liver damage and disturbances in fat metabolism in the body. Several PFASs can inhibit fetal development (Haug et al., 2020). The half-life of these compounds in the human body depends on whether it is long or short chain, and can therefore be between 4 days or 5 years (American Water Works Association, 2019).
Due to the toxicity of PFAS, many regulatory bodies have imposed stricter regulations concerning its use and mitigation guidelines (Sinclair et al., 2020).

1.2 PFAS restrictions

As more and more information about this "new" contaminant and its bioaccumulative, persistent and highly mobile properties has become known, the restrictions and laws on production and use of these compounds have become increasingly stricter.

Over the years the Norwegian Environmental Protection Agency have included different PFAS compounds in the List of Priority Compounds, as a measure to reduce and eventually stop the emission of the contaminant. This list includes compounds that "*pose a serious threat to health and the environment*" (Miljødirektoratet, 2020a; Miljødirektoratet, 2021a). Perfluorooctane sulfonate (PFOS) was added in 2002 as the first PFAS compound, followed by perfluorooctanoic acid (PFOA) in 2007, perfluorohexane sulfonate (PFHxS) in 2017, perfluorobutane sulfonate (PFBS) in 2019 and perfluorohexanoic acid (PFHxA) in 2020.

So far, PFOS is the only PFAS substance regulated substance through European and global legalisation. PFOS and related compounds have since 2009 been listed under Appendix B of the Stockholm Convention for Persistent Organic Pollutant (POPs) and listed in Annex I of the POPS Regulation (EU 2020/784) (European Comission, 2020; OECD, 2015; OECD, u.k.). PFOA was considered to be listed in 2017 (Chohan et al., 2021)

Norway adopted a national regulation banning PFOS and PFOS-related compounds in fire-fighting foams, impregnating agents and textiles on the 21st March 2007, complying with the EU directive on PFOS from 2006 (2006/122/ECOF) (Regjeringen, 2010).

Other countries have also introduced strict regulations and guidelines to either phaseout or limit the use of PFAS (Sinclair et al., 2020). Additionally, the international ski federation (FIS) has prohibited any product containing PFAS at all FIS events from season 2021/2022 onwards, complying with the EU regulation (FIS, 2021).

With all of the regulations on long-chain PFASs, a variety of non-regulated compounds with shorter chain length have been developed and gradually used as a replacement (Brendel et al., 2018; OECD, 2013)

Perfluorobutane sulfonic acid (PFBS) is an example of a four-carbon PFAS compound developed to replace PFOS (EPA, 2018). However, the introduction of short-chain PFAS

compounds has led to an increased PFAS concentration in the environment (Ateia et al., 2019).

The EU drinking water directive has adopted limit values for all PFAS compounds. The sum of PFASs in drinking water should not exceed 0.5 µg/L. Each of the compounds should not occur at levels above 0.1 µg/L (1000 ng/L) (Miljødirektoratet, 2021b). Table 2 shows an overview of the current quality standards in Norway for PFOS and PFOA in waters. The standards are divided into five classes, background, good, moderate, bad, and very bad. The classes represent an expected degree of damage to the organism community. Good is no toxic effects, moderate is chronic effects with long-term exposure, bad is acute toxic effects with short-term exposure, and very bad is extensive toxic effects (Miljødirektoratet, 2020b).

The criteria used to determine classification limits are based on internationally established standards for environmental quality and risk assessment of chemical exposure in the EU, while the limit values have been derived from the Technical Guidance Document for Deriving Environmental Quality Standards (Miljødirektoratet, 2020b).

Table 2. *The Norwegian quality standards for PFOS and PFOA in both fresh water and costal water. Only Class II and Class III values are defined (Norwegian Environmental Agency, 2020)*

	Class	Class I	Class II	Class III	Class IV	Class V
	Classification	Background	Good	Moderate	Bad	Very bad
PFOS	Fresh water (µg/L) Costal water (µg/L)	*ND*	0-0000.65 0-0.13	0.00065-36 0.00013-7.2	*ND*	*ND*
PFOA	Fresh water (µg//L) Costal water (µg//L)	*ND*	0-9.1 0-9.1	*ND*	*ND*	*ND*

ND= not defined

1.3   Fate of PFAS in the environment.

PFAS can be divided into two groups, "long-chain" and "short-chain", which have different properties and fates in the environment (Zhao et al., 2016).

"Long-chain" PFAS is a term used if the carbon chain length contains >6 carbons. Long-chained PFASs are known to be very stable, and were found to be more predominant in sediment samples from Norwegian landfills than the short-chained ones, due to their ability to bioaccumulate (Knutsen et al., 2019). This ability increases gradually with the length of the carbon-chain (Krafft & Riess, 2015).

The most researched and well-known long-chain PFAS compounds are PFOS and PFOA. Of these two, PFOS has a slightly higher ability to bioaccumulate than PFOA, due to the different functional groups (Ahrens & Bundschuh, 2014). It is said that sulfonated fluorocarbons sorb stronger than carboxylate ones (Zareitalabad et al., 2013). Sulfonates have a larger molecular volume, making them more hydrophobic (Zhang et al., 2019). PFAS compounds are often present in anionic form but can also exist in neutral and cationic states. The state is pH-dependent (Buck et al., 2011).

"Short-chain" PFAS is a term used if the carbon chain contains <6 carbons. They are less bioaccumulative and are more dominant in the aquatic environment and landfill leachates. Some short-chain PFAS compounds, that can be found in landfill leachates, are PFBS and perfluorobutanoic acid (PFBA) (American Water Works Association, 2019; Knutsen et al., 2019). Additionally, short-chain variants tend to be more water-soluble and adsorb less strongly to particles than long chained variants, thus being more persistent and likely to remain in the environment for decades or centuries. (Brendel et al., 2018; Cousuns et al., 2016; Kjølholt et al., 2015; Krafft & Riess, 2015).

Although long-chain PFAS compounds are known to be very stable and resistant to biotic transformation, upon breakdown, they will degrade into shorter chain forms. (Sinclair et al., 2020). Thus, PFAS will always be present in the environment.

1.4   Discharge of PFAS

Since the production of PFAS in the early 50s, anthropogenic activities have resulted in a large-scale contamination worldwide. From typical point emission sources, PFAS compounds have been found in remote locations, as in artic wildlife, far from human settlements (Lindstrom et al., 2011). The ability to move over long distances and contaminate new areas far from the original emission point, makes the PFAS emission picture complicated. Even though it is still very unclear how the environment and humans have been exposed to PFAS, some major pathways have been identified, and are shown in figure 3.
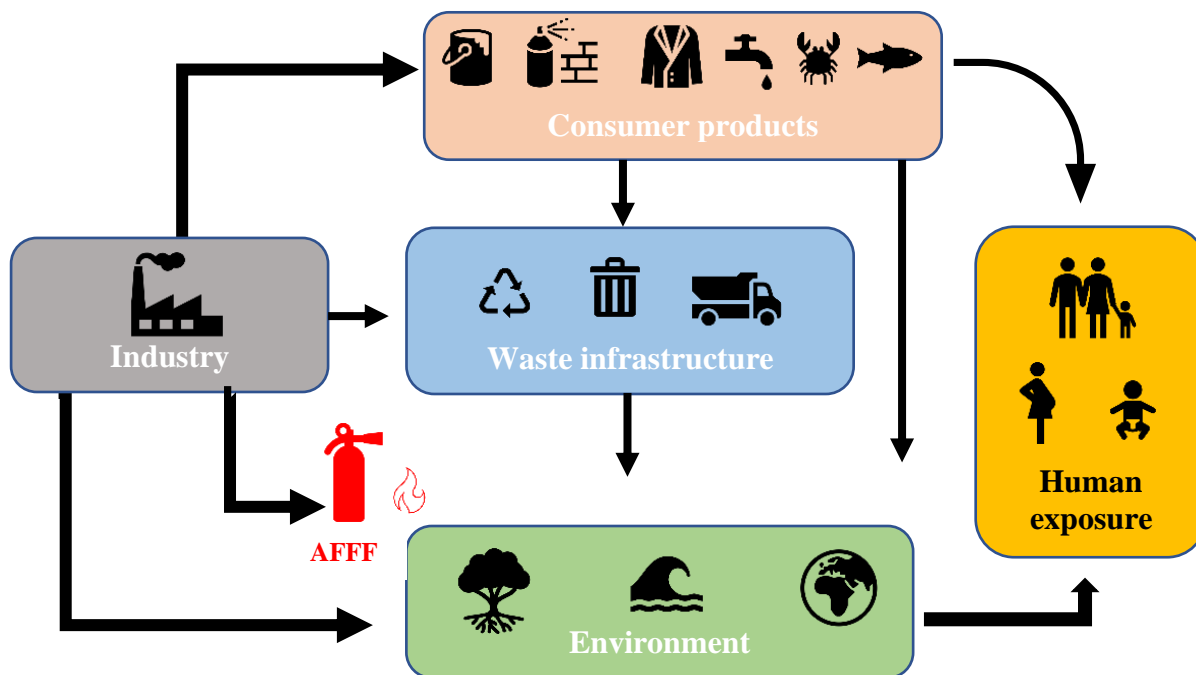
*Figur 3: Overview over the major exposure pathways of PFAS, inspired from (Sunderland et al., 2019).   Humans are primarily exposed to PFAS trough ingestion of contaminated drinking water and seafood. Fish and shellfish are the primary food exposures (Chohan et al., 2021; Jian et al., 2018; Sunderland et al., 2019) *AFFF are aqueous film forming foams*

Typical point emission sources of PFAS are runoff from airports, and industrial emissions from plants that produce and/or use PFAS (American Water Works Association, 2019). PFAS have long been used in fire-fighting foams, due to its stability and surface-active properties, that enables it to form a film on top of the burning liquids and extinguish the fire (Brendel et al., 2018).

It has been concluded that the extensive use of aqueous film forming foams (AFFFs) is one of the most important point sources of PFAS to lakes and ground water within urban areas (Filipovic et al., 2015). The primary usage of AFFFs is at military bases, airports and oil rigs. (Moody & Field, 2000). One of the first studies to address the contamination of groundwater due to the leakage of AFFFs and PFASs was done by Moddy & Field in 1999 . Several other studies have pointed out the usage of AFFFs as a potential source of PFAS to the environment (Ahrens, Lutz et al., 2010; Moody & Field, 2000; Moody et al., 2003).

PFOS and PFOA have been detected in several environmental matrices, such as biota, water, sediment and sludge, whereas the major source of this introduction was from discharge of municipal and industrial wastewaters.(Yu et al., 2012)

Another discharge source that appears to be minor, but not insignificant, are landfills, in the form of leachates. While landfills are gradually moving from being traditionally "dumping grounds" to a place where there is more focus on recycling and improved waste treatment methods, waste fractions in former landfill sites may have the potential to leach significant PFAS amounts. It has been reported that the concentration of these emissions are considerable but varying across Norway (Knutsen et al., 2019).

Leachate is produced at landfills from the infiltration of precipitation through solid waste and from liquid waste migrating down (Masoner et al., 2020). With no suitable treatment methods to reduce the concentration of PFAS in the leachate, it will eventually result in contamination of nearby waterbodies and/or groundwater. There is still a lack of research about the fate and transport of PFAS in groundwater, but if not treated, the contamination will pose a potential risk and hazard to the public health and to the ecosystem (K.Y.Foo & B.H.Hameed, 2009; Sinclair et al., 2020).

1.5   Potential material for PFAS remediation

The method which has shown to be the most efficient in the treatment of PFAS contaminated leachate over the past decades is the use of activated carbon (AC). A surface phenomenon, with its large surface area, thermostability, low acid/basic reactivity and pore structure, makes it a significant sorbent. Due to these properties, activated carbon has a great ability to remove a large variety of organic and inorganic pollutants, that are dissolved both in the aquatic and gaseous environment (K.Y.Foo & B.H.Hameed, 2009).

There are currently limited number of technologies available for removing PFAS from water *in-situ* (on site) , with many lacking technical maturity such as PFAS destruction or long-term sequestration (Riegel et al., 2020). Although the AC doesn't remove the

PFAS contaminants from the environment, it contributes to limit its leaching within an already affected area and reduce the potential of damage to the surrounding environment. Environments such as groundwater, surface waters, rivers, ecosystems, etc are common recipients (Darlington et al., 2018).

Figure 4 is given a simplified and idealized scheme of the immobilization of PFAS by adding a sorbent.
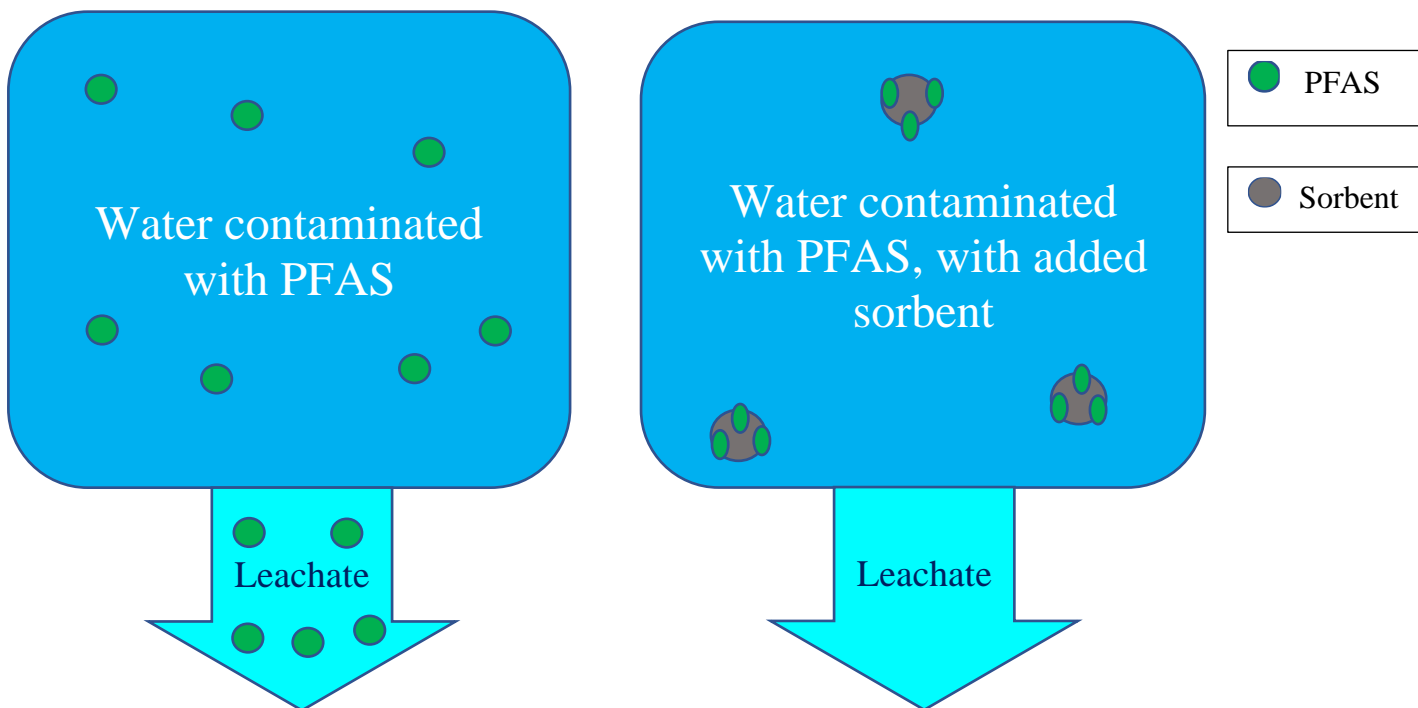
Figure 4. Schematic presentation of the immobilization of PFAS (sorption and stabilization) method. The leaching of PFAS from contaminated water is prevented by mixing suitable sorbent into the water, thus protecting the surrounding environment from PFAS contamination.

While there will most likely be technological advances in rehabilitation methods for PFAS contaminated areas in the nearest future, but until then AC filters will remain the most common method worldwide to prevent PFAS compounds from leaching into the environment (Hale et al., 2017).

AC is a non-renewable resource which is derived predominately from coal sources, and its extraction and activation process has quite a large environmental footprint. While greener alternatives such as activated biochar made from waste-based materials are surfacing as a more sustainable option (Chen et al., 2019; Thompson et al., 2016),

it may be possible for spent AC from numerous industrial applications (e.g., water and gas/odour purification) to be reused in the treatment of PFAS contaminated water. It is speculated that spent AC from some industries is sent to landfills before the theoretical sorption capacity has been exhausted. With an increasing focus on circular solutions, it may be possible to reduce the consumption of virgin AC by introducing the spent version into PFAS treatment regimes.

Lindum is one of the leading companies in Norway that manages various types of organic waste and landfilling of contaminated masses. They have designed a landfilling method for PFAS- contaminated soils from fire-fighting areas in Norway. These soils are deposited in their own sealed cells which include the use of a sorbent layer of slag. This slag comes from the metallurgical industry in Norway and is a suitable sorbent due to its relatively good sorbing properties to PFAS compounds, in addition to maintaining its permeability under high loads (Lindum, 2019). During the filling phase of the PFAS cells, precipitation percolates through the deposited soil and takes with its soluble PFAS compounds.

Most of the PFAS in this water is filtered by the slag layer, however AC is used in backup filters which are positioned at the outlet of each of the cells to further reduce the amount of PFAS being sent to the landfill leachate system. Once a cell is filled, it is then covered with a water repellent membrane so that the leachate amounts eventually are completely abated.

The potential for using spent AC as a filter medium at PFAS-contaminated locations depends not only on its ability to filter PFAS compounds. There is also potential for the contamination, that the AC has adsorbed from its original assignment, to desorb (wash off) from the surface. It is therefore essential to ensure that this contamination is properly bound to the carbon so that they do not pose any additional risk for the environment.

1.6 Various PFAS treatment

There are several ex-situ (off-site) treatment forms for PFAS removal from groundwater. The most used methods are sorption to AC, reverse osmosis, ion exchange resins and nanofiltration, where AC sorption is the most widely used treatment. Other technologies like chemical oxidation, chemical reduction, electrochemical, sonochemical and Surface Active Foam Fraction (SAFF) are being developed, and have so far shown some promising results (Kucharzyk et al., 2017)

The latest technique (SAFF) is something Australia and Sweden are working with, where they use air bobbles to treat PFAS. (Burns et al., 2021; Envytech, n.d)

So far, there has been limited activity regarding the application of *in-situ* PFAS treatment. Full-scale in situ technologies have not yet been documented, and pilot-scale applications have been few (Kucharzyk et al., 2017).

Several studies in Norway examine how different sorbents act as soil amendments in PFAS contaminated soil.

One study compared the adsorption efficiency between AC and montmorillonite in a PFAS-contaminated airport site. The AC reduced the leaching of PFAS by 94 to 99%, while montmorillonite reduced it by 28 to 40% (Hale et al., 2017).

Another study compared the adsorption between AC and biochar. The AC removed almost all PFAS from the water phase, while the biochar did not noticeably affect the PFAS concentration (Kupryianchyk et al., 2015). Other studies with activated biochar have revealed promising results in reducing PFAS contamination from soil (Qvale, 2021; Sørmo et al., 2021). EarthresQue, a "newly" founded centre for excellent innovation will create and improve new methods and processes for reusing, recycling, and treating contaminated soil. This new focus on recycled materials has increased the value and interest in recyclable materials and thus increases the interest in the circular economy. This is also in line with Lindum's (see section 1.5) effort in their research and development to increase the value of the waste material, by recycling it for newer purposes, such as energy production and stabilization of contaminated soils, etc. Hence, the objectives of this study (next section) fall exactly in line with these efforts and values, as the present study explore the potential use of spent AC as a PFAS sorbent.

2 Objectives and hypotheses

2.1 Research Objectives

The overall objective for this study was to investigate the potential of recycling spent-AC from various industrial applications in the use of filtering PFAS-contaminated water.

The specific objectives of this thesis were to

- Investigate the PFAS sorption properties of several different spent AC samples from 3 different companies via laboratory batch-tests.
- Investigate the extent of desorption of already bound contaminants in the spent AC samples.
- Test samples which show potential from laboratory experiment in a large scale *in-situ* experiment.

If some of spent AC samples tested successfully bind PFAS in the batch test, the primary output will be knowledge that can lead to their potential use in reducing PFAS leaching from PFAS contaminated areas and landfills, thus making good economic use of spent AC and by that increase its circular economy.

2.2 Scientific Approach and Hypothesis

In this study PFAS sorption capacity was tested on spent AC samples from five companies in addition to one anthracite sample from another company. The samples were collected from wastewater treatment plants, air purification plants, and biogas plants. The study utilized batch tests with water containing a high concentration of PFAS, and where possible unused samples of the same AC type were tested in parallel to allow for easier comparison. Samples containing high concentration of contaminations were also tested separately in batch tests in order to assess the leaching potential of these contaminants.

Depending on the type of previous industrial application, the spent AC will have a varying degree of PFAS sorption efficacy, and thus are following hypotheses are defined:

(i)   Samples from applications involving filtering other organic compounds will be the least effective as less sorption sites will be available.

(ii)   PFAS sorption capacity for the spent AC samples will be less compared to the unused AC samples.

(iii)   Desorption of contaminants already bound to the spent AC will be the most prominent for contaminants with low carbon to water partition coefficients (K_{OC}).

The hypotheses will be tested by making use of several batch tests involving different amounts (liquid/solid ratios) of spent AC samples to test their sorption properties. Analysis of the different AC samples for the content of other contaminants will also be performed for characterisation purposes.

Theory

2.3 PFAS remediation

The strong C-F bond of PFAS compounds and their electron-withdrawing ability of their functional groups makes them resistant to ordinary water treatment disinfections like UV irradiation and the addition of free chlorine. Additionally, biological treatment will, with and without oxygen result in breakage of the C-C bond that only will lead to the formation of short-chain PFAS, making remediation of PFAS a challenge (Gagliano et al., 2020). On the other hand, adsorption is considered a preferable way to remove PFAS from wastewater or water. An overview of PFAS sorption patterns and the sorbent patterns will be given in the next section.

2.4 Adsorption mechanisms of PFAS

Adsorptions mechanisms such as electrostatic interactions, hydrophobic interactions, van der Waals forces (dipole-dipole interaction), hydrogen bonds and electron exchange are possible interactions involving PFAS adsorption to various sorbents (Deng et al., 2012). The major PFAS adsorption mechanisms are electrostatic and hydrophobic interactions.

Electrostatic interactions are those between the anionic PFAS and the positively charged sorbent. The electrostatic negativity is originated from the functional head of the PFAS compound (Du et al., 2014). Hydrophilic interactions are those between PFAS and non-polar hydrophobic surfaces (Dixit et al., 2021). The occurrence of hydrophobic interactions depends on the length of the C-F chain. It has been found that the presence of additional C-F chain results in a stronger hydrophobic adsorption (Gagliano et al., 2020).

Considering that the electrostatic interactions are taking place at the hydrophilic functional "head", while the hydrophobic interactions are taking place at the hydrophobic C-F "tail" of PFAS (Du et al., 2014; Gagliano et al., 2020), it seems most likely that the electrostatic interactions are more specific to the short-chained PFAS compounds, while the longer-chained compounds are more prone to hydrophobic interactions.

2.5   Adsorption behaviour

Factors like solution pH, co-existing compounds in the water and the properties and characteristic of the different PFAS compounds and the sorbent (i.e., functional groups, porosity and polarity) will affect the adsorption behaviour of PFAS.

Changes in pH in the solution will affect the PFAS molecules and the surface properties of the sorbent, it has been observed a inversible correlation between PFAS sorption and the pH in the solution (Du et al., 2014).

As reported in several studies, the adsorption affinity to PFAS decreases with increasing pH in the solution (Bolan et al., 2021; Gao et al., 2017; Qian et al., 2017).

Increasing pH in a solution leads to a more negatively charged sorbent surface, resulting in a weaker attraction to the PFAS anions (Du et al., 2014).

Co-existing compounds such as inorganic anions and organic matter can hinder the adsorption to PFAS by changing the solution's pH and competing for the adsorption sites on the sorbent (Du et al., 2014; Zhang et al., 2019).

PFAS sorption behaviour can be quantified by using partitioning coefficients such as K_d and K_{OC}. The coefficient values differ depending on the environment, chemical properties and the method used. In general, the higher the K_d/K_{OC}, the stronger the affinity to the sorbent (Bolan et al., 2021). A high K_d indicates that the PFAS compound is immobile and very strongly absorbed into the soil, thus resulting in a high concentration of PFAS in the sediment. Alternatively, PFAS compounds with a low K_d values, which mainly exists in the dissolved phase, are very mobile and capable of dispersing throughout an aquatic environment (Ahrens et al., 2011).

2.6   Other contamination in spent AC

BTEX and sulphur are two volatile compounds typically found in AC from air/gas purification. These can cause environmental harm if released through leachate from the spent AC. They may desorb if they are exposed to a new load of organic compounds like PFAS, that can compete with the sorption sites.

2.6.1 BTEX

Benzene, toluene, ethylbenzene and xylene (BTEX) are volatile organic compounds (VOC) that are commonly found in landfill gasses. Landfill gasses are made by decomposition of organic waste in the landfill. BTEX is classified as a hazardous air pollutant (HAP), due to the variety of health problems upon exposure. BTEX can cause cancer, respiratory irritations and nerve damage (Durmusoglu et al., 2010).

By using activated carbon in landfills, it can reduce or remove the emissions of BTEX. Since both BTEX and AC are organic compounds, they will have some kind of affinity to each other, and BTEX will be absorbed to the AC trough electrostatic interactions (Daifullah & Girgis, 2003).

2.6.2 Sulphur

Sulphur (S) is an element and an essential nutrient for microorganisms and plants. It is present in several inorganic and organic forms and oxidation states from -2 to +6.

Its neutral state does not cause any environmental harm, but there are several factors influencing the fate of sulphur in the environment, such as oxygen availability and the microbial community.

For example, sulphate (SO_4^{2-}) will be reduced under anaerobic conditions. This reduction results in gaseous emissions such as hydrogen sulphide (H_2S) and dimethyl sulphide (CH_3). H_2S is toxic to aquatic life, because it contributes to acidification of the water, and it will corrode engines if used in biogas (Mamun & Torii, 2015; vanLoon & Duffy, 2017).

Sulphur occurs in water as sulphate (SO_4^{2-}). This anion has four oxygen atoms attached to it, with a recommended maximum concentration of 250 mg/L in drinking water in Norway (Drikkevannsfroskriften, 2016). Water containing high concentrations of sulphate can cause diarrhoea and dehydration due to its laxative effect (Miao et al., 2012).

As sulphur occurs in several forms, it is difficult to describe precisely how it is adsorbing to the AC, but several studies suggest electrostatic interaction is the dominant mechanism and the presence of basic surface functional groups enhances the sorption of sulphur forms like sulphur dioxide and hydrogen sulphide (Bagreev et al., 2002; Lee et al., 2002; Yan et al., 2002).

2.7 Activated carbon

Activated carbon (AC) (also called activated charcoal or activated coal) is a manufactured type of charcoal, which has very strong absorbing properties, due to its high porosity, extensive interparticle surface area that can be up to 1.000 m^2/g, controllable internal pore structure, consisting of macro-, meso- and micropores and together with its wide spectrum of functional groups on the surface, such as oxygen, hydrogen, sulphur and nitrogen (Bansal & Goyal, 2005; Bhatnagar et al., 2013; Chingombe et al., 2005; Foo & Hameed, 2009; McDougall, 1991). AC has a non-polar surface, that makes it suitable for absorption for hydrophobic compounds (Du et al., 2014). Figure 5 illustrates a particle of AC and the location of the different pore structure characteristics.
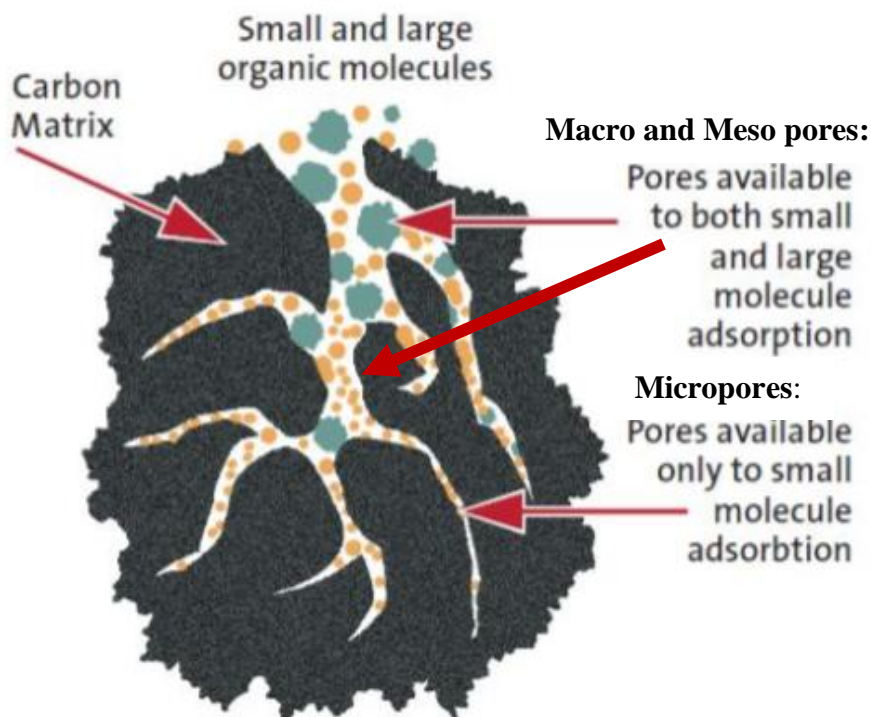
Figure 5: Illustration of the different pore sizes and where they are found in AC, inspired from (ELGA, 2021; McDougall, 1991).

AC comes in several forms, such as pellets, powdered activated carbon (PAC) or granulated activated carbon (GAC), all with different properties and fields of application.
The most important applications are removal of volatile compounds, treatment of industrial waste water, purification of gas and air and treatment of leaching in landfills (Foo & Hameed, 2009). Samples used in this thesis are pellets (4mm diameter) and GAC variant.

GAC has many irregular shapes and has a particle size ranging from 0.2 to 5mm. GAC is mainly used for water treatment, while pellets with an 4mm diameter (Sc40) are mainly used for air and gas purification (Silcarbon, n.d.). This is due to the different densities and pore sizes of the forms.

2.7.1 Production of activated carbon

AC is produced by chemical or steam activation of carbonaceous rich materials, like wood, coal, peat, lignite or coconut shell (Amstaetter et al., 2012; Bhatnagar et al., 2013). The coal variant (fossil-based-material) is the most traditionally produced and used.

Activation is the most important step in the production of AC. In this step, the raw material is converted into a crystalized form of carbon that contains the greatest possible number of randomly distributed pores of various shapes and sizes resulting in an increased pore volume, improved pore diameter, pore structure and porosity (Bansal & Goyal, 2005; McDougall, 1991; Sinha et al., 2020). The activation is typically performed in an atmosphere of air and CO_2 or steam and involves two steps. The first step involves carbonization at 400-500 ^{O}C to remove most of the volatile matter, followed by incomplete gasification at 800-1000 ^{O}C to develop porosity and surface area (Bansal & Goyal, 2005; Yang, 2003).

2.7.2 GAC in water treatment

Although GAC is an effective filter of PFAS in water treatment, the presence of dissolved organic material (DOM) has the potential to interfere with the absorption of PFAS during the treatment. Due to its molecular structure, natural organic material (NOM) such as humus can compete with PFAS for the same sorption sites on the AC (Kothawala et al., 2017). The adsorption of organic compounds have also shown to increase the negative charge on the AC (Newcombe et al., 1993).

It is known that NOM influence the adsorption of organic compounds to AC, through two significant competitive mechanisms; direct competition on surface site and pore blockage (Yu et al., 2012). GAC used in water treatment can thereby have different absorption properties depending on the amount/presence of organic material.

2.7.3 Pellets inn air and biogas purifications

Pelletized activated carbon has a cylindrical form, with diameters ranging from 0.8mm to 5mm, where the range 3 and 4mm are most commonly used in air and gas purification (Silcarbon, n.d.). The uniform shapes and different manufacturing conditions that can manipulate the pore structure makes it perfect for a variety of uses, like controlling odour, removing mercury from natural gas and removing volatile organic compounds (VOC) like BTEX (Chemviron, n.d).

Methane (CH_4), carbon dioxide (CO_2), and hydrogen (H_2) are the main components of biogas, with some traces of hydrogen sulphide (H_2S) and dimethylsulfide (C_2H_6S). AC in biogas purification is primarily used to remove sulphur compounds such as H_2S from the biogas generator since they can cause corrosion on both the engine and metal components (Mamun & Torii, 2015).

2.7.4 Adsorption mechanisms

The adsorption properties of AC can be divided into two types, physical and chemical. Physical adsorption involves weak van der Waals forces and hydrogen-bonding, while chemical adsorption involves exchanging or sharing of electrons (covalent bonding). Chemical adsorptions are much more stable and generally more irreversible than the physical ones (Bansal & Goyal, 2005; McDougall, 1991).

However, several factors influence the adsorption behaviour of AC, such as the pH value in the solution, temperature (increasing temperature will decrease the adsorption), the functional groups and the physical properties of the AC (e.g. particle- size distribution) (McDougall, 1991). Phenols (-OH), carboxyl (COOH), carbonyl(C=O) and others are the main functional groups that are responsible for uptake of pollutants in the carbon structure.

However, the adsorption properties and concentration of surface functional groups can be altered by several surface modification mechanisms. These techniques purposely attach some functional groups to the surface, to enhance the reactivity and enhance the adsorption property for specific contaminations (Bhatnagar et al., 2013; Sinha et al., 2020).

AC has been widely reported as an effective sorbent for various pollutants, but has a bigger affinity to organic contamination than inorganic or metals (Bhatnagar et al., 2013).

The modification of the chemistry of the surface of activated carbon is regarded as an optimistic and attractive way towards use in new applications. Modification involves oxidation on the AC surface by chemical, electrochemical and/or microwave methods to introduce new functional groups (Bhatnagar et al., 2013). For example, acidic treatment has shown to be very efficient in removing heavy metals from water.

Most of these mechanisms require a lot of energy and thereby have a high economic cost. The average energy demand of producing AC has been found to be 97 MJ/kg, whereas biochar demands only (6.1 MJ/kg) (Alhashimi & Aktas, 2017).

2.7.5 AC vs. biochar

While AC is produced from coal and goes through an energy consuming activation process, biochar is produced when biomasses undergo thermal decomposition with no to little oxygen supply in a process called pyrolysis. The biomasses that are used in the pyrolysis process are carbon rich waste products, such as crop residues, straws and waste timber (Chen et al., 2019). The temperature needed to produce biochar is substantially lower than what is needed for AC production. Depending on the biomass and type of char wanted to be produced, the required temperature ranges go from 200 to 700 ^{O}C, whereas the temperature for making AC is between 400 to 1000 ^{O}C (Aller, 2016; Yang, 2003).

Although the use of biochar may steadily increase in the coming years, fossil-based AC is likely to remain the most popular alternative. Considering the significant energy difference of 93% between AC and biochar, it is important to maximise AC's utilisation, by ensuring that it is used to its maximum potential before being sent to landfills.

3   Materials and Methods

To determine how spent AC can immobilise PFAS in contaminated water, PFAS-contaminated water was collected and mixed with various ratios (L/S) of different AC materials (see table 3).

The different spent AC was sent to a Eurofins, an external laboratory to be analysed for their content of sulphur (S), various heavy metals and organic substances, such as BTEX, polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH), total organic carbon (TOC) and dissolved organic carbon (DOC).

The analysis was performed at the Norwegian University of Life Science (NMBU). An overview of the series of operations is given in figure 6.

Sampling of water and AC

Water sampling
- Water from leachate from PFAS contaminated soils

AC sampling
- AC retrieved from various companies

AC analysis

Chemical analysis for existing contaminants including:
- Sulphur content (external laboratory)
- Heavy metals content (external laboratory)
- PFAS content (external laboratory)
- BTEX content (external laboratory)
- PCB content (external laboratory)
- PAH content (external laboratory)
- TOC/DOC content (external laboratory)

Batch leaching/shaking test

Batch shaking test for sorption of PFAS to AC

Batch shaking test for leaching of BTEX and sulphur from AC

***In-situ* experiment**

PFAS adsorption to the most promising spent AC sample observed from lab experiments

Figure 6: Flowchart that gives a summary of this thesis 'materials and methods'

3.1 Consideration when working with PFAS

When working with PFAS contaminated materials, some consideration must be taken to avoid unnecessary contamination of the sample. Using the right materials and equipment are essential in this case. Materials that can absorb or contain fluorinated compounds must be avoided, both during the sampling and the handling process. Equipment made of high-density polyethylene (HDPE) or polypropylene (PP), glass or stainless steel are recommended materials that should be used when coming in direct contact with the PFAS- contaminated samples. To minimise the risk of contamination, these materials were cleaned. Methanol is a solvent that is recommended to use as a washing solution, since it can extract potential fluorinated compounds (Ahrens, L. et al., 2010; Standard Norge, 2003).

In addition to the choice of the materials and equipment, the way they are cleaned and handled can be a significant source of error. Water samples should be stored at -20 ^{O}C if it is not possible to analyse the samples within a few days. Due to the potential of biotransformation in biologically active samples, they should be analysed within two weeks (Ahrens, L. et al., 2010).

In this study, the water samples were stored at 4 ^{O}C and were often not analysed within the two weeks as they did not contain living organisms.

3.2 Sampling site

The sampled of PFAS-contaminated water was collected from Lindum Oredalen, one of Lindum's landfills on the Hurum peninsula, approximately 60km south of Oslo.

As previously mentioned, this landfill receives and stabilises masses from various PFAS contaminated sites around Norway, with soil from airport fire-fighting practice areas being the most common. The treatment method was described in Section 1.4, and leachate samples were taken directly from their PFAS cell (cell1-2) and stored in 20L HDPE containers.

3.3 Sorbent materials – Activated carbon

Spent AC samples were retrieved from three different industrial applications and six different companies.

The samples were retrieved from the air purification companies CLAIRS and Yara, the biogas

purification plants "*The magical factor (TMF)* and Lindum and the waste-water treatment plants *"Nedre Romerike Vassverk"* (NRV) and *"Kristiansand Kommune vannverk"* (KKV). Pellets were used in the air and biogas purification plants, while GAC was used in NRV and anthracite in KKV. It should be noted that the CLAIRS AC (SC40 type) was used by both biogas plants.

Where possible, unused AC samples were also sampled for comparative reasons. Table 3 outline the AC characteristics and sources.

Iodine number and specific surface area are two important factors which determine the adsorption capacity of AC. The iodine number indicates the microporosity of AC and is defined as the amount of iodine absorbed per 1g of carbon at mg-level (Saka, 2012). Higher iodine values will result in higher adsorption efficiency due to higher porosity.

The specified surface area is the total surface area of solid particles divided by the total mass of solid particles. A larger surface area generally increases the potential for adsorption.

Table 3: Activated carbon samples and their sampling locations.

Companies	CLAIRS	Yara	TMF	Lindum	NRV	KKV
Setting	Air purification (Pellets)		Biogas purification (Pellets)		Waste-water treatment (GAC) and (anthracite)	
Iodine number (mg/g)	>1050		>1050	>1050	850	
Specific surface area (m²/g)	1100		1100	1100	850	

3.4 Experimental setup

The experimental setup of this study is composed by three separate parts.

(1) The first part is designed to investigate the degree of leaching of these contaminants, particularly BTEX and total sulphur. A leaching test with PFAS contaminated water was executed in this part.

(2) The second part is focused on the sorption capacity of PFAS to the spent AC, samples via the use of batch leaching tests. These samples contained varying concentrations of contaminants from their original assignments.

(3) The third and final part includes the completion of a large scale *in-situ* experiment at Lindum's Oredalen landfill.

The one-step batch leaching test for part 1 and 2 were done according to the NS - EN 12457-2 method (Standard Norge, 2003), with a few modifications. The samples were put on a shaking table with a lateral shaking motion, instead of shaking end-over-end. The leaching test was left to settle for 1 hour instead of centrifuging before filtration.

3.4.1 BTEX and sulphur leaching test (1)

To test the impact of PFAS contaminated water on the potential leaching of sulphur an BTEX from the spent AC, a simplified batch experiment was conducted.

One-step leaching test was conducted in 2L glass bottles (containers), where 1800 ml PFAS-contaminated water was amended with 180g spent AC (L/S 10).

The samples were left on the shaking table for 24 hours as per the standard EN 12457-2.

Three samples were investigated from each spent AC sample from both biogas plants and both air purification companies, with a total of 12 samples (3 x 4).

After shaking, the samples were left to rest (settle) for about 1 hour and then filtered through a 0,45 µm, cellulose acetate filter. After the filtration, the samples were transmitted to 500mL HDPE bottles for sulphur analysis and 500mL glass bottles for BTEX analysis. Then samples were then sent for analysis to the Eurofins accredited laboratory.

From the average leachate concentration of BTEX and sulphur in the eluate samples, it was then possible to estimate how much of the original content in the AC samples had leached out. See appendix F for calculation

3.4.2 PFAS sorption test (2)

The one-step batch shaking test was conducted in 1L High Density Polyethylene (HDPE) bottles, where PFAS-contaminated water and AC were added at three different liquid to solid ratios (L/S). This was because a minimum of three data points is required to create a linear test of K_d developed by different L/S ratios (Eq. 1, section 3.7)

As the L/S ratios used (80/100/200) all resulted in near 100% removal of PFAS compounds from the water, it was decided to increase them to100/500/2000 and finally 1000/2000/8000 in order to give a better indication of sorption capacity.

The samples were left on the shaking table (Edmund Bühler, SM-30) at a speed at 100 shakes/min for 14 days (as opposed to 24 hours in the standard) to allow sufficient equilibrium time for PFAS compounds.

Samples were prepared in triplicates for each of the three L/S ratios. Triplicates were used to ensure the results were valid and so spoiled samples would not ruin the study. Triplicates were also used for control samples (i.e., only PFAS water, no AC), making it 12 samples for each AC sample. In total, 120 samples were tested.

The samples were put to rest for 24h (in room temperature) after shaking and then transferred to 50mL (methanol- cleaned) falcon tubes. These falcon tubes were then centrifuged for 10 minutes at 28000 rpm (MEGA STAR 1.6 VWR) to separate the solids phase from the liquid. A total of 72 centrifuge tubes were used for each AC sample, making it a total of 720 tubes. The particle-free water in the upper part of the tube was thereafter carefully poured into 100-mL HDPE PFAS bottles specifically designed by the Eurofins laboratory, and pH was measured using a VWR symphony SB7OP pH meter. The samples were then packaged and sent by a postal service the same day to Eurofins' accredited laboratory in Moss, Norway for PFAS and DOC analysis.

3.5   *In-situ* experiment using spent AC sample (3)

Once all testing was completed in the laboratory, it was decided that a large *in-situ* experiment would be done on one of the spent AC samples which showed the most promising lab results. The main aim with this experiment was to test this samples sorption capacity compared with an unused sample of the same AC type

3.5.1   Experimental site

The experiment was done at Lindum's Oredalen landfill site in Hurum, Norway. This landfill has received PFAS contaminated soil from areas around Norway into three sealed cells (as mentioned in Section 1.5). All three cells contained soils from PFAS contaminated airport and military grounds, however the third cell also contained a large percentage of PFAS contaminated drilling mud. "Drilling mud is crushed masses of rocks that have been transported to the surface from the drilling after oil and gas at an oil platform" (Hammer, 2011). PFAS is used as a foaming agent to minimize the amount of fluid lost during drilling and to reduce the potential formation damage (Norwegian Environmental

Agency, 2021). AC is normally filled into concrete catchpits placed directedly after the cell outlets to remove PFAS from the leachate which is not filtered by the slag layer in the cells. For this experiment, the unused AC was replaced with spent AC.

3.5.2 Experimental setup

Cells 1 and 2 have a common outlet for the leachate, whereas cell 3 has its own, as can been seen in figure 7. Spent AC was filled in the final catchpit while unused AC was filled in the catchpit directedly after cell 3. This is because the drilling muds contained considerable amounts of organic salts and heavy metals which can negatively interfere with the adsorption of PFAS, thus making the spent ACs only job to absorb the PFAS.

The leachate from the cells flows through four different sampling points (#1, # 2a, #2b, #3), before it flows into the common leachate network for the landfill (figure 7). Site #1 consisted of leachate directly after flowing out of cell 3, while #2a samples were taken after the leachate had flowed through catchpit full of unused AC. The #2b site sampled leachate directedly after cell 1-2, and site # 3 consisted of mixed leachate from cell 1-2 and 3 after flowing through a catchpit full of spent AC. Table 5 gives an overview of the various analyses done on samples at each sampling site.

Table 5: Overview over what was sampled for det separate sites.

Sampling sites			
1	**2a**	**2b**	**3**
PFAS	PFAS	PFAS	PFAS
pH	pH	pH	pH
Conductivity	Conductivity	Conductivity	Conductivity
	Sulphur	Sulphur	Sulphur
	BTEX	BTEX	BTEX

Figure 7: An overview over the sampling site. Cell 1-2 containing PFAS contaminated mases form airports and military bases, Cell 3 containing PFAS contaminated masses from drill cuttings. The blue line indicates the pipes from the cell to the wells, white arrows show the direction of the water flow. Firs well containing unused/new AC, second well containing used AC. 1, 2a, 2b and 3 indicates det separate sampling sites.

3.5.3 Leachate from cell 1-2 vs. cell 3

Since the cells contain different types of PFAS masses, the composition of the leachates will most likely vary. To compare this variation, a batch test was performed on the spent AC samples with the leachate from cell 3 (sampling site 1) in addition to the leachate from cell 1-2 (sampling site 2b). The batch tests were conducted as mentioned in section 3.4.1, using the same L/S ratios.

3.6 Analytical methods

Different types of standards and analytical methods were used for the different parameters. Table 4 gives an overview of the different types of standards and appendix A includes more detailed description of the different methods.

Table 4: Analysis methods for PFAS, DOC, BTEX and Sulphur. Limit of Quantification (LOQ), LC/MS/MS stands for the type of triplicates. SS-EN-ISO stands for a European standard written in Swedish (SS → Swedish standard)

Parameter	Analysis code	Analysis method	Description	Instrument and conditions
PFAS	PLWXZ	DIN 38407-42, UNEP Chemicals Branch 2015 modified		LC-MS/MS (QqQ) LOQ = 1ng/L
DOC	MM152	NS-EN 1484	Dissolved organic carbon (DOC) is determined on samples filtered through a membrane filter with a pore width of 0.45 m	Spectrophotometry (INDIR) LOQ = 0,3 mg/L
BTEX	SLL18	SS-EN ISO 10301		HS-GC-MS LOQ = 0,1 µg/L except for m, n- Xylen that had LOQ =0,2 µg/L
Sulphur	SLM01	SS-EN ISO 15587-2:2002 for processing samples and SS-EN ISO 11885:2009 for calculation	Concentrated nitric acid (HNO3) is added to 20ml of the samples. The mixture is boiled in an autoclave at 120 degrees for 30 min, the analysis takes place in the clear phase	ICP-OES LOC= 0,1 mg/L

SS-EN-ISO 15587-2 is standard for water quality- digestion for the determination of selected elements in water , considering part two of the standard focusing on nitric acid (SIS, 2002). ICP-OES stands for "Inductively coupled plasma-optical emission spectrometry"

3.7 Quality control

All batch tests were performed in triplicates. The unused equipment for PFAS analyses was always cleaned with methanol and dried in a drying cabinet to minimise the chances of sample contamination. It was also important to use only recommended materials when handling and storing PFAS samples - such as glass and high-density polyethene (HDPE). In retrospect, it would have been preferable to use distilled water to determine whether any of the equipment leaches or retain PFAS, before testing with the PFAS contaminated water.

3.8 Partitioning coefficients for describing PFAS sorption

The distribution coefficient, K_d (L/g), for PFAS sorption to the tested AC is calculated for the different test ratios (L/S) by the following equation:

$$Kd = \frac{Cs}{Caq} \qquad\qquad (1)$$

Where C_s is the PFAS concentration (ng/g) sorbed on the different AC materials, and C_{aq} (ng/L) is the PFAS in the sample water before mixing (C_s= PFAS concentration in leached water-PFAS measured in supernatant)

This study had a constant concentration of the contaminant in the sample water, while the sorbent concentration increased three times, resulting in liquid/solid (L/S) ratios of 8000, 2000 and 1000 or 2000, 500 and 100.

4   Results and discussions

The overall aim of these tests was to determine which of the spent AC samples had the most potential to be used in PFAS filtration, and hence most suitable for further testing in a large-scale *in-situ* experiment.

The contamination content results for the different AC samples are presented in subchapter 4.1. Subchapter 4.2 presents the concentration of PFAS and their compounds in the sample water. Results from the batch leaching tests are presented in 4.3, and adsorption batch tests are in 4.4. The PFAS capacity for the sorbents is presented in 4.5, while the concentration of DOC is presented in 4.6. Subchapters 4.7 presents the results from the in-situ experiment, and the final subchapter 4.8 gives a small summary of the different sorbents.

4.1   Concentration of contaminants in the spent AC samples

All samples were found to have concentrations of PCB, PAH and heavy metals, which were under normative values according to the classification guidelines for contaminated soils in Norway (Norwegian Environmental Agency, 2020) and were therefore not evaluated further in this study. Results for these contaminants can be found in Appendix C. PFAS, BTEX and sulphur, on the other hand, had more significant concentrations in several of the different samples (table 6). The classification guidelines for contaminated soils outlines what is considered acceptable levels in topsoil and deeper soil layers according to different land uses and uses the same classification system as mentioned in Section 1.2 (Miljødirektoratet, n.d.).

Table 6: An overview of the contaminants in the sampled AC contents. Σ PFAS, BTEX (with its different substances) and sulphur, with colour coding values. Green is good, yellow is moderate, orange is moderately bad and red is very bad. N.d = No data. Un-spent AC samples are not represented, because here re-usability is valued. Note that PFAS values are presented in µg/kg DM, while BTEX and Sulphur values are presented in mg/kg DM.

Sample	Σ PFAS (µg/kg)	BTEX (mg/kg)					Sulphur (mg/kg)
		Benzene	Toluene	Ethylbenzene	m-/-p Xylene	o-Xylene	
CLAIRS spent	49	10	220	13	44	15	5600
Yara spent	84	11	1500	18	42	14	62300
TMF spent	5	4	85	27	75	44	48350
Lindum spent	1500	9	540	100	740	110	8200
NRV spent	2,1	N. d	N. d	N. d	N. d	N. d	N. d
KKV spent	31	3	8		4		N. d

4.1.1 PFAS content in different spent AC materials tested

PFOS is the only PFAS compound with a quantified norm value in Norway, being 0.1 mg/kg dry matter (100 µg/kg DM) (NGI, 2020).

Concentrations of 1500, 84, 49, 31, 5 and 2.2 µg/kg DM of sum PFAS were detected in the spent AC samples from Lindum, Yara, CLAIRS, NRV, TMF and NRV, respectively (figure 8). The most prevalent PFAS compounds were perfuorohexanoic acid (PFHxA) and pefluorobutanoic acid (PFBA) as shown in figure 9.
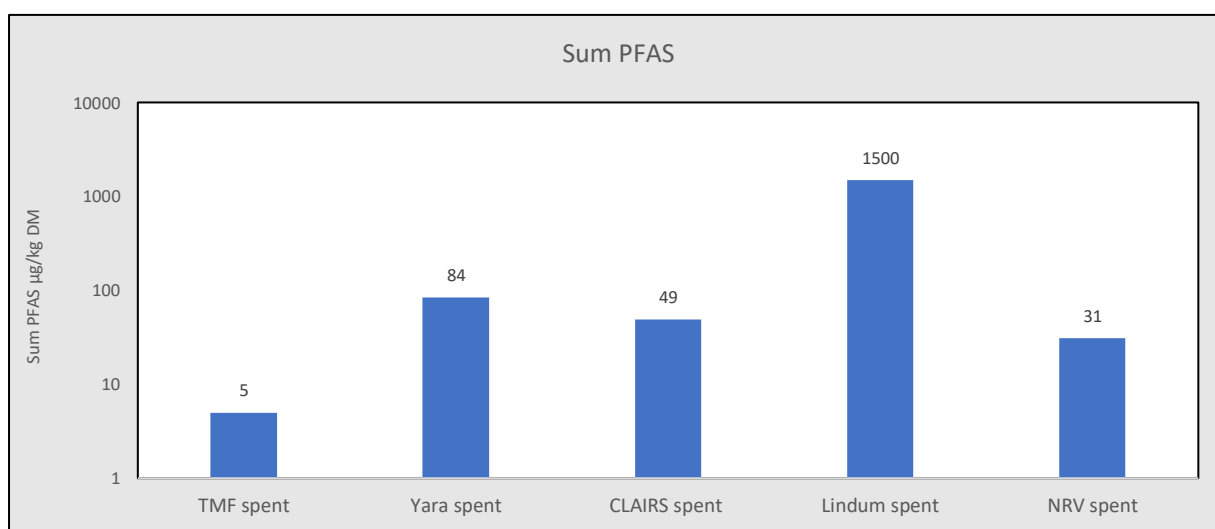
Figure 8: Total concentration of sum PFAS compounds (µg/kg DM) from the contamination analysis
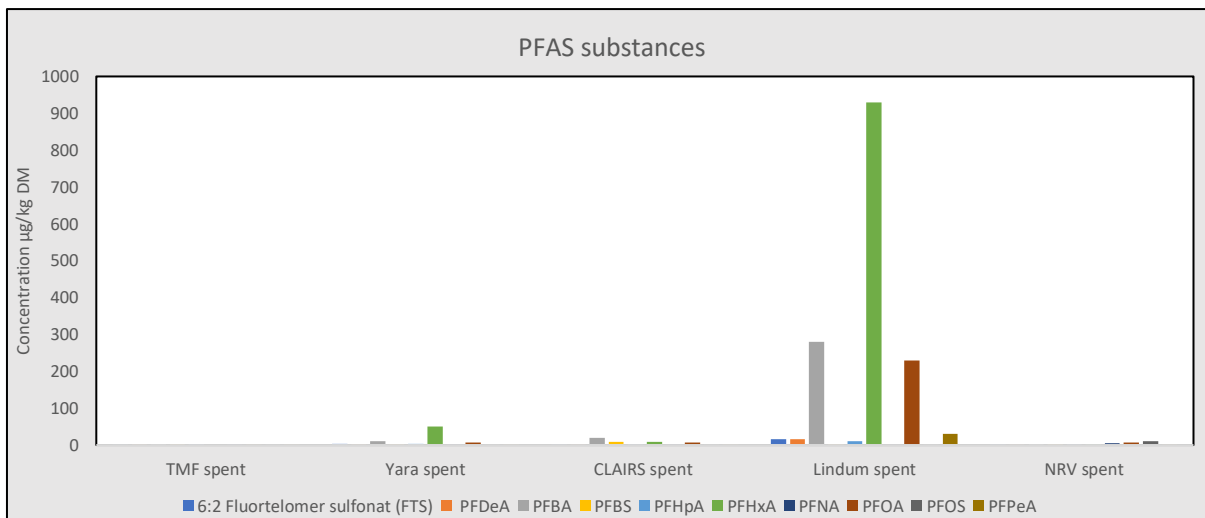
Figure 9: Overview over the prevalent PFAS compounds from each spent AC (µg/kg DM) from the contamination analysis

4.1.2 BTEX concentration

The normative values for the different BTEX compounds for contaminated soils are 0.01, 0.3, 0.2 and 0.2 mg/kg for benzene, toluene, ethylbenzene and xylene, respectively (Forurensningsforskriften, 2004). Benzene is the only of these compounds included in the classification guidelines for contaminated soils compiled by the Norwegian Environment Agency (Miljødirektoratet, n.d.).

Concentrations of 1600, 1500, 300 and 240 mg/kg DM of sum BTEX were detected in the spent AC samples from Yara, Lindum, CLAIRS and TMF, respectively (fig. 10). There are no normative values for the total concentrations of BTEX in the Norwegian guidelines, so figure 11 displays the prevalent BTEX compounds for each of the spent AC samples where, toluene is the dominating compound in the air purification samples. Both toluene and m-/p- xylene are most prevalent in the biogas samples.

The concentration of the different BTEX substances and how they are classified from the condition classes for polluted land can be seen in table 6.
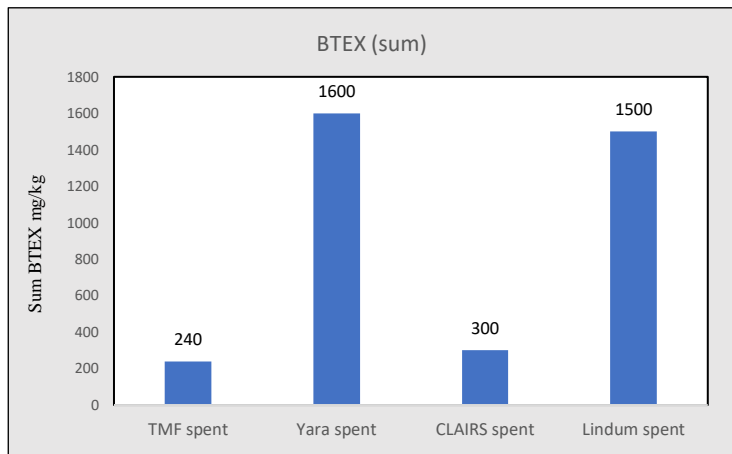
Figure 10: Total concentration of BTEX (mg/kg DM) from the contamination analysis (n=3)
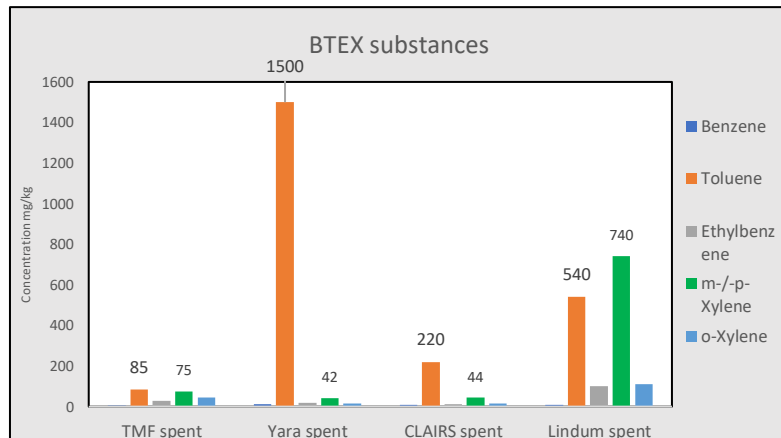
Figure 11: Concentrations of the BTEX substances, benzene, toluene, ethylbenzene, m-/-p xylene, and o- xylene (mg/kg DM) from the contamination analysis. (n=3)

4.1.3   Sulphur concentration

There are no established limits for the element sulphur in the environment, only for the harmful forms in which it occurs, such as hydrogen sulphide (H_2S), sulphate (SO_4^{2-}), sulphate dioxide (SO_2), and dimethylsulfide (C_2H_6S).

Concentrations of 5600, 62300, 48350, and 8200 mg/kg sulphur were detected in the spent AC samples from CLAIRS, YARA, TMF and Lindum, respectively (figure 12). Yara consists of almost 11 times more sulphur than CLAIRS (air purification), and TMF consists of nearly five times more sulphur than Lindum (biogas purification). These differences are most likely related to the content of sulphur compounds in the different substrates producing the gases.
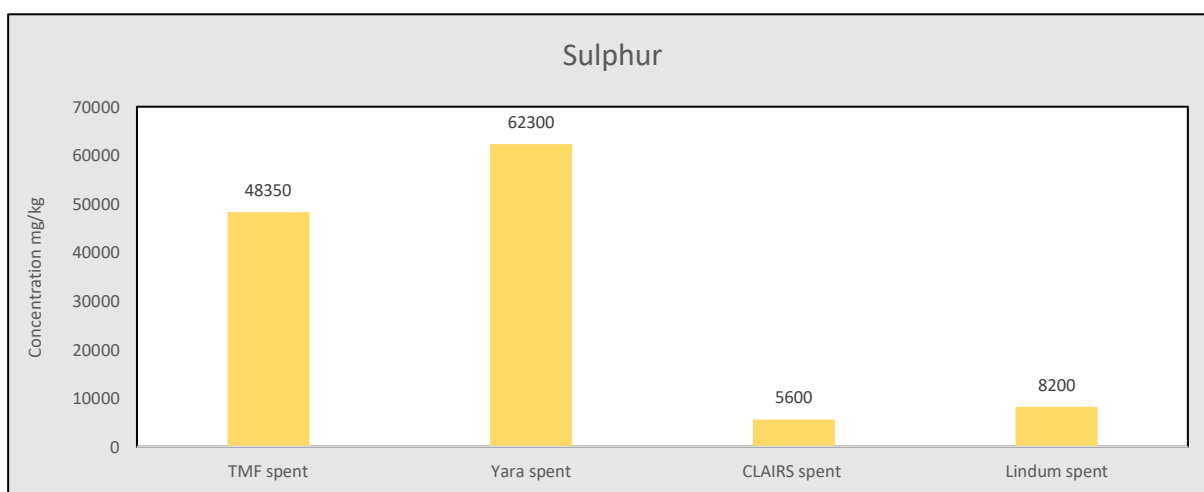
Figure 12 Total concentration of sulphur (mg/kg) from the contamination analysis. (n=3)

4.2   PFAS concentration in control water

Control samples were included in the batch tests which allowed for measurement of the difference in PFAS concentration before and after the addition of the sorbents. While there were some variations in sum PFAS concentrations in the various water samples, the concentration was relatively stable around 11000 ng/L, with the average being 10880 ng/L, see figure 13.
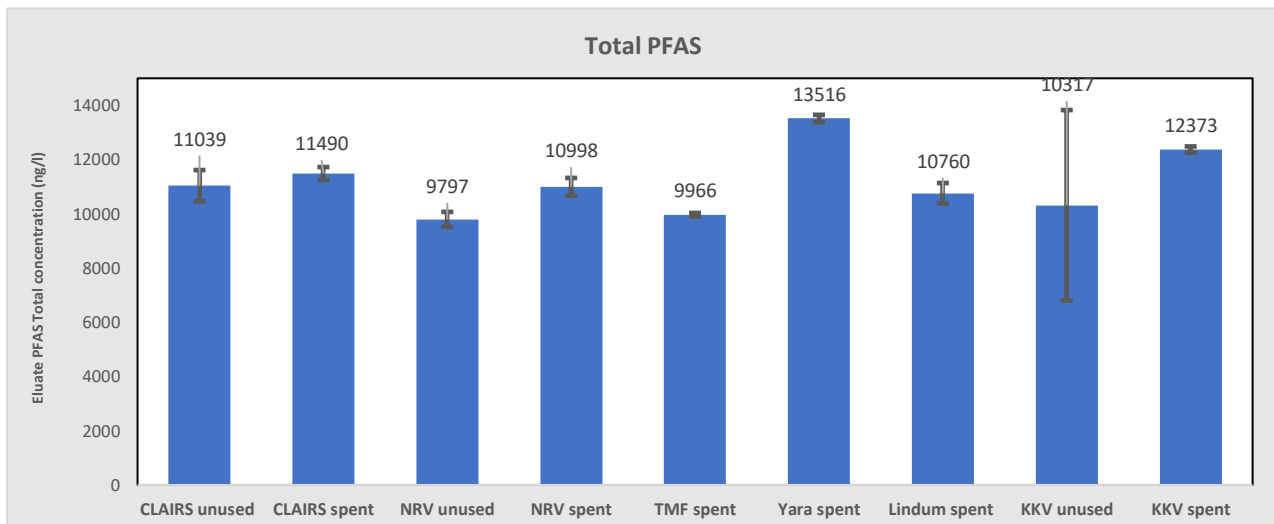
Figure 13: Total PFAS concentration from the control water for the various batch tests. Error bare are included line, (n=3)

It is unknown why the variations in PFAS concentration were so large in the different water samples, since the source is the same. However, the various soils from several different airport firefighting facilities in the cell may react differently to the physicochemical properties such as pH, the content of organic carbon, temperature and climate conditions (Gallen et al., 2017; Knutsen et al., 2019; Wei et al., 2019) producing slightly different leachate compositions with time. The sampling water was collected in 3 20L containers from cell 1-2. Changes may also have occurred in the water while it was stored in these containers throughout the testing period. The different bottles of leachate water used in the batch experiment can thus likely be connected to some of the variability seen in the lab work. Variations in the lab work, e.g., changing the size of the sample bottles, may also have caused some of the variations we see in the results.

The results in this study show a correlation between the DOC and PFAS concentrations (discussed in section 4.6), but no correlation between the pH and the PFAS concentration. There have been several studies which have discovered different results in how pH and organic content correlates with the concentration of PFAS in the leachate. An Australian study from 2017 discovered a correlation between increasing concentration of some PFAS compounds with the increased pH and dissolved organic carbon (DOC) values (Gallen et al., 2017), while a Norwegian study showed no significant correlation between the same factors (Knutsen et al., 2019).

PFAS concentrations can also be affected by lab work. People carrying out the analysis can do different things, affecting the results.

It should be noted that both the materials and the methods in the experiment were slightly altered during the experiment, the bottles used in the adsorption batch-test have been changed from 1L to 500mL bottles, but the plastic type remained the same, and the rest time before centrifugation has sometimes exceeded one day.

This difference in PFAS concentration in the water samples is important to have in mind when evaluating the effectiveness of spent AC adsorption.

4.2.1   Types of PFAS

From the total of 33 screened PFAS compounds, only 10 were found to be above detection levels in all samples. In reducing order of concentration, the detected PFAS compounds were, perfluoropentanoic acid (PFPeA), perfluorobutanoic acid (PFBA), perfluorohexanoic acid (PFHxA), perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluoroheptanoic acid (PFHpA), perfluoropentane sulfonate (PFPeS), fluorotelomer sulfonate (6:2 FTS), perfluorooctanoic acid (PFOA) and perfluorooctyl sulfonate (PFOS) (figure 14).
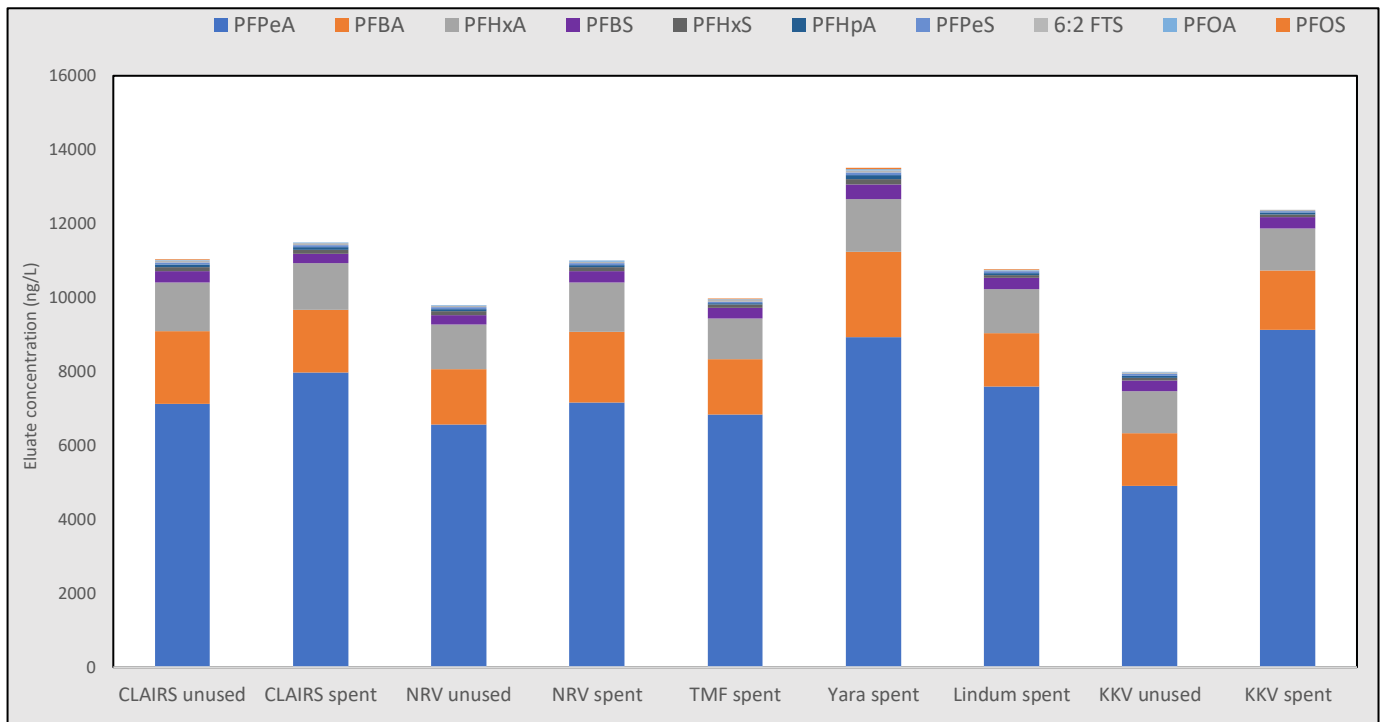
Figure 14: Ten quantified PFAS compounds from the total 33 screened. (n=3).

There were differences between the short- and long-chained PFAS concentrations, ranging from 37 ± 20 ng/L to 9133 ± 4000 ng/L for PFOA (long-chain) and PFPeA (short-chain), respectively. The short-chained PFAS types, PFPeA, PFBA and PFHxA, were overall more abundant in the leachate, representing 54%, 21%, and 13% of the total PFAS, respectively. The remaining PFAS compounds accounted for <0,61% each. A chart of the concentrations and standard deviations of PFAS can be found in Appendix D.

The dominant abundance of short-chained PFAS in leachate has also been reported from several other studies. A Chinese study observed a significant amount of PFBS (C4) present in Chinese landfills. After being treated with a two-step process, containing an external membrane bioreactor and nanofiltration unit, PFBS was still the most abundant PFAS fraction, consisting of 40% of the leachate (Yan et al., 2015). A German study observed a significant abundance of PFBS (C4) and PFBA (C4) in their leachate. After treating the leachate with AC, reverse osmosis, nanofiltration, biological treatment and wet air oxidation, they discovered a greater filtration influence in AC, reverse osmosis and nanofiltrations, than in the biological treatment and wet air oxidation (Busch et al., 2010). A Swedish case-study

observed a dominant abundance of PFHxA (C6) and PFPeA (C5) in the leachate from a waste-to-energy stockpile (Björklund et al., 2021). Based on these four studies (including this one), there is no pattern in which short-chain types are the most dominant in the leachate, although short-chained types are always more abundant. As mentioned in section 4.1, this study focuses on leachate from PFAS contaminated soil from fire-fighting facilities.

Short-chained PFAS have a lower sediment-water partitioning coefficient (K_d) than the long-chained PFAS, meaning that they are significantly less bioaccumulated than the long-chained PFAS, and they are more mobile in the environment. Thus, the relatively high abundance of short-chained PFAS observed in this and other studies can partly be due to the high solubility in water. Short-chained PFAS compounds are also occurring more in consumer products after the restriction of long-chained PFAS compounds were imposed in the Stockholm Convention, as mentioned in section 1.2.

4.3   Contaminant leaching tests

The contamination analysis reveals the composition of various contaminants in the spent AC. However, it does not reveal how well the AC retains these contaminants when immersed in water. The following section will demonstrate the leaching of BTEX and sulphur from the various spent AC materials, which were the main reasons of concern in the air purification and biogas AC samples. More information about the calculation and concentration can be found in appendix E.

4.3.1   BTEX leaching

Although four of the spent AC samples had a high concentration of BTEX, as shown in table 6, the leaching of BTEX from the samples was only 0 to 0.04% in comparison to the contamination analysis. This percentage of leaching is further explained in Appendix F. Figure 15 shows the average leaching of BTEX (µg/kg AC) at an L/S 10, while figure 16 shows the concentration of the different substances of BTEX.
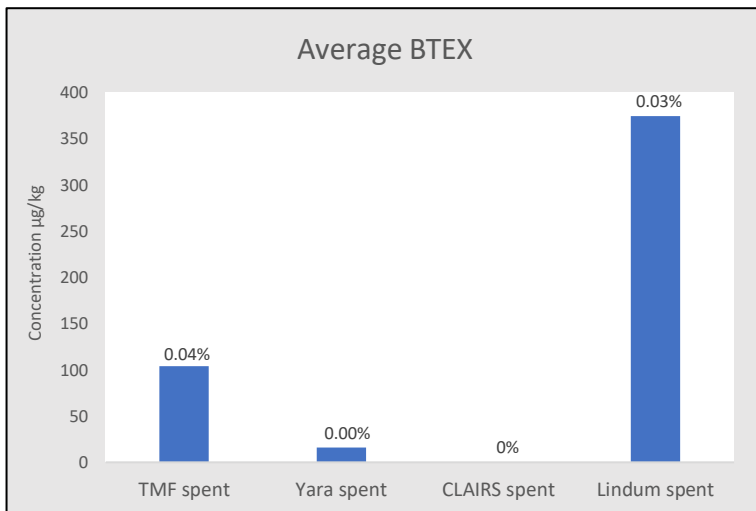
Figure 15: Average BTEX concentration from the spent AC from TMF, Yara, CLAIRS and Lindum, with the total % leakage from each of the samples (n=3)
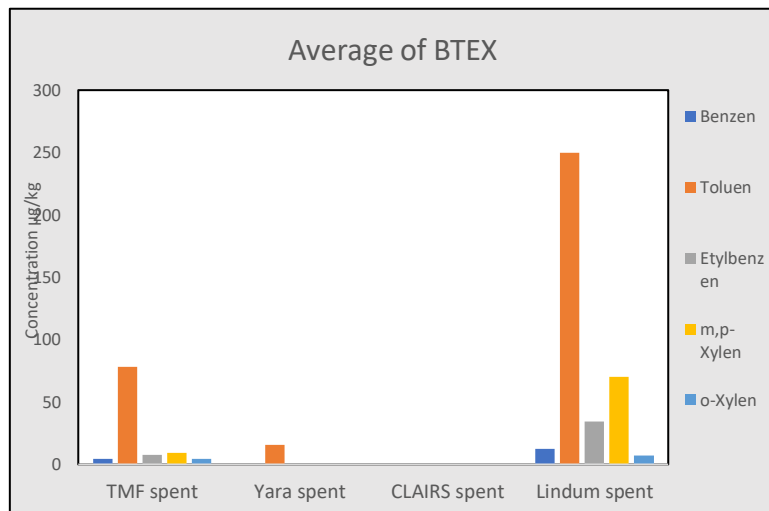
Figure 16: Average concentration of the BTEX substances: benzene, toluene, ethylbenzene, m, p xylene and o- xylene (n=3)

Toluene represents a significant portion of the BTEX concentration in the eluate, which can be explained due to the higher concentration of toluene in the AC samples (appendix B), or due to the low soil-water partition coefficient (K_{OC}) value of 178. As stated earlier, in section 2.5, the higher the K_{OC} value, the stronger the affinity to the sorbent is. Note that toluene has the second lowest K_{OC} value of the BTEX substances, which partly explains why toluene desorbs easier than the others. However, it is noteworthy that benzene, with an even lower K_{OC} of 134, does not desorb quite as much as toluene (Earl et al., 2003). The other K_{OC} values are 432, 424, 492 and 448 for ethylbenzene, o-xylene, m-xylene and p-xylene, respectively (Earl et al., 2003).

As previously mentioned, biogas and air purification plants are the common source of these spent AC materials.

The variation of BTEX concentration from the different AC materials may be explained due to the different substrate used in the biogas production (Durmusoglu et al., 2010; Lakhouit et al., 2014). TMF is mainly using food residues and manure, while Lindum mostly uses sewage sludge. However, it is unknown why sewage sludge is leaches out more BTEX than food waste and manure. The amount of the BTEX in the AC samples from water plants, NRV and KKV, was significantly lower (Appendix B).

4.3.2 Leaching of sulphur

Figure 17 shows the results for sulphur from the leaching tests. Major differences were observed between the different spent AC materials tested.
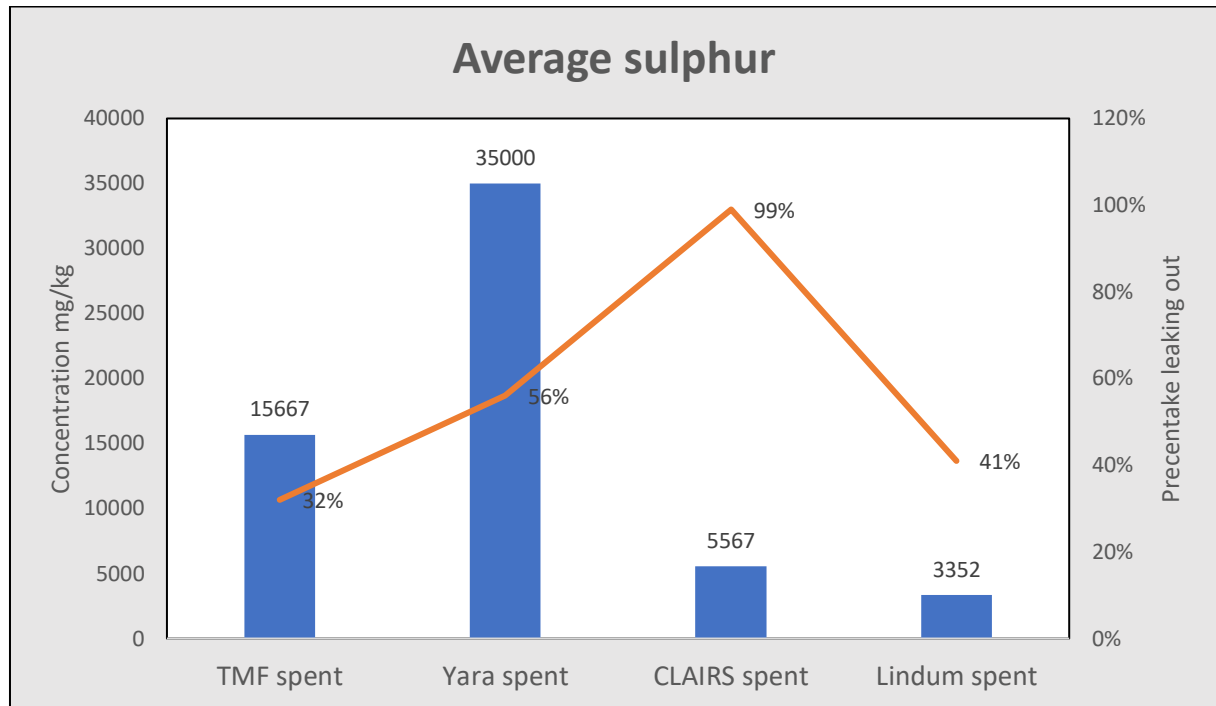
Figure 17: The blue columns show average leakage concentration of sulphur (mg/kg), and the orange line shows percentage leakage from the total sulphur concentration.

Sulphur leaching from the two different spent AC from air purification plants (CLAIRS and Yara) was substantially different (fig. 17). This difference is also evident in the total component analysis (Appendix B), in which YARA has approximately 11 times more sulphur than CLAIRS, with 62300 mg/kg and 5600 mg/kg, respectively. A similar difference can be seen between the spent AC from biogas purification (TMF and Lindum). In this case TMF contains approximately 5 times more sulphur than Lindum, with 48350 mg/kg and 8200 mg/kg, respectively.

The AC from CLAIRS, which contained the lowest amount of sulphur (see appendix B) leached 5570 mg/kg of its total 56600 mg/kg sulphur, representing approximately a 99% leakage.

Although spent AC from Yara contained the highest average sulphur concentration in the leachate (approx. 84% higher than CLAIRS), it only leached out 56% of its sulphur content (figure 17). It is unknown what caused this substantial sulphur concentration in the spent AC sample from YARA, however the difference between TMF and Lindum may be due the origin of the waste. A study from California concluded that food waste contains higher levels of sulphur due to the presence of sulphur-containing compounds compared with dairy manure and municipal solid waste (Li et al., 2019). Appendix B provides a more detailed information on the concentrations.

4.4 PFAS adsorption batch tests

Batch adsorption tests were performed with the different types of spent AC samples, with PFAS contaminated water at different L/S ratios. Where available, unused AC samples were also tested for comparative reasons. The L/S ratio started at 100, 500, 2000 and then increased to 1000, 2000, 8000, as unused AC samples were more effective in sorbing PFAS. More information about the PFAS concentration can be found in appendix G.

Figure 18 shows a considerable variation in the adsorption ability of AC depending on its application. This section discusses the different applications and how they differ from unused to spent.
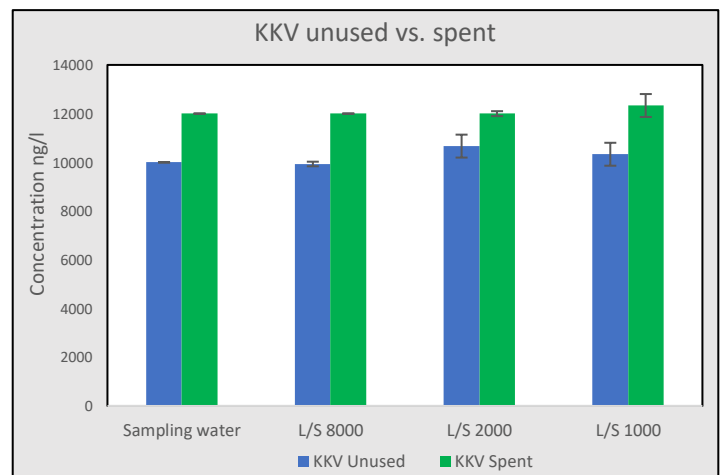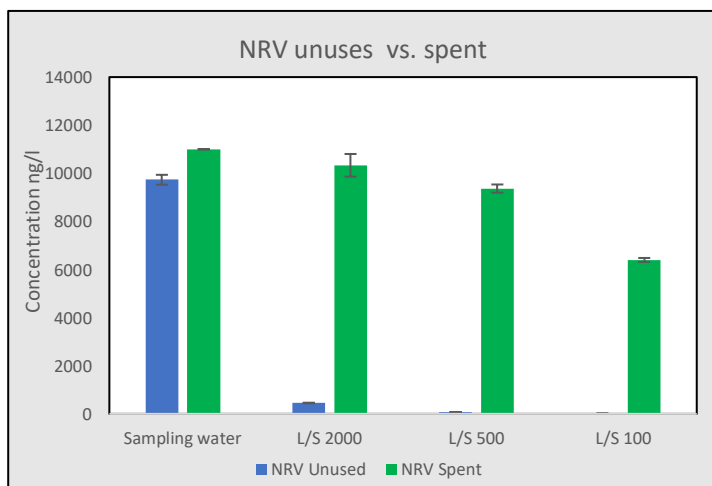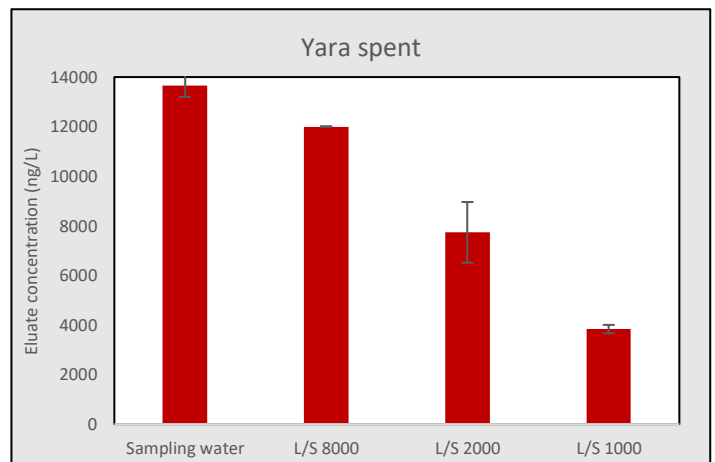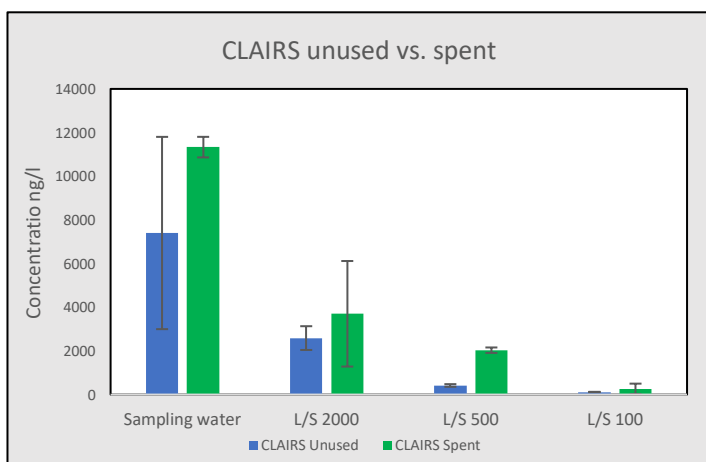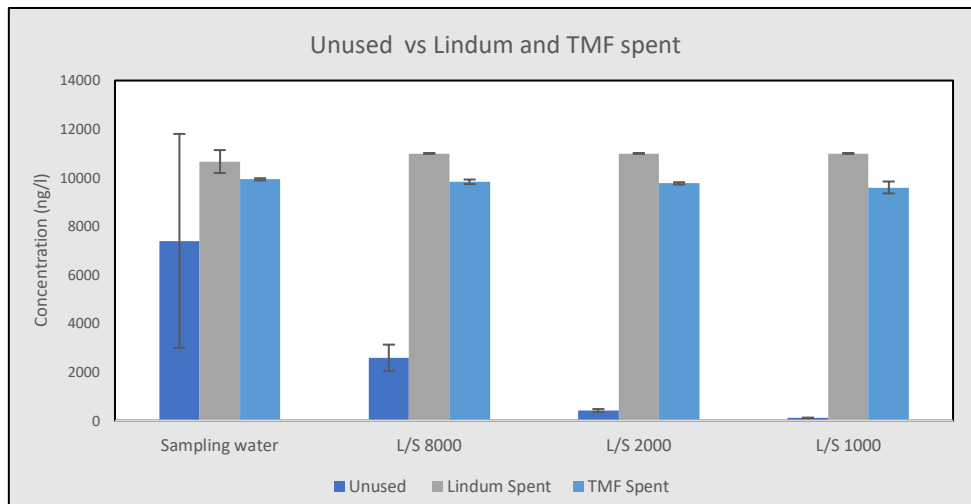
Figure 18: PFAS concentrations (ng/L) from the batch tests of the different AC samples with the different L/S ratios. This is grouped after which type of filter is used. CLAIRS, Lindum and TMF all use the same type of pellet AC filter it is unknown which type of AC Yara uses, however, it was a similar pellet to CLAIRS. KKV uses an anthracite carbon while NRV uses a GAC filter. Error bars based on the standard deviations of the PFAS concentration (n=3) are given for each column. Note that the L/S ratio differs for some of the columns.

4.4.1 AC from Wastewater treatment

There was a major difference in adsorption efficiency between the unused and the spent AC from NRV. The unused had an adsorption efficiency of 95%, while the spent AC only sorbed 6%. These are from the test which were added the lowest amounts of AC (L/S 2000). This major difference may come from the organic compounds already occupying the sorption sites in the spent AC, making it very difficult for the AC to sorb additional substances, such as PFAS. The KKV samples showed less differences between the unused and spent samples.

AC from the wastewater treatment plants, NRV and KKV, showed a major difference in their adsorption capacity at L/S 100. AC from NRV reduced the PFAS concentration in the leachate by 99% (unused) and 42% (spent), while the anthracite from KKV increased the PFAS concentration by 3,3% (unused) and 2,7% (spent). This major difference is likely due to the different natures of the sorbents. NRV utilizes granulated AC, a carbon that has gone through an activation process, which manipulates its pore and surface area, KKV, on the other hand, uses an anthracitic carbon type variant which is a natural occurrence of coal, also called black carbon. Despite its poor performance in this study, it may seem like anthracite is an ideal filter when combined with an existing sand filter or as a multi-media filtration system component, while the GAC work fine on its own (Genesis, 2019). Anthracite was used as a single filter in this experiment, so perhaps the conditions for the sorbent were not optimal.

4.4.2 AC from Biogas purification:

Unused CLAIRS showed considerable adsorption capacity for PFAS compared to the spent samples from Lindum and TMF, with 65%, -3% and 1%, respectively. These results come from the L/S 2000 tests, which is the common L/S ratios for these samples. The low levels of PFAS adsorption from Lindum and TMF make the spent samples unsuitable for PFAS filtration. The reason these three are compared, even though the CLAIRS sample stems from an air purification plant is, that all three facilities use the same type of AC.

TMF reduced the PFAS concentration by 1%, while Lindum increased the concentration by 3%. There may be two plausible reasons for the difference in PFAS adsorption between the two AC's.

The first reason could be the difference in the total concentration of PFAS already present in the material. Spent AC from Lindum contained >99% more PFAS than spent AC from TMF, 1500 µg/kg and 5 µg/kg, respectively. Thus, the potential for desorption is higher.

A second reason could be the difference in the total concentration of BTEX in the carbon. Spent AC from Lindum contained 84% more BTEX than spent AC from TMF, 1500 mg/kg and 240 mg/kg, respectively. The sorption sites for Lindum AC are probably less available for additional substances. The low adsorption efficiency from TMF, despite seemingly having more sorption sites available, may be due to other organic compounds sorbed by the AC, since TMF mostly uses food waste for biogas production.

The total concentration of PFAS and BTEX in the carbon can be seen in appendix B.

4.4.3   AC from Air purification:

The unused and spent AC from CLAIRS showed similar adsorption efficiency. The unused had a 65% adsorption efficiency, while the spent AC had 67% at the L/S 2000 ratio, which had the lowest amount of added AC.

This slight difference in adsorption efficiency between unused and spent could make the spent AC a potential PFAS sorbent for the leachate. However, there are some uncertainties about how the sulphur leaching from the AC can affect the environment (as mentioned in section 5.3.1).

Pellets from the air purification plants, CLAIRS and YARA, significantly reduced the concentration of PFAS in the sample water. Spent CLAIRS reduced the PFAS concentration by 67%, while spent AC from YARA reduced it by 43%. These are from the common L/S ratio 2000. The difference in adsorption efficiency between these AC can have several explanations.

The first explanation can be due to the different concentration of PFAS in the total analysis. AC from YARA contained 41% more PFAS than CLAIRS, with 84 µg/kg and 49 µg/kg, respectively.

A second explanation can be the concentration of sulphur. As seen in figure 7, the average sulphur concentration is higher in the AC from Yara than in CLAIRS. These concentrations can influence the sorption efficiency of the AC.

The third explanation, that is important to have in mind, is the difference in the L/S ratios. CLAIRS was measured at 2000,500 and 100, while Yara was measured at L/S 8000, 2000 and 1000.

4.5 Adsorption capacity

Although many AC samples were shown to have a good sorption efficiency for PFAS, the sorption capacity gradually increased with the decreasing amounts of sorbents.

The capacity was estimated by calculating the K_d value (equation 1). It is represented by two set of graphs, each representing the different L/S ratios. The K_d values with C_s and C_{aq} can be found in appendix H.

Figure 19 shows the K_d value for the AC samples filtered with the L/S ratios 2000, 500 and 100. These apply for the unused and spent AC samples for both CLAIRS and NRV.

The decreasing amount of AC in the sample water, increases the K_d value in all the samples. The unused and spent AC from CLAIRS showed a quite similar capacity for PFAS, with a K_d value of 3.27 and 3.37, respectively, at L/S 2000, and a K_d value of 0.25 for both at L/S 100. The unused AC from NRV had a K_d value of 4.74 at L/S 2000, while the spent sample only had 0.30 at L/S 2000 (see table 7).
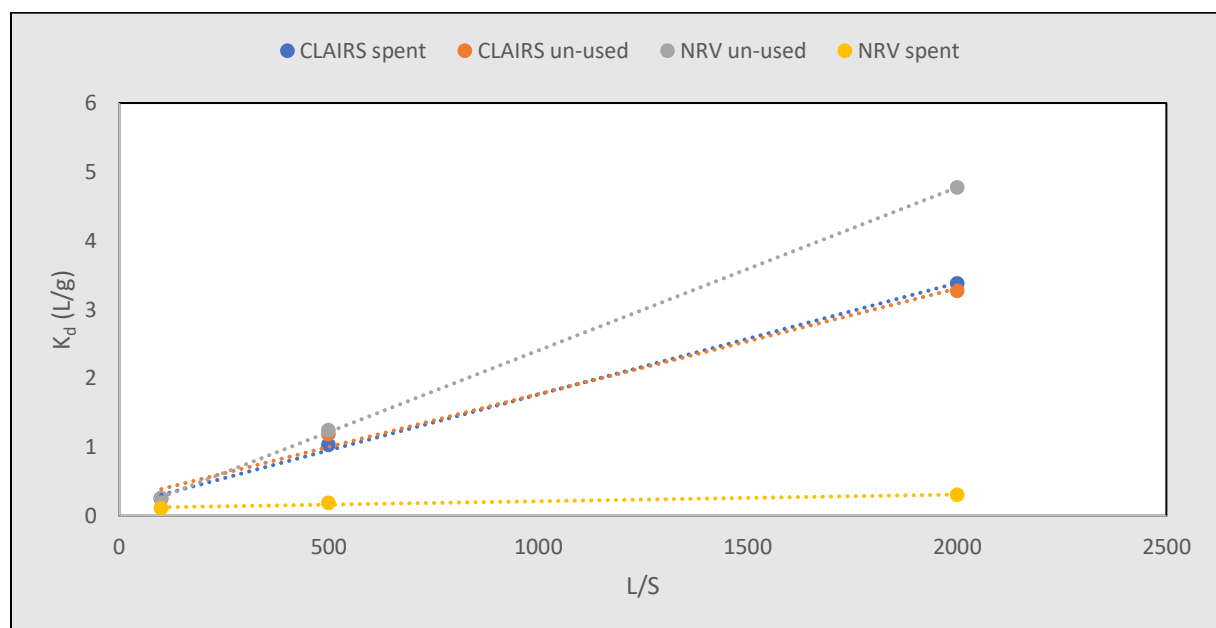
Figure 19: The K_d value for the average sum of PFAS to the various AC samples, with the AC addition of 0.2, 0,8 and 4 grams.

Table 7: Kd values for PFAS in the sample water after mixing with 0.2, 0.8 and 4 grams for the unused and spent samples from CLAIRS and NRV.

AC material	Sample water (ng/L)	L/S 2000	L/S 500	L/S 100
CLAIRS unused	7400	**3.27**	1.18	0.25
CLAIRS spent	11333	**3.37**	1.03	0.25
NRV unused	9733	**4.76**	1.24	0.25
NRV spent	11000	**0.30**	0.19	0.10

This large difference in K_d value between the unused and spent NRV samples may be due to, as stated earlier in section 4.4.1, that organic compounds occupy the sorption sites from previous uses, making no available space for new substances. The small difference in the K_d value between unused and spent CLAIRS, is suggesting that the spent CLAIRS can potentially be used as a PFAS filter.

Figure 20a shows the K_d value for the AC samples filtered with L/S ratios 8000, 2000 and 1000. These apply for the spent AC from Yara, TMF, Lindum and unused and spent anthracite from KKV. There was a major difference in the PFAS capacity between the different spent samples. Spent Yara showed a relatively good PFAS sorption capacity, compared to the other sorbents. The others showed a negative or low trend (see figure 20b and table 8). Figure 20b excludes the spent Yara sample, since it is an outlier in this group.
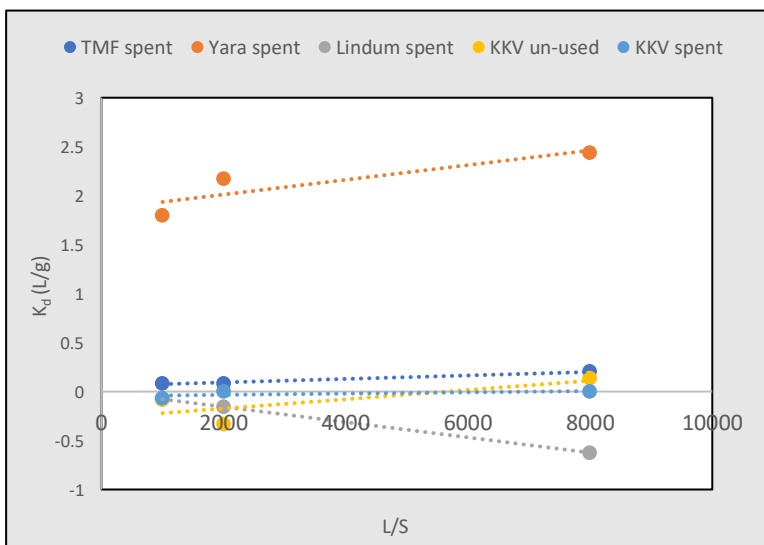
Figure 20a: Kd values for the average sum of PFAS to the various AC samples with 0.05, 0.2- and 0.4-grams AC added
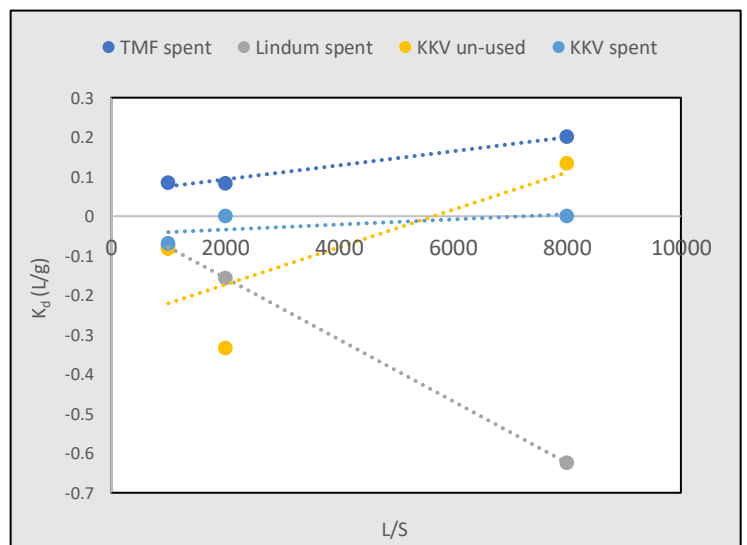
Figure 20b: Kd value for the average sum of PFAS without the spent AC from Yara

Table 8: K_d value for PFAS in the sample water after mixing with 0.05, 0.2 and 0.4 grams spent AC from Yara, TMF, Lindum, and unused and spent anthracite from KKV.

AC materials	Sample water (ng/L)	L/S 8000	L/S 2000	L/S 1000
Yara spent	13667	**2.44**	**2.13**	**1.80**
TMF spent	9933	0.20	0.08	0.08
Lindum spent	10667	-0.62	-0.16	-0.08
KKV unused	10000	0.13	-0.33	-0.08
KKV spent	12000	0	0	-0.07

The low K_d value for the spent AC from TMF and Lindum may be due to, as mentioned earlier in section 4.4.2, the high concentration of BTEX occupying the sorption sites from previous use. The leakage of PFAS from the unused and spent anthracite from KKV (figure 18) and the low K_d value, suggests that the anthracite is not suitable to be reused as a PFAS filter.

All K_d values in this study are comparatively low, e.g., in another study PFOA had a K_d value of 49, indicating that the longer-chained PFOA sorbs more easily to the AC. The majority of PFAS compounds in this study are, as mentioned in section 4.2.1, short-chained compounds. The low K_d value can be reflected in the physiochemical properties for short-chained PFASs, such as low hydrophobicity and high water solubility (Milinovic et al., 2015).
It was observed that the sorption efficiency for each gram AC decreases with the increased amount of AC is added, but figure 18 shows that a certain amount of AC is needed for an effective filtration of PFAS.

4.5.1   Substances affecting sorption

The concentration of BTEX and TOC in the AC may compete with the PFAS for sorption sites, reducing the PFAS sorption to the AC (see also section 2.5 and 2.6.1). To see exactly how BTEX and TOC correlates with the PFAS sorption affinity (expressed as K_d) for the different AC samples, the total concentration of BTEX and TOC extracted from the AC

samples, were correlated with PFAS-K_d values at L/S 2000 (figure 21a and 21b). The L/S 2000 ratio is selected, as it is the only L/S ratio tested across all types of AC samples. Generally, the range in PFAS-K_d values is very, very low, and the test is for that reason not expected to provide strong correlations. As indicated before in section 1.3, the short-chained PFAS compounds binds poorly to any material, which is confirmed by these generally low K_d-values. But the tests were conducted anyhow, to check if any clues could indicate possible competition for AC surfaces, worth studying more at a later stage. The datapoints for the BTEX PFAS-K_d values (figure 21a) are fewer (n=5) than the number of AC samples (n=9), because only 5 samples contained detectable BTEX concentrations. They are also dispersed and give no clear trend on how BTEX correlates with the PFAS-K_d.
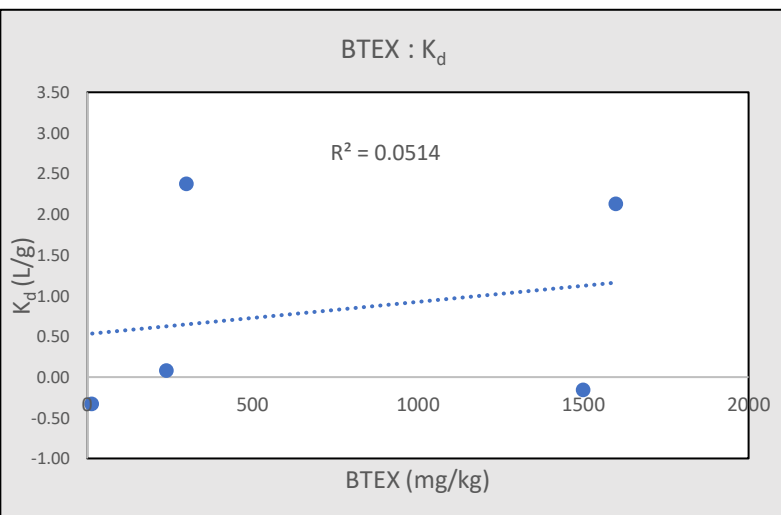
Figure 21a: K_d value and BTEX concentration
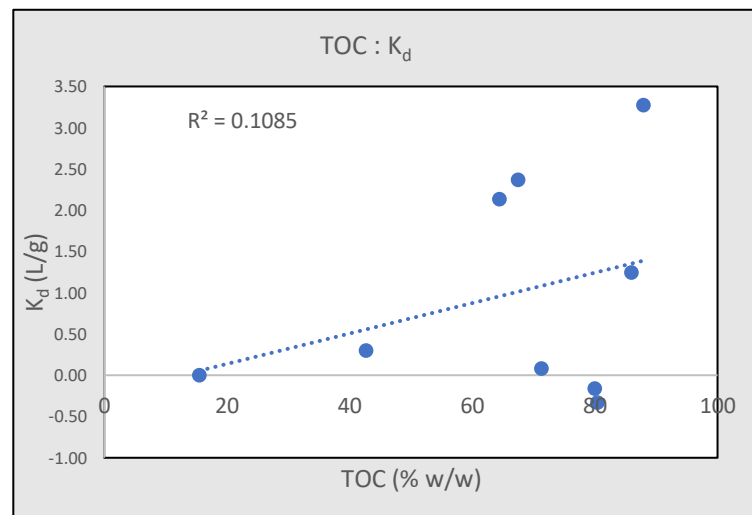
Figure 21b: K_d value and TOC percentage

Figure 21b indicates a poor, but positive, correlation between PFAS-K_d and the increasing concentration of leached TOC.

This is interesting, as such a trend correspond with other studies, reporting increasing K_d value with increasing concentration of organic matter (Ahrens, Lutz et al., 2010; Milinovic et al., 2015).

However, the result of Milanovic's 2015 study relates to long-chained PFAS, suggesting that the concentration of TOC increases the hydrophobic interactions in the hydrophobic C-F chain. As outlined in section 1.3, the hydrophobic interactions increase with the chain length. This study makes use of leachate water dominated by short-chained PFAS compounds, making the TOC concentration naturally less prominent. More information about the K_d value and BTEX and TOC concentrations can be found in appendix I.

4.6   DOC concentration

The concentration of dissolved organic compounds (DOC) in water is an important factor for the PFAS adsorption capacity of the sorbent because it competes with the sorption sites on the AC sorbents. Due to different L/S ratios used, two different graphs have been made. More information about the DOC concentration can be found in appendix J.

4.6.1   L/S 2000, 500, 100

Figure 22 shows the DOC concentration in the water after the use of AC samples with the L/S ratios 2000, 500, and 100. These involves the unused and spent AC samples for both CLAIRS and NRV.
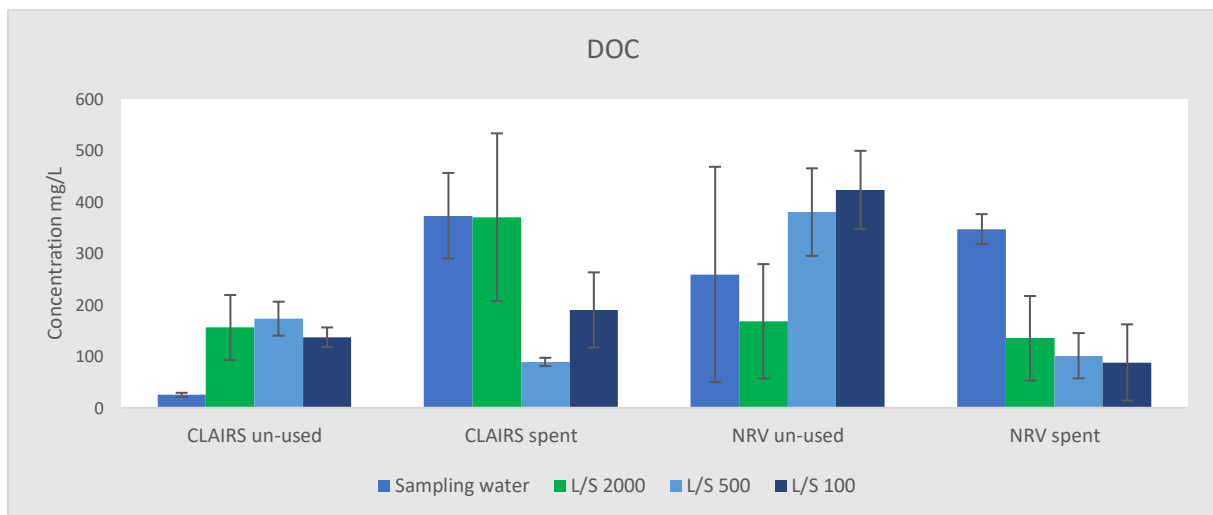
Figure 22: Overview over the DOC concentration in the water after the use of AC filter with the L/S ratio 2000, 500 and 100s have been tested. (=3)

The leaching of DOC varied between the different AC samples (figure 22).
The unused samples showed an increasing trend for the DOC concentration in the water. This may indicate that the organic matter leaches from the AC.
The spent samples showed a decreasing trend for the DOC concentration in the water. This may indicate that the DOC adsorbs into the AC.
The difference in DOC adsorption efficiency between the unused and spent may be due to the significant difference in the PFAS adsorption efficiency, as seen in figure 18.

In addition, as stated in the previous section, the unused AC samples have a considerably higher K_d value than the spent samples, leading to greater sorption affinity for PFAS.

The spent ACs sorption affinity for DOC is very unexpected, because, as mentioned in section 3.6.3, it was expected that organic matter from previous use would compete and/or block the pores in the carbon, making it difficult for "new" substances to be absorbed.

It is uncertain why the spent AC's have an affinity for DOC, but I think it may be that the original organic matter sorbed in the AC from previous use (e.g., humus) has a greater sorption affinity for" new" organic matter.

However, humus is just occurring in the NRV sample and not in CLAIRS. Spent CLAIRS shows a good adsorption efficiency for both PFAS and DOC, thus probably having available sites for both substances. The DOC concentration in the spent NRV sample has likely caused pore blockage and occupied sorption sites, thereby hindering the adsorption of PFAS, see figures 18 and 21.

4.6.2   L/S 8000, 2000, 1000

Figure 23 shows the DOC concentration in the water after the use of AC samples with the L/S ratios at 8000, 2000, and 1000. These involved spent AC from TMF, Lindum, and YARA and spent and unused anthracite from KKV.
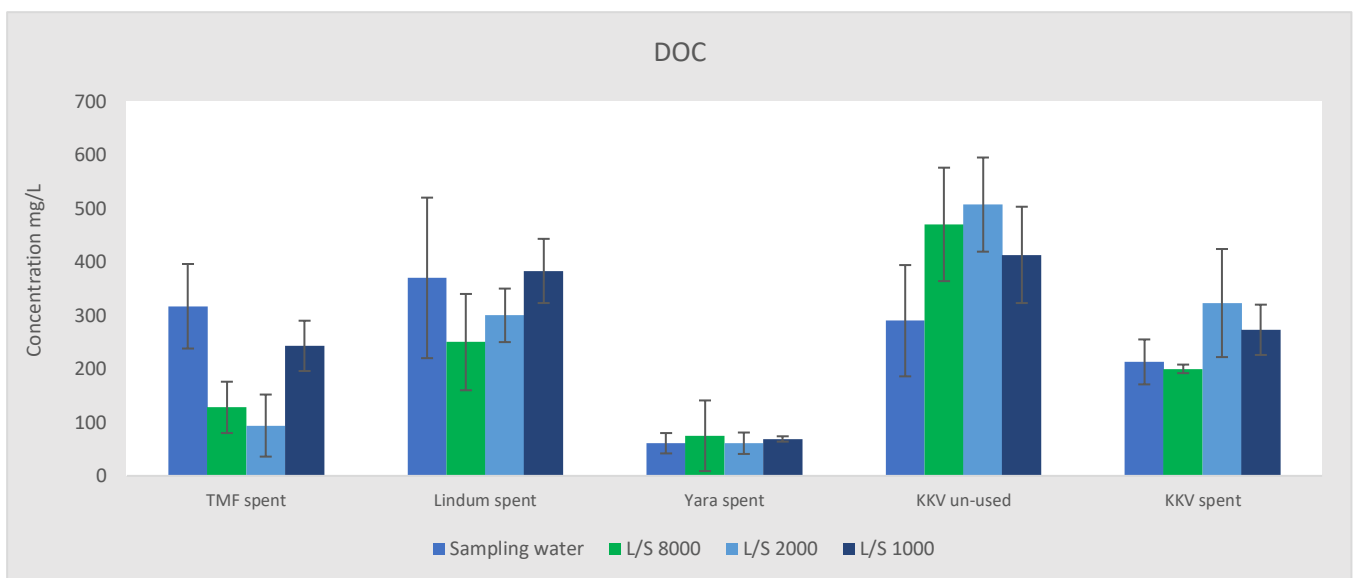
Figure 23: Overview over the DOC concentration in the water after AC filters with L/S ratio at 8000, 2000 and 1000 have been tested (n=3)

The adsorption of DOC from L/S ratios 8000, 2000, and 1000 varies significantly between the AC filters. The unused sample from KKV follows the same trend as the other unused samples, by releasing DOC to the water. There are no clear trends for the remaining spent AC samples, with some adsorbing DOC and other releasing it. It may look like the biogas samples (TMF and Lindum) follow the same adsorption trend, with some minor differences.

The different adsorption patterns may be due to the high content of PFAS and BTEX in the spent AC from Lindum (see appendix B), making it more difficult for the organic matter to be adsorbed to the AC. The concentration of organic matter in the spent TMF sample, from primarily from food waste, may be a factor that contributes to the DOC adsorption, because "old" organic matter attracts "new" organic matter, as speculated earlier.

KKV samples are releasing DOC to the water, and one possible explanation could be that anthracite doesn't adsorb DOC well.

The unused and spent AC samples showed different DOC trends. The unused samples showed a clear leaching of organic matter from all samples. In contrast, the spent AC samples demonstrated a greater degree of DOC adsorption. The higher DOC adsorption from the spent samples was unexpected, but one explanation could be that organic matters have some sort of an affinity for each other. Although the spent samples adsorb a lot more DOC, it does not affect the adsorption of PFAS, as can be seen in figure 18, for the spent CLAIRS and Yara. Early on in this study it was evident that the spent CLAIRS samples had a good PFAS affinity and DOC adsorption rate, and it was decided to further test this sample. The next chapter is going to explore this further.

4.7 *In-situ* testing with spent CLAIRS AC

The field test using the spent AC samples from CLAIRS should ideally be conducted with the same leachate used in the laboratory test (i.e., from cell 1-2 of the Lindum Oredalen landfill) since this is a typical firefighting contaminated water sample that contains PFAS. Unfortunately, it was not possible to separate the leachate from cell 3, which had a high concentration of organic matter, so the spent CLAIRS AC had to be tested with a mixture of the two leachates.

4.7.1   PFAS concentration variation

As mentioned in section 3.4.2, a batch test was completed in the laboratory to show the effect of the different leachate types with regards to PFAS adsorption effectiveness of CLAIRS spent AC. There was a 64% variation in the sum PFAS concentration in the leachates from cell 1-2 and 3 with 11333 ng/L and 4067 ng/L, respectively (figure 24). Increasing the AC concentration in the water samples, resulted in a major difference in the PFAS adsorption efficiency. Cell 1-2 had an increasing adsorption efficiency with increasing AC concentration, by 67% at L/S 2000 and 97% at L/S 100, while cell 3 had an increased efficiency from 15 to 78% at the same L/S ratios.

These major variations in PFAS concentration in the water sample are due to the different source materials in the cells. Cell 1-2 contains PFAS contaminated masses from fire-fighting areas from airports, while cell 3 contains masses from drill cuttings, which contain a lot of organic matter. The organic matter from the drill cuttings may occupy the sorption sites of the AC, making it more difficult for the PFAS to be attached to the surface, which is reflected in figure 24. See appendix K for concentrations and standard deviation.
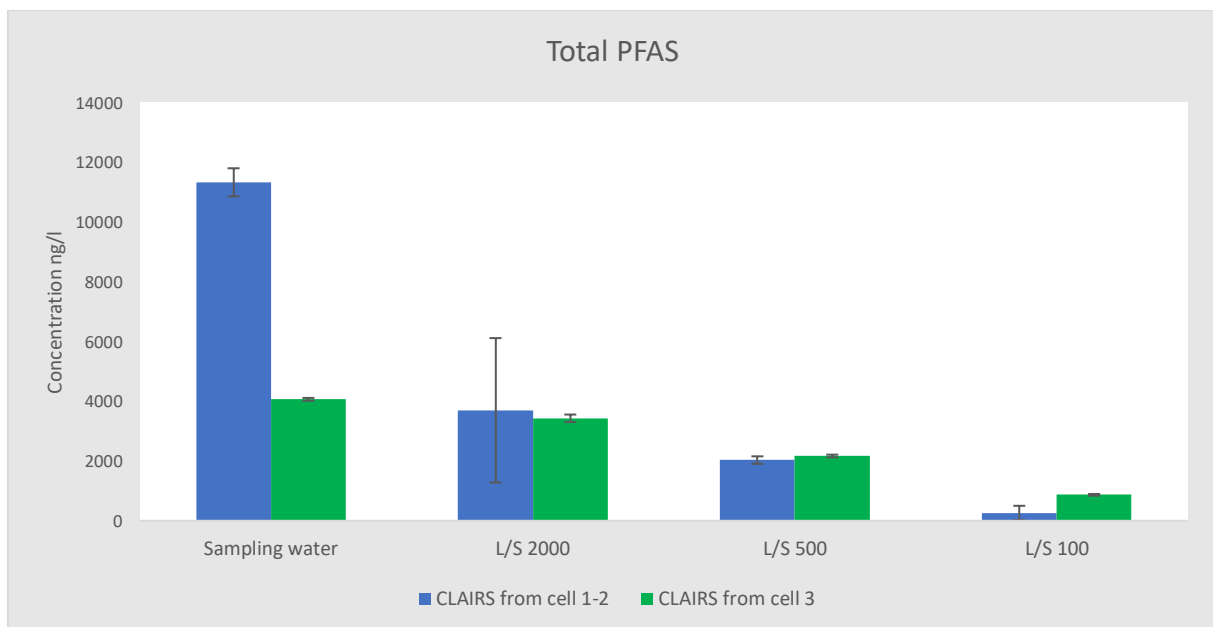
Figure 24: Total PFAS concentration between the cells. At L/S 100, 97% sum PFAS was removed from the cell 1-2 leachate, while only 78 % was removed from the cell 3 leachate. Error bares are included (n=3).

4.7.2 DOC concentration variation

The major concentration of organic matter from cell 3 is well reflected in figure 25, which shows the difference in concentration of DOC between the cells.

The sampling water shows a 57% variation between cells 1-2 and 3 with 373 ng/L and 884 ng/L, relatively. By increasing the concentration of AC in cell 3, it increased the concentration of DOC in the water by 11 % at L/S 2000 and 15% by L/S 100, relative to the sampling water. Another pattern is expressed in cell 1-2, where increasing the concentration of AC from L/S 2000 to 100, decreased the DOC concentration from 0,8 to 49%, relative to the sampling water. See appendix K for concentration.
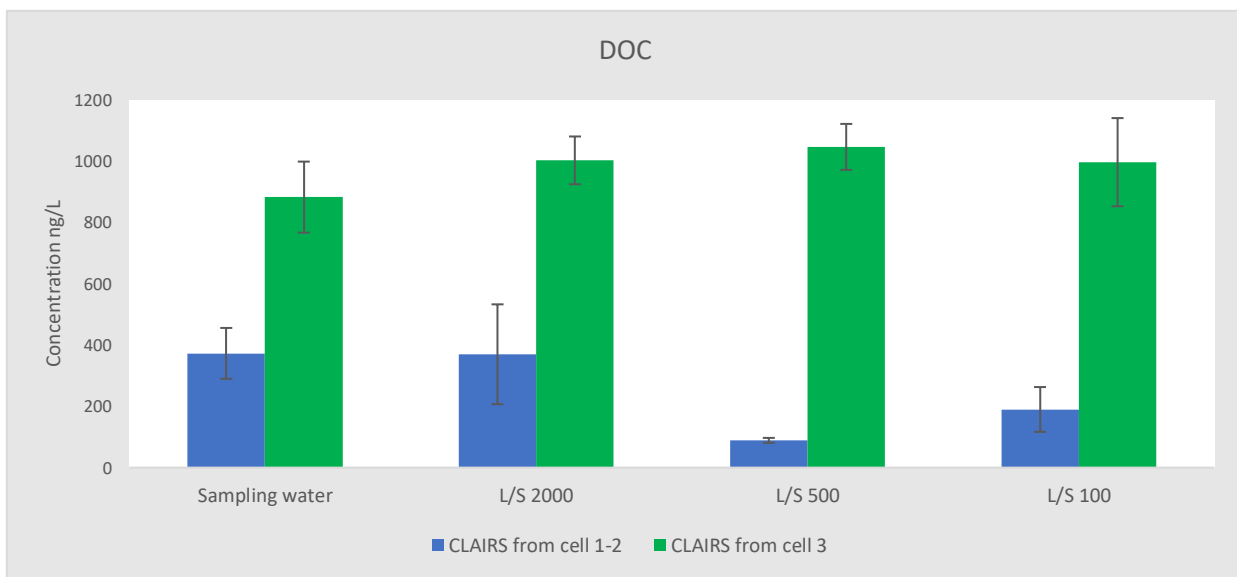
Figure 25: Total DOC concentration in the leachate from the different cells, with increasing concentration of AC. With error bars (n=3).

Comparing the results from cells 1-2 and 3 show how the high concentration of TOC/DOC in cell 3 are affecting the sorbents' ability to adsorb PFAS. The high concentration of DOC in the water from cell 3 reduces the PFAS sorption of the spent AC significantly.

It is, however, uncertain how high a DOC concentration is needed to affect the PFAS adsorption rate significantly, but this could be an area for further studies and research.

4.7.3 *In-situ* testing results

It was decided that the spent AC from CLAIRS was the most suitable for the *in-situ* experiment, due to the promising results with the PFAS sorption and minimal leaching of BTEX. As previously mentioned, it was planned to be used with the leachate from PFAS cell 1-2 at Lindum's Oredalen landfill, however due to unavoidable circumstances, the organic rich leachate from cell 3 had to be filtered simultaneously. Initially, attempts were made to limit the leachate from cell 3. The cell was covered with a water-repellent membrane to reduce the leaching of the cell, and to eventually reduce the concentration of TOC in the leachate. This attempt, however, was not successful, and TOC rich water continued to leach from cell 3.

The PFAS and TOC concentration were measured in sampling water from sampling site #1-3 (figure 7) and the result are seen in figure 26 and 27.

Figure 26 shows TOC concentrations from the 5 weeks of testing. Sample sites #1 and #2a both analyse the leachate from cell 3, #1 was from before the leachate flowed through a catchpit with unused CLAIRS AC, and #2a was from right after. Sample site #2b measures the leachate from cell 1-2 before filtration. The leachate from both cells combines after the sampling sites #2a and #2b, then flow together through a catchpit with spent CLAIRS AC, before being measured at sample site #3 (see figure 7). A major concentration of TOC is showed at the sample sites #1 and# 2a, at the first week with 750 mg/L and 670 mg/L, relatively, and this remained relatively constant.

By the end of the five weeks the TOC concentration had slightly decreased, and this small reduction may reflect the adsorption of PFAS by the AC. Sample site #2b showed marginal concentrations of TOC, while #3 showed a TOC concentration which is roughly halfway between the concentrations in #2a and #2b, which is not surprising, since the sample water in #3 is a mixture of #2a and #2b.
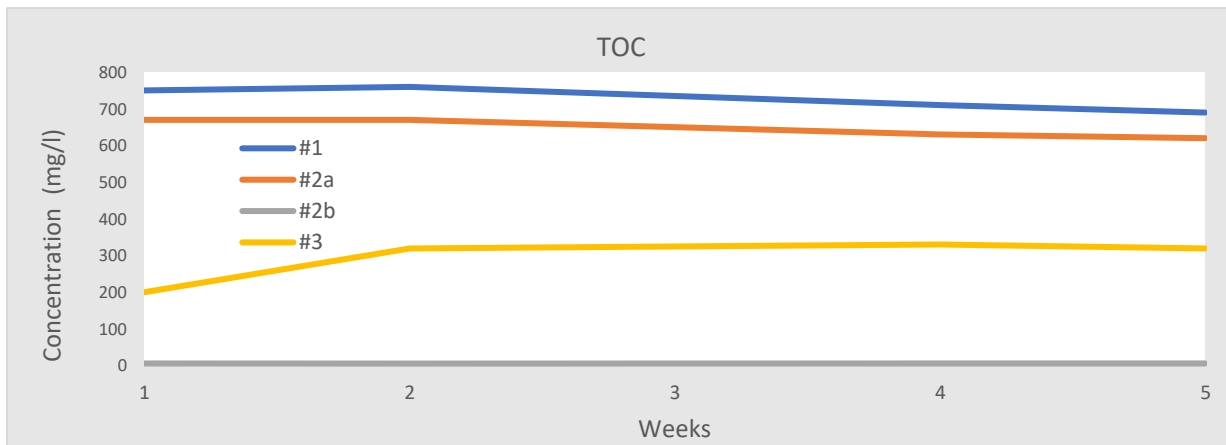
Figure 26: TOC concentration during the 5 weeks of the in-situ experiment. Sample site #1 shows cell 3 leachate before filtration by unused CLAIRS AC, and #2a shows the concentration after the filtration. Sample site #2a shows the concentration from cell 1-2 before combination with the leachate from cell 3 and the filtration by spent CLAIRS AC. Sample site #3 shows the combined leachate of cell 1-2 and 3, after it's been filtrated by spent CLAIRS AC.

Figure 27 shows the PFAS concentrations in the leachate. It varied significantly between the different sample sites. High concentrations of PFAS was shown in cell 1-2 (# 2b), with a concentration between 13000 ng/L and 11000 ng/L.

PFAS concentration from cell 3 (# 1) showed an unusually high concentration in week 1 before returning to its relative average values at 3000 ng/L, and then decrease to 500 ng/L in the last week. It is unknown why there were such high concentrations of PFAS during the first week and low during the last week. The PFAS concentration in #2a remained stable at 17-1900 ng/L throughout the testing period. The catch-pit with unused AC might have had a stabilizing effect on the PFAS concentration, but the reason remains uncertain.

Sample site #3 showed an effective filtering PFAS compounds from the leachate in the first two weeks, however it could be seen that after 4-5 weeks the effectivity reduced resonably quickly. More information about the TOC and PFAS concentration can be found in appendix L.
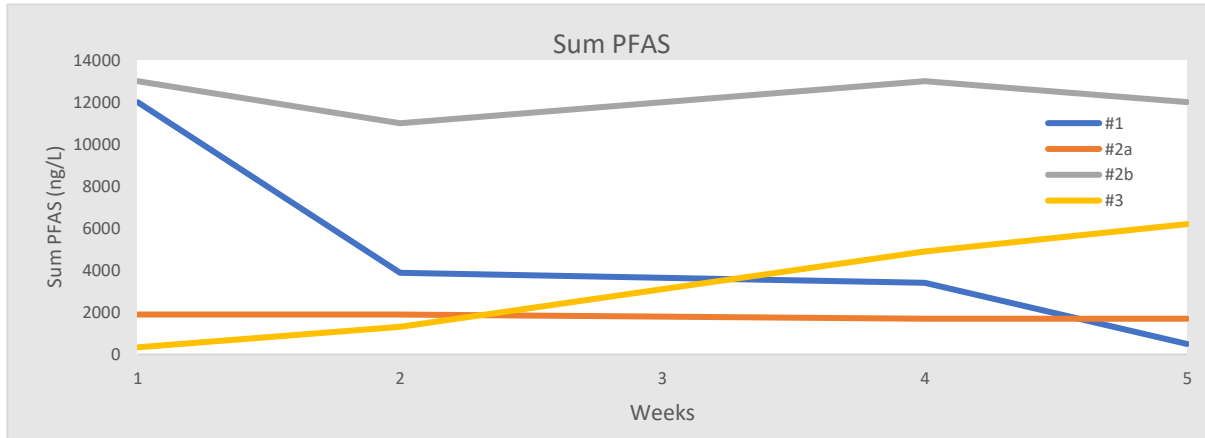
Figure 27: PFAS during the 5 weeks of the in-situ experiment. Sample site #1 shows cell 3 leachate before filtration by unused CLAIRS AC, and #2a shows the concentration after the filtration. Sample site #2a shows the concentration from cell 1-2 before combination with the leachate from cell 3 and the filtration by spent CLAIRS AC. Sample site #3 shows the combined leachate of cell 1-2 and 3, after it has been filtrated by spent CLAIRS AC.

While it may be possible that the efficiency reduction was due to the AC being previously used in air purification, it was hypothesized that the large amount of TOC in the cell 3 leachate stole the sorption sites for the PFAS compounds. Measures were taken to reduce the TOC concentrations from cell 3, but they had less than desired effect.

The *in-situ* experiment was therefore temporarily terminated and planned for a later date when testing with only leachate from cell 1-2 was possible. This was unfortunately after the time scope of this study. It is difficult to speculate on the mechanisms taking place here, as the *in-situ* experiment was not designed specifically to investigate this complexity. In the *in-situ* experiment, the TOC seems to have outcompeted the PFAS, whereas in the batch test, the TOC seems to increase the AC´s affinity for PFAS (figure 21b). This is a matter for more detailed studies, certainly as the PFAS chain-length adds to the complexity.

4.8   Sorbents suitability for PFAS sorption

This chapter summarises the findings in the result and discussion part of this study. The primary focus is to value the suitability of the different spent AC filters for PFAS filtration.

4.8.1   Air purification spent AC

The spent AC samples from air purification (CLAIRS and Yara) showed promising results in the PFAS sorption, with a 67% (CLAIRS) and 43% (Yara) PFAS adsorption efficiency, relative to the control water at the L/S 2000 ratio. They showed minimal leaching of BTEX,

but had a high rate of sulphur leakage, 99% (CLAIRS) and 56% (Yara) (figure 17). The sulphur leaching may not be a problem, because the amounts in the AC samples were small, 5.6 g/kg DM (CLAIRS) and 62.3 g/kg DM (Yara), with the respect to the amount of water they may filter at a commercial scale (figure 12). It is unknown which form of sulphur is leaching out, and harmful forms cannot be ruled out, thus making the sulphur in the spent AC samples an area for future research and studies.

4.8.2 Biogas spent AC

The spent AC samples from biogas purification (TMF and Lindum) did not show promising results as a potential PFAS sorbent. With only a 3% adsorption efficiency from spent TMF and a 3% PFAS leakage from the spent AC from Lindum.

4.8.3 Water treatment spent AC

The spent AC and anthracite samples from water treatment (NRV and KKV) also showed less than promising results as a potential PFAS sorbent, with only 6% adsorption efficiency from the spent AC from NRV and a negligible adsorption efficiency form the spent anthracite from KKV. While samples from other wastewater treatment plants should be tested, it is presumed that the high organic matter taken out of the water at these plants will reduce the likelihood of their potential use as PFAS filter.

5 Conclusion and future perspective

This study aimed to identify the potential of spent AC from three different applications (water, air- and biogas purification) for being reused as a filter for PFAS-contaminated water. The study provided both insufficient and sufficient data to verify the hypotheses defined in section 2.2.

The sorption efficiency of the spent AC samples from the three different applications varied significantly.
Samples from water treatment and biogas purification showed an insignificant PFAS adsorption efficiency compared to the AC samples from air purification plants. Neither spent AC samples from water treatment and biogas purification were suitable for reusing as PFAS sorbents, due to other substances like BTEX and organic matter occupying the sorbent's sorption sites, thus verifying the sorbent hypothesis (i). As a result of these factors, spent AC is less effective than the unused AC, making hypothesis (ii) valid.

Whether the K_{OC} value explains the desorption of the different contaminants from the spent AC, as suggested in hypothesis (iii), could not be fully verified. E.g. toluene had a higher desorption rate than benzene, even though toluene has a higher K_{OC} value than benzene, 173 compared to 134.

This study showed that spent AC from air purification plants had the best potential to be reused as a PFAS sorbent, with an adsorption efficiency of 67-97%, depending on the amount of sorbent added. However, due to the significant amount of sulphur leakage from the AC, further testing on other air filtration samples is needed. Focusing on PFAS adsorption and sulphur leaching in large-scale *in-situ* tests are also needed.
The concentration of TOC had a profound effect on this study, showing how much it can affect PFAS, and would also be worthy of further study.

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Appendix overview

Appendix A: Description of the method, instruments, and conditions.

DIN 38407-42 is a German standard method for the examination of water, waste and sludge, part 42 consisting of determination of selected polyfluorinated compounds in water. This method are using high performance liquid chromatography and mass spectrometric detection (HPLC/MS-MS) after an solid-liquid extraction (DIN, 2011), with some modification from United Nations Environment Programme (UNEP) (Weiss et al., 2015). NS-EN 1484 is an Norwegian translated European standard for Water analysis, guidelines for the determination of total organic carbon (TOC) and dissolved organic carbon (DOC) (Standard Norge, 1997). SS-EN-ISO 10301 is a standard that specifies on two methods for the determination of highly volatile halogenated hydrocarbons using gas chromatography (SIS, 1997). HS-GC/MS stands for head space gas chromatography/mass spectrometry

Apenndix B Contamination content in the various AC samples

Table B1.: Content of various contaminations in the different sorbents from the different companies. These contaminations were PFAS (µg/kg DM), metals (mg/kg), BTEX (mg/kg), PAH (mg/kg), PCB (mg/kg) and TOC (w/w).

	Sample reference	CLAIRS unused	CLAIRS spent	Yara spent	TMF spent	Lindum spent	NRV unused	NRV Spent	KKV unused	KKV spent
PFAS										
4:2 Fluortelomersulfonat (FTS)	µg/kg TS	<0,20	<0,10	<0,10		<0,10	<0,10	<0,11	N.d	N.d
6:2 Fluortelomer sulfonat (FTS) (H4PFOS)	µg/kg TS	<0,20	<0,10	4	2,2	15	<0,10	0,31	N.d	N.d
8:2 Fluortelomersulfonat (FTS)	µg/kg TS	<0,20		1,4	<0,10	0,7	<0,11	<0,11	N.d	N.d
7H-Dodekafluorheptansyre (HPFHpA)	µg/kg TS	<0,20	<0,10	<0,10		<0,10	<0,10	<0,11	N.d	N.d
Perfluor -3,7-dimetyloktansyre (PF-3,7-DMOA)	µg/kg TS	<1	<0,50	<0,50	<0,50	<0,50	<0,50	<0,55	N.d	N.d
Perfluordekansyre (PFDeA)	µg/kg TS	<0,20	1,9	1,7	0,16	15	<0,10	2	N.d	N.d
Perfluorbutansyre (PFBA)	µg/kg TS	<0,20	19	10	<0,10	280	<0,10	0,49	N.d	N.d
Perfluorbutansulfonat (PFBS)	µg/kg TS	<0,20	8,4	1,3	<0,10	<0,10	<0,10	0,35	N.d	N.d
Perfluordodekansyre (PFDoA)	µg/kg TS	<0,20	<0,10	0,3	<0,10	1	<0,10	<0,11	N.d	N.d
Perfluortridekansyre (PFTrA)	µg/kg TS	<0,20		0,34	<0,10	<0,10	<0,11	<0,11	N.d	N.d
Perfluordekansulfonsyre (PFDS)	µg/kg TS	<0,20	<0,10	<0,10	<0,10	<0,10	<0,10	<0,11	N.d	N.d
Perfluorheptansyre (PFHpA)	µg/kg TS	<0,20	0,87	3,1	<0,10	11	<0,10	1,8	N.d	N.d
Perfluorheptansulfonat (PFHpS)	µg/kg TS	<0,20	<0,10	<0,10	<0,10	<0,10	<0,10	0,18	N.d	N.d

Perfluorheksansyre (PFHxA)	µg/kg TS	<0,20	7,8	50		930	<0,10	0,44	N.d	N.d
Perfluorheksansulfonat (PFHxS)	µg/kg TS	<0,20	0,55	<0,10	<0,10	<0,10	<0,10	1,3	N.d	N.d
Perfluornonansyre (PFNA)	µg/kg TS	<0,20	0,22	1	<0,10	2,1	<0,10	5,5	N.d	N.d
Perfluoroktansyre (PFOA)	µg/kg TS	<0,10	6,2	7,2	0,84	230	<0,050	6,4	N.d	N.d
Perfluoroktylsulfonat (PFOS)	µg/kg TS	<0,10	1,4	0,14	<0,050	<0,050	0,22	11	N.d	N.d
Perfluoroktansulfonamid (PFOSA)	µg/kg TS	<0,20	0,1	0,59	<0,10	<0,10	<0,10	0,18	N.d	N.d
Perfluorpentansyre (PFPeA)	µg/kg TS	<0,20	0,71	0,82	<0,10	31	<0,10	<0,11	N.d	N.d
Perfluortetradekansyre (PFTA)	µg/kg TS	<0,20		<0,10	<0,10	0,25	<0,11	<0,11	N.d	N.d
Perfluorundekansyre (PFUnA)	µg/kg TS	<0,20	<0,10	0,57	<0,10	0,17	<0,10	1	N.d	N.d
N-etylperfluoroktansulfonamid-HAc (EtFOSAA)	µg/kg TS	<0,20	<0,10	<0,10	<0,10	<0,10	<0,11	<0,11	N.d	N.d
N-metylperfluoroktansulfonamid-HAc (MeFOSAA)	µg/kg TS	<0,20	<0,10	<0,10	<0,10	<0,10	<0,10	<0,11	N.d	N.d
N-metylperfluoroktansulfonamidetanol (MeFOSE)	µg/kg TS	<0,20		<0,10	<0,10	<0,10	<0,11	<0,11	N.d	N.d
N-metylperfluoroktansulfonamid (MeFOSA)	µg/kg TS	<0,40	<0,20	0,33	<0,20	<0,20	<0,20	<0,22	N.d	N.d
Perfluoroktansulfonamid-HAc (FOSAA)	µg/kg TS	<0,20	<0,10	<0,10	<0,10	<0,10	<0,11	<0,11	N.d	N.d

Perfluorheksadekansyre (PFHxDA)	µg/kg TS	<1		<0,50	<0,50	<0,50			N.d	N.d
N-etylperfluoroktansulfonamidetanol (EtFOSE)	µg/kg TS	<0,20		<0,10	<0,10	<0,10	<0,11	<0,11	N.d	N.d
N-etylperfluoroktansulfonamid (EtFOSA)	µg/kg TS	<0,40		<0,20		<0,20	<0,22	<0,22	N.d	N.d
Σ PFAS	µg/kg TS	0	49	84	5	1500	2,1	31	N.d	N.d
METALS										
Chlorine (Cl2)	mg/kg	73	<50	<50	<50	<50	69	250	170	460
Sulphur (S)	mg/kg	250	5600	62300	48350	8200				
Arsenic (As)	mg/kg	2	3,6	3,8	6	4,3	5	6,3	0,9	6,4
Lead (Pb)	mg/kg	<2	9	3	<2	< 2	5	< 2	5	35
Cadmium (Cd)	mg/kg	< 0,2	<0,2	< 0,2	<0,2	< 0,2	< 0,2	0,9	< 0,2	< 0,2
Chromium (Cr)	mg/kg	6	10	11	11	13	16	21	3	21
Copper (Cu)	mg/kg	6	12	9	12	10	143	154	4	21
Nickel (Ni)	mg/kg	23	15	22	16	20	26	59	6	49
Zinc (Zn)	mg/kg	4	25	4	16	14	17	167	10	75
Mercury (Hg)	mg/kg	< 0,07		< 0,07		< 0,07	< 0,07	< 0,07	0,46	0,27
Calcium (Ca)	mg/kg	5900	8440	7330	3720	3580	697	6880	2150	35000
Potassium (K)	mg/kg	235	324	388	1660	3280	867	786	490	1950
Magnesium Mg)	mg/kg	638	1190	648	623	561	83	251	247	1150

Sodium (Na)	mg/kg	3570	1400	2020	811	869	434	400	340	762

BTEX										
Benzene	mg/kg	< 2	10	11	4	9	<2	<2	3	< 2
Toluene	mg/kg	< 2	220	1500	85	540	<2	<2	8	< 2
Ethylbenzene	mg/kg	< 2	13	18	27	100	<2	<2	< 2	< 2
m-/-p xylene	mg/kg	< 2	44	42	75	740	<2	<2	4	< 2
o-xylene	mg/kg	< 2	15	14	44	110	<2	<2	< 2	< 2
BTEX sum	mg/kg	< 2	300	1600	240	1500	< 2	< 2	15	<2

PAH										
Naphthalene	mg/kg	0,8	2,2	< 1,0	5,6	< 1,0	0,2	2,4	25	0,3
Acenaphthylene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	0,2	< 0,1	< 0,1
Acenaphthene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	<0,1	<0,1	2,7	< 0,1
Fluorene	mg/kg	<0,1	<0,1	< 1,0	1,5	< 1,0	< 0,1	0,2	7,9	< 0,1
Phenanthrene	mg/kg	0,3	0,2	< 1,0	<0,1	< 1,0	< 0,1	1,6	34	< 0,1
Anthracene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	0,1	< 0,1
Fluoranthene	mg/kg	0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	0,5	3	< 0,1
Pyrene	mg/kg	<0,1	<0,1	< 1,0	0,1	< 1,0	< 0,1	0,2	3,5	< 0,1
Benz(a)anthracene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	0,9	< 0,1
Chrysene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	6,3	< 0,1
Benzo(b)fluoranthene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	5,7	< 0,1

Benzo(k)fluoranthene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	0,5	< 0,1
Benzo(a)pyrene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	1	< 0,1
Indeno(1,2,3-cd) pyrene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	0,6	< 0,1
Dibenz (a, h) anthracene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	0,7	< 0,1
Benzo (g, h, i) perylene	mg/kg	<0,1	<0,1	< 1,0	<0,1	< 1,0	< 0,1	< 0,1	1,4	< 0,1
Total 16 EPA-PAH excl. LOQ	mg/kg	1,2	2,4	<1,0	7,2	<1,0	0,2	5,1	93,3	0,3
PCB										
PCB 28	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 52	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 101	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 118	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 138	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 153	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
PCB 180	mg/kg	<0,5	<0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	<0,5	<0,5
Total 6 ndl-PCB (lower bound)	mg/kg	< 0,5	< 0,5	<0,5	< 0,5	<0,5	<0,5	< 0,5	< 0,5	< 0,5
PCB (7) Sum	mg/kg	< 0,5	< 0,5	0,5	< 0,5	<05	< 0,5	<0,5	< 0,5	< 0,5
TOC										
TOC (total organic carbon - fossil fuels)	% (w/w)	87,9	67,5	64,5	71,3	80	86	42,7	80,5	15,5

Appendix C: Concentration of PAH, PCB, and heavy metals for the different samples

Table C1.: Quality standards for PAH 16, PCB7 and heavy metals (mg/L) in contaminated soil in Norway. Heavy metals is left without values and only normative values, because all of the metals were below the normative values. (Norwegian Environmental Agency, 2020).

Sample	PAH (mg/kg)	PCB (mg/kg)	Heavy metals (mg/kg)
CLAIRS spent	2,4	N. d	
Yara	N. d	N. d	
TMF	7,2	N. d	
Lindum	N. d	N. d	
NRV spent	5,1	N. d	
KKV spent	0,3	N. d	

Appendix D: PFAS concentration in sample water used for the various batch tests

Table D1: Concentrations of the various PFAS compounds with standard deviation in the sample water (ng/L) for the different AC samples. The results shown are averages from triplicate tests.

PFAS	Chain-length	AC								
		CLAIRS		NRV		TMF	Yara	Lindum	KKV	
		Unused	Spent	Unused	Spent	Spent	Spent	Spent	Unused	Spent
PFPeA	5 (short-chain)	7133 ± 499	7967 ± 170	6567 ± 249	7167 ± 125	6833 ± 47	8933 ± 94	7600 ± 294	7233± 3394	9133 ± 47
PFBA	4 (short-chain)	1967 ± 47	1700	1500	1900 ± 82	1500	2300	1433 ± 47	1433 ± 47	1600
PFHxA	6 (short-chain)	1300	1267 ± 47	1200	1333 ± 47	1100	1433 ± 5	1200	1133 ± 47	1133 ± 47
PFBS	4 (short-chain)	323 ± 9	260 ± 8	260 ± 8	317 ± 12	293 ± 5	400	297 ± 5	290 ± 14	303 ± 5
PFHxS	6 (long-chain)	103 ± 5	107 ± 5	98 ± 11	98 ± 11	82 ± 4	143 ± 5	82 ± 8	80 ± 4	84 ± 3
PFHpA	7 (long-chain)	69 ± 2	71 ± 5	65 ± 4	66 ± 42	44 ± 7	102 ± 6	51 ± 5	43 ± 1	43 ± 3
PFPeS	5 (short-chain)	54 ± 2	57 ± 2	51 ± 1	53 ± 1	42 ± 1	71 ± 3	45 ± 2	43± 1	39 ± 6
6:2 FTS	8 (long-chain)	49 ± 8	49 ± 1	37 ± 7	42 ± 5	38 ± 2	61 ± 19	31 + 16	53 ± 2	38 ± 2
PFOA	8 (long-chain)	24 ± 4	19 ± 1	19 ± 4	22 ± 2	17 ± 1	37 ± 2	11 ± 1	10	-
PFOS	8 (long-chain)	17 ± 1	-	-	-	17	36 ± 2	10	-	-
Total		11039 ± 577	11490 ± 236	9797 ± 276	10998 ± 327	9966 ± 67	13516 ± 136	10760 ± 378	10317 ± 3511	12373 ± 113

Appendix E: Average BTEX and sulphur concentration in the leachates

Table E1: *The average concentration for BTEX (μg/L) and sulphur (mg/L) in the sample water, after the leaching tests with L/S 10. The results shown are averages from triplicate tests.*

	BTEX							Sulphur
	Benzene	Toluene	Ethylbenzene	M, p-Xylene	o-Xylene	Xylene (sum)	Σ BTEX	
Sample reference	μg/L	μg/L	μg/L	μg/L	μg/L	μg/L	μg/L	mg/L
TMF	4.57	78.00	7.73	9.10	4.27	13	10	15667
Yara	N. d	25.67	N. d	N. d	N. d	N. d	2	35000
CLAIRS	N. d	N. d	N. d	N. d	N. d	N. d	N. d	5567
Lindum	13.67	250.00	34.33	70.33	7.20	77.33	37	3352

Appendix F: BTEX and sulphur leaching (in %)

The next two tables indicate the BTEX (table F1) and sulphur (table F2) leakage from 180g carbon in a 18000ml water bottle (L/S 10) and the conversion to 1 kg to determine the extent of the leakage.

Table F1: An overview over the leakage percentage of BTEX from 1 kg AC compared to the total content in the AC sample

BTEX (L/S 10)							
TMF	240000 µg/kg	YARA	1600000 µg/kg	CLAIRS	300000 µg/kg	Lindum	1500000 µg/kg
180g	18.66 ug	180g	2,8 ug	180	N. d	180g	67.41 ug
1 kg	103.66 ug	1 kg	15.55 ug	1 kg	N. d	1 kg	374.5 ug
0.004% leaks out		0% leaks out		N. d		0.03% leaks out	

Table F2: An overview over the leakage percentage of sulphur from 1 kg AC compared to the total content in the AC sample

Sulphur (L/S 10)							
TMF	48.35 g/kg	YARA	62.6 g/kg	CLAIRS	5.6 g/kg	Lindum	8.2 g/kg
180g	2820 mg	180g	6300mg	180	1002.6 mg	180g	603,43 mg
1 kg	15,67 g	1 kg	35g	1 kg	5.57	1 kg	3,35
32% leaks out		56% leaks out		99 % leaks out		41% leaks out	

The percentage calculation was calculated like:

E.g., Yara

- 3500 mg/L · 1.8L = 6300 mg

To make the result easier to compare, the concentration was converted to 1 kg before the percentage calculation was done

- (1000/180) · 6300mg = 35000mg = 35g

The percentage value:

- 35g/62g · 100 = 56%

Appendix G: PFAS concentration in the eluate for the different AC samples

Table G1: The concentration of PFAS in the sample water (ng/L) with standard deviation for the different L/S ratios from the different AC samples. The results shown are averages from triplicate tests.

Sample reference	L/S ratio	PFAS ng/L	St.dev ng/L
L/S 2000/500/100			
CLAIRS unused	Control water	7400	4403
	2000	2587	544
	500	417	61
	100	117	9
CLAIRS spent	Control water	11333	471
	2000	3700	2417
	500	2033	125
	100	262	239
NRV unused	Control water	9733	205
	2000	453	12
	500	76	3
	100	12	1
NRV spent	Control water	11000	0
	2000	10333	471
	500	9367	170
	100	6400	82
L/S 8000/2000/1000			
Yara spent	Control water	13667	471
	8000	12000	0
	2000	7733	1226
	1000	3833	170
TMF spent	Control water	9933	47
	8000	9833	94
	2000	9767	47
	1000	9600	245
Lindum spent	Control water	10667	471
	8000	11000	0
	2000	11000	0
	1000	11000	0
KKV unused	Control water	10000	0
	8000	9933	94
	2000	10667	471
	1000	10333	471
KKV spent	Control water	12000	0
	8000	12000	0
	2000	12000	0
	1000	12333	471

Appendix H: K_d value for the different concentration of the AC samples.

Table H1: Concentrations of the average L/S ratios after triplicate tests, for the different AC sample and how much AC (g) is added for the different L/S ratios. Then the PFAS concentration in water (Caq) and concentrations in the sorbent (Cs), and then calculating the K_d value (L/g) from these.

Sample reference	L/S	AC (Gram)	Caq (Ng/L)	Cs (Ng/g)	K_d (L/g)
L/S 2000, 500, 100					
CLAIRS spent	2000	0.2	3700	38165	3,37
	500	0.8	2033	11625	1,025
	100	4	262	2767,75	0,25
CLAIRS unused	2000	0.2	2567	24165	3,27
	500	0.8	417	8728,75	1,18
	100	4	117	1820,75	0,25
NRV spent	2000	0.2	10333	3335	0,30
	500	0.8	9367	2041	0,19
	100	4	6400	1150	0,10
NRV unused	2000	0.2	453	46400	4,77
	500	0.8	76	12071,25	1,24
	100	4	12	2430,25	0,25
L/S 8000, 2000, 1000					
Yara spent	8000	0.05	12000	33340	2,44
	2000	0.2	7733	29670	2,17
	1000	0.4	3833	24585	1,80
Lindum spent	8000	0.05	11000	-6660	-0,62
	2000	0.2	11000	-1665	-0,16
	1000	0.4	11000	-832,5	-0,08
TMF spent	8000	0.05	9833	2000	0,20
	2000	0.2	9767	830	0,08
	1000	0.4	9600	832,5	0,08
KKV spent	8000	0.05	12000	0	0
	2000	0.2	12000	0	0
	1000	0.4	12333	-832,5	-0,07

KKV unused	8000	0.05	9933	1340	0,13
	2000	0.2	10667	-3335	-0,33
	1000	0.4	10333	-832,5	-0,08

Appendix I: BTEX-PFAS K_d value and TOC-PFAS K_d value

Tabell I1: Concentration of BTEX (mg/kg) and TOC (% w/w) from the contamination analyses correlates with the PFAS-K_d value (L/g)

Sample reference	BTEX mg/kg	TOC (% w/w)	K_d (L/S 2000)
CLAIRS unused		87.9	3.27
CLAIRS spent	300	67.5	2.37
Yara spent	1600	64.5	2.13
TMF spent	240	71.3	0.08
Lindum spent	1500	80	-0.16
NRV unused		86	1.24
NRV spent		42.7	0.3
KKV unused	15	80.5	-0.33
KKV spent		15.5	0

Appendix J: Concentration of DOC

Table I1: DOC concentrations (ng/L) from the triplicate average with standard deviation in the sample water.

Sample reference	L/S ratio	DOC	St.dev
		ng/L	ng/L
L/S 2000/500/100			
CLAIRS unused	Sample water	25	4
	2000	156	63
	500	173	33
	100	137	19
CLAIRS spent	Sample water	373	83
	2000	370	163
	500	89	8
	100	190	73
NRV unused	Sample water	259	209
	2000	168	111
	500	380	85
	100	423	76
NRV spent	Sample water	347	29
	2000	135	82
	500	101	44
	100	88	74
L/S 800/2000/1000			
Yara spent	Sample water	61	19
	8000	75	66
	2000	61	20
	1000	69	5
TMF spent	Sample water	317	79
	8000	128	48
	2000	94	58
	1000	243	47
Lindum spent	Sample water	370	150
	8000	250	90
	2000	300	50
	1000	383	60
KKV unused	Sample water	290	104
	8000	470	106
	2000	507	88
	1000	413	90
KKV spent	Sample water	213	42
	8000	200	8
	2000	323	101
	1000	273	47

Appendix K: PFAS and DOC concentration from the different cells

Table J1: Concentration variations of PFAS and DOC for cell 1-2 and 3 with spent CLAIRS AC. The results shown are averages from triplicate tests.

Sample reference	L/S ratios	PFAS	St.dev	DOC	St.dev
		Ng/L	Ng/L	Ng/L	Ng/L
CLAIRS from Cell 1-2	Sample water	11333	471	373	83
	2000	3700	2417	370	163
	500	2033	125	89	8
	100	263	239	190	73
CLAIRS from call 3	Sample water	4067	47	883	116
	2000	3433	125	1003	78
	500	2167	47	1047	75
	100	870	28	997	144

Appendix L: Concentration over PFAS and TOC for the different sample sites in the *in-situ* experiment.

Tabell K1: Concentrations of PFAS and TOC (ng/L) in the leachate during the five weeks of the in-situ experiment.

Sample sites	Week	PFAS	TOC
#1	1	12000	750
#1	2	3900	560
#1	3		
#1	4	3400	710
#1	6	500	690
#2a	1	1900	670
#2a	2	1900	670
#2a	3		
#2a	4	1700	630
#2a	6	1700	620
#2b	1	13000	5.5
#2b	2	11000	5.2
#2b	3		
#2b	4	13000	5.5
#2b	6	12000	5.5
#3	1	340	200
#3	2	1300	320
#3	3		
#3	4	4900	330
#3	6	6200	320