



## Excavated vs novel *in situ* soil washing as a remediation strategy for sandy soils impacted with *per*- and polyfluoroalkyl substances from aqueous film forming foams

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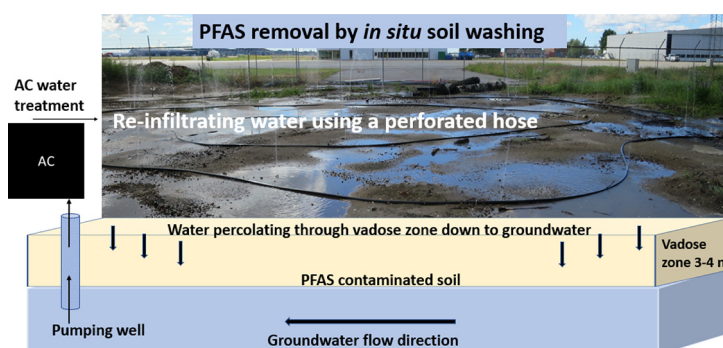
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### HIGHLIGHTS

- Soil washing, *in situ* vs excavated was tested for PFOS contaminated sandy soil.
- First real field scale test of *in situ* soil washing of PFAS contaminated soil
- 73% removal of PFOS in test of *in situ* soil washing, though 100% is attainable.
- Non-calibrated 1-D first order saturated model predicts removal within a factor 2.
- Model and methods applicable to other PFAS contaminated sandy soils

### GRAPHICAL ABSTRACT



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### ABSTRACT

*In situ* soil washing at the field scale has not yet been investigated as a remediation strategy for soils impacted by *per*- and polyfluoroalkyl substances (PFAS). This remediation strategy is a promising low-cost alternative to other costlier remediation options like excavating, transporting and landfilling large amounts of PFAS contaminated soil. However, it is unclear if it is effective at the field scale, where large areas of heterogeneous soil can be challenging to saturate with infiltration water and then pump to a treatment facility. To address this for the first time, herein we established three different trials involving *in situ* washing of an undisturbed, 3 m deep, sandy vadose zone soil contaminated with aqueous film forming foam (AFFF). The trials were performed at a site with an established pump and treat system for treating PFAS contaminated groundwater. *In situ* soil washing was compared to the more conventional practice of washing excavated soil on top of an impermeable bottom lining where the PFAS contaminated water was collected and monitored in a drainage system before treatment. The measured amount of perfluorooctane sulfonate (PFOS) removed was compared with expectations based on a non-calibrated, 1-D first order rate saturated soil model using only the local soil-to-water distribution coefficient as well as the volume and irrigation rate of wash water as input. This model predicted results within a factor of 2. The suspected reasons for small discrepancies between model predictions and excavated vs *in situ* washing was a combination of the heterogeneity of PFOS distribution in the soil as well as preferential flow paths during soil washing that prevented full saturation. This analysis showed that *in situ* soil washing was more efficient and less costly than washing excavated sandy soil, particularly if a pump-and-treat system is already in place.

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## 1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a large group of chemicals which are resistant to chemical, biological and physical degradation processes in their natural environment (Glüge et al., 2020). In 2018 there were more than 4700 registered PFAS CAS numbers (UNEP, 2018). Due to their valuable properties like hydrophobicity, lipophilicity and thermal stability, PFAS have been used increasingly in consumer products and industry since the 1950s (Buck et al., 2011). PFAS are extremely persistent and once present in the environment human intervention is needed to remove them, if possible (Goldenman et al., 2019). The EU's Chemicals Strategy for Sustainability Towards a Toxic-Free Environment, published in October 2020, presents extensive action points to remove pollutants from the environment, one of which is to restrict the use of PFAS, unless their use is essential (EU, 2020).

PFAS has been used in aqueous film forming foam (AFFF) production starting from the 1960s (Norden, 2013; Dauchy et al., 2017). The surface tension lowering properties of PFAS gave PFAS-AFFFs superior firefighting capabilities compared to other, previously designed non-PFAS firefighting foams (Schaefer et al., 2008). Therefore, PFAS-AFFFs have been used extensively at civil and military airports, resulting in PFAS contamination in soil, groundwater, surface waters and biota worldwide (Guelfo and Higgins, 2013; Houtz et al., 2013; Ahrens et al., 2015; Filipovic et al., 2015; Anderson et al., 2016). In the unsaturated zone (also called the vadose zone) PFAS can sorb to soil, accumulate in terrestrial biota, be taken up in plants, and leach to the underlying groundwater (Sharifan et al., 2021). The fate and transport of different PFAS in the vadose zone are influenced by the chemical properties of PFAS, but also properties of the site specific soil such as grain size, dry bulk density, porosity, organic content, mineral content, hydraulic conductivity, air-water interface, saturation rate and the solution ionic strength, some of which have been thoroughly investigated (Lyu et al., 2019; Brusseau, 2020; Guelfo et al., 2020; Van Glubt et al., 2021). Individual PFAS have different sorption affinities to soil particles and soil organic matter (Hale et al., 2017; Schaefer et al., 2021) which results in PFAS from AFFF hotspots percolating through the vadose zone to groundwater at different rates. Retention and sorption processes for PFAS in the vadose zone are complex and are still the focus of substantial research (Brusseau, 2020).

Although, some studies have found the majority of PFAS at AFFF sites have leached to the surrounding environment, these tend to have water tables close to the soil surface, and therefore have only a thin vadose zone (e.g. Adamson et al., 2020). PFAS-AFFFs contaminated soil sites with long distances to the water table are essentially long term sources of PFAS emissions into the underlying groundwater below (Høisæter et al., 2019). Previous investigations on the transport of PFAS-AFFFs in pristine, unsaturated soil columns concluded that the retention of certain PFAS, such as perfluorooctanesulfonic acid (PFOS), in the upper vadose zone can be quite substantial, and that accurate predictions of PFAS leaching rates are difficult to predict (Høisæter et al., 2019). One theory to account for this retention of PFOS is enhanced sorption on the air/water interface within in the vadose zone, which slows down the overall leaching under unsaturated conditions (Brusseau et al., 2015; Brusseau, 2018; Lyu et al., 2018; Brusseau et al., 2019; Brusseau, 2020; Guo et al., 2020). Since the vadose zone is rarely saturated under natural conditions, the PFAS sorbed to the air/water interface will continue to be retained within the soil. Due to the complexity of this and other PFAS sorption processes in the vadose zone, the quantification of PFAS leaching rates in the vadose zone to the groundwater is a challenge for accurate risk assessment; there is nevertheless a necessity of appropriate remediation designs to account for this, ranging from mobilization to immobilization (Bolan et al., 2021).

Remediation of PFAS-AFFF contaminated soils has proven to be challenging and expensive due to the chemical and thermal stability of PFAS itself (Ross et al., 2018; Lu et al., 2020). Environmental authorities have

implemented low target concentration values in soil and low environmental quality standard values for water and biota (NEA, 2020). Due to this, large amounts of soil must be remediated at several AFFF contaminated sites worldwide to reach local environmental goals. Dig and dump solutions are expensive remediation techniques, where PFAS contaminated soil is removed and transported long distances to designated landfill sites, where they may pose less risk.

As an alternative to the excavation and transportation of PFAS contaminated soil, sites could be remediated using pump and treat systems, where the PFAS contaminated groundwater is pumped up and treated in a water treatment facility for PFAS (e.g. filtration on activated carbon (AC)), before incineration of the carbon filter mass). However, due to the slow leaching of PFAS, this would require a long-term resource commitment, as such facilities would likely need to run on the scale of decades. The overall range of costs related to environmental remediation of PFAS contamination for five Nordic countries is estimated to be up to 11 billion Euros (Goldenman et al., 2019). There is therefore an urgent need for feasible, rapid, and cheaper, on-site remediation solutions for PFAS contaminated soil at AFFF sites like airports and military bases.

Bolan et al. (2021) recently reviewed studies on immobilization and mobilization for soil remediation of PFAS. Stabilization *via* tilling or mixing excavated soil with a sorbent to immobilize the PFAS compounds within the soil and further prevent leaching has been considered as a promising method. Sorbents tested for PFAS contaminated soils include – but are not limited to AC (Hale et al., 2017), biochar (Kupryianchuk et al., 2016), modified clay (Das et al., 2013) and a sorbent mix containing AC and aluminium oxyhydroxides (Bräunig et al., 2021, Mahinroosta and Senevirathna, 2020).

In contrast to immobilization, mobilization of PFAS *via ex situ* soil washing may prove a promising method to actually remove PFAS rather than slow emissions. Soil washing is a separation technique for excavated soil often carried out off site, which uses water with or without solvents or surfactants to wash PFAS from the soil and into a water treatment facility. In contrast to unsaturated conditions, under saturated conditions, or during soil washing, the leaching behavior is more predictable and can be accounted for by the soil-specific soil-to-aqueous phase partitioning coefficient (Kd) (Hale et al., 2017; Higgins and Luthy, 2006; Wei et al., 2017). This is partly due to sorption to the air-water interface no longer being a relevant consideration. *In situ* soil washing would have logistical advantages over excavated *ex situ* soil washing, for the simple reason that it avoids excavation and additional transportation.

The aim of this study was to test and model *in situ* soil washing at a firefighting training facility with heavily PFAS contaminated sandy soil from the use of PFAS-AFFF, and to compare this to locally excavated soil washing. To our knowledge, this is the first study of its kind to look at *in situ* soil washing of undisturbed soil as a remediation option. Contaminated water from the soil washing tests was collected and treated in AC filters to prevent spreading. An expected limitation is the difficulty in ensuring homogenous soil saturation and flow rates during washing, due to the potential uneven spreading of infiltration water and the occurrence of preferential flow paths. Therefore, our hypothesis is that the washing effect would be similar to that of soil saturation and homogenous flow of washing water; such that deviations from this hypothesis would be accounted for by soil heterogeneity. This hypothesis is applied to both locally excavated and *in situ* soil washing setups, each differing soil washing area and soil depth, by investigating if PFOS removal through washing could be predicted by the local Kd value and the volume and rate of water added, using a non-calibrated, 1-D first order rate model for fully saturated soil.

## 2. Materials and methods

### 2.1. Site description

The PFAS-contaminated firefighting training facility (FTF) used for the field test of soil washing is located 35 km north east of Oslo,

Norway, and is described in detail in Høisæter et al. (2019). The site area is a flat plain of glaciofluvial deposits dominated by sand. The groundwater level at the site fluctuates throughout the year and is between 3 and 4 m below the surface (Hale et al., 2017). The average groundwater chemistry at the site is Fe of 1.56 mg/L, Mn 0.93 mg/L, Ca 20 mg/L, pH 7.0 and CEC 181 µS/cm. The yearly precipitation in the area is approximately 800 mm, where half of it (400 mm) is lost to evapotranspiration (Jørgensen and Østmo, 1990). Soil sampling at the site in 2016 revealed that PFOS accounted for 96% of the total Σ12-PFAS in the soil, ranging from <0.3 µg/kg to 6500 µg/kg (Høisæter et al., 2019). PFOS is therefore the only PFAS discussed in this paper, but concentrations of Σ12-PFAS for each soil sample are given in the SI.

The FTF had previously established a pump and treat system for groundwater remediation of PFAS. There are several operating AC water treatment plants at the site, both stationary and mobile with different treatment capacities for PFAS contaminated water. PFAS contaminated water is collected from the base of each firefighting training platform, which contains an impermeable bottom liner, before treatment in an AC treatment plant. The groundwater downgradient from the entire FTF is collected in pumping wells and treated in a pump and treat system with another AC treatment plant. This was established to ensure PFAS contaminated groundwater does not spread from the site. The groundwater is treated in an AC filter and monitored for PFAS before being either returned to the soil or used for soil washing. After treatment, the PFOS concentration in the groundwater has been consistently under the detection limit of 10 ng/L.

Grain size distributions from the site show that the soil in the area is generally medium fine sand with some variations in organic content in soil taken from different depths (0–4 m), ranging from 0.17 to 1.2% with an average of  $0.36 \pm 0.27\%$ . The organic content is generally higher in the top meter of the soil, ranging from 0.31 to 1.2% with an average of  $0.71 \pm 0.3\%$ . In the deeper soil samples (1–4 m), the organic content is generally lower varying from 0.17 to 0.36% with an average of  $0.23 \pm 0.05\%$ . (Hale et al., 2017). The hydraulic conductivity (Ks) at the site ranges from  $10^{-3}$  to  $10^{-5}$  m/s (French et al., 2009). Batch leaching tests with contaminated soil samples from the site resulted in an average Kd value for PFOS of 10 L/kg (Hale et al., 2017). A bulk density of 1.3 g/cm<sup>3</sup> is used for the excavated sandy soil and a porosity of 0.51. A bulk density of 1.68 g/cm<sup>3</sup> was determined in earlier studies for sandy soil at the site under *in situ* conditions, with a corresponding porosity of 0.37 (Kłonowski et al., 2008).

## 2.2. Soil washing on excavated sandy soils

Excavated soil washing (Fig. 1a) was carried out at three different firefighting training platforms, Sites A, B and C. Excavated soil was placed and packed in a 1 m thick layer on top of a sloping impermeable high-density polyethylene (HDPE) geomembrane over a swelling bentonite layer that led infiltrating water down slope to a collection pipe. The top soil was slightly compacted and leveled. A detailed description including site specific parameters (test area, soil depth, average PFOS concentration in soil before soil washing, rate of added water, local soil-water partitioning coefficient Kd, bulk dry density, porosity and the duration of soil washing) are given in the Supporting Information (SI, Table S1). In brief, local tap water (pH 8.2 and conductivity of 12.1 mS/m) was added at the soil surface (Fig. 1a) using a water sprinkling system consisting of a perforated hose placed in a spiral on the top soil. Water was applied at different rates that would appear to visually reach saturation levels, as well as for various durations to control for the liquid-to-solid (L/S) ratio. The PFAS contaminated water infiltrating through the soil was sampled in the manhole shown in Fig. 1a regularly during the duration of soil washing (Table S3). The PFAS concentration in the soil was determined before and after the soil washing using a sampling strategy of a minimum of 10 sub-samples per individual soil sample, following ISO 18400-104:2018. The soil layer was 1 m in depth. Depending on the site, 2–7 soil samples were taken before soil washing

and 5–8 after. The test area, soil depth, average concentration and amount of PFOS in the soil, rate of water added, duration the test and total amount of water added at each test locations are given in Table 1. The soil washing results are calculated based on applied water infiltration rates, excluding the natural precipitation during the experimental period. Detailed results from all soil samples are given in Tables S2 and S4.

## 2.3. Field tests of *in situ* soil washing using a pump and treat system

Novel *in situ* washing of the undisturbed vadose zone using a pump and treat system (Fig. 1b) was applied at sites D, E and F. In this system groundwater from the pumping well was treated by AC filtration (Table S6), and then uniformly infiltrated into the topsoil at the site using a perforated hose at a controlled water rate that visually indicated saturation *via* slight pooling at parts of the surface. The level of the groundwater table fluctuated throughout the year and is at the highest expected to be approximately 3 m below terrain; hence the infiltrated water will percolate through at least 3 m of unsaturated zone. The PFAS contaminated percolated water was allowed into the groundwater, so that it could be extracted pumping well that was then AC filtered and recycled as infiltrate water (Table S6). It should be noted that the FTF has a row of pumping wells from intercepting PFAS contaminated groundwater flowing out of the contaminated site in addition to infiltration wells for clean water downgradient to prevent the spreading of PFAS contaminated groundwater. After being treated in the AC treatment plant, the PFOS concentration was monitored and found to be well below the detection limit of 10 ng/L. The pump and treat remediation system at the site is described in by Høisæter et al. (2019). It should be noted that based on this pump-and-treat system causing a hydraulic gradient towards the pumping wells, it is likely that water outside the test area was mixed with that within the test area; however, it is not possible to quantify how much this dilution could have been occurring.

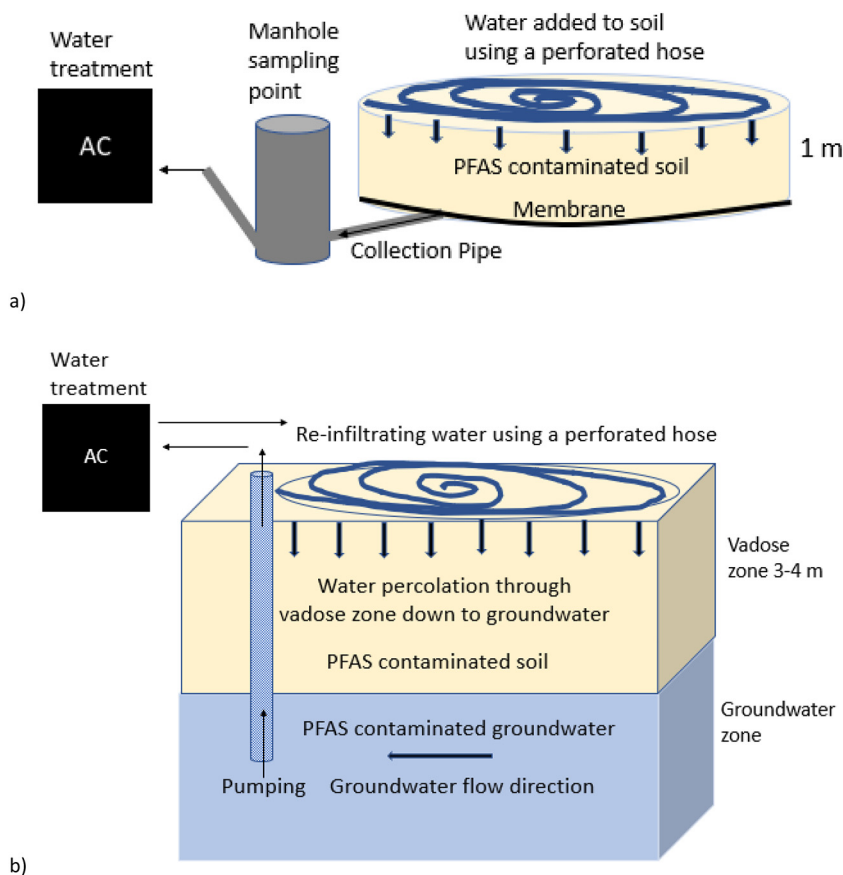
The soil at all three locations was sampled every meter down to groundwater at ca 3 m below the surface, both before and after the soil washing, using a sampling strategy of a minimum of 10 sub-samples per individual sample of 1 m soil, following ISO 18400-104:2018. Depending on the location, 7–10 samples were taken before soil washing and 9–24 after. Groundwater was sampled regularly in the pumping wells at all three sites. The duration of irrigation was sufficient to get a L/S ratio from 5.0 to 5.5 for the different sites (Table 1). Detailed information on the locations, soil depth, average PFOS concentration and amount of PFOS in the soil, the rate of water added, duration of the test and total amount of water added is given in Table 1. Detailed results from all soil samples are given in Tables S5, S7 and S8.

## 2.4. Soil sampling

At all the sites, the soil was sampled using an excavator to dig a trial pit in the sandy soil. The different sites were sampled in a systematic way with different amount of trial pits spread out uniformly at the site. The number of trial pits was different at each site due to the different area of each site, as presented above. Sampling itself followed ISO 18400-104:2018. Further details of samples for each site are given in the SI Tables S2, S4, S5, and S7 to S14. Gloves and a clean spade were used to sample the soil. Before sampling at each point, gloves were changed, and the spade was rinsed with methanol. Soil was transferred into sampling bags (polyamide 1 L) and stored at 4 °C. The samples were homogenized using clean gloves before shipping to an accredited laboratory for PFAS analysis (see below).

## 2.5. Water sampling

At sites A, B and C water was sampled from the collection systems draining the platforms and collected as a grab sample. For the *in situ*



**Fig. 1.** a) Field testing of soil washing of excavated AFFF contaminated soil with an impermeable bottom liner (membrane), allowing for collection of PFAS contaminated water for treatment. Water is added to the soil surface using a perforated hose. b) *In situ* soil washing, also with a perforated hose, where wash water that percolates through the vadose zone enters the groundwater and is then extracted by a pumping well and treated via AC filtration. The AC-filtrated water is then re-infiltrated into the perforated hose.

test sites, groundwater was sampled from either a monitoring well in the washing area (site D), pumped groundwater entering the AC treatment plant (site E) or the extraction well (site F). The groundwater level at site D was measured to be 3 m under terrain when the soil washing started, with a pH of 7 and conductivity of 200  $\mu\text{S}/\text{cm}$ . During the soil washing period, the water was sampled every day the first week and then once a week for the rest of the soil washing period. Water samples (1 L) were collected at 1 m below the groundwater level, after removing stagnant water in the well (25 L).

2.6. 1-D first order rate saturated soil model

The soil washing results were predicted using a non-calibrated, 1-D first order rate saturated soil model. The 1-D model is based on a simplified soil column, where there is no new PFAS entering the column, and with the boundary condition that at time (t, days) = infinity, all PFAS

will be leached out. The soil depth ( $h_{\text{soil}}$ ) (m), soil porosity ( $n$ ) and bulk dry density ( $\rho_b$ ) (kg/L), the amount of water added (**Added water**) ( $\text{m}^3/\text{day}$ ), the infiltration factor (**IF**) (%), the sorption coefficient of PFAS (**Kd**) (L/kg) and the initial PFAS concentration in the soil ( $C_{\text{PFOS,soil,0}}$ ) ( $\mu\text{g}/\text{kg}_{\text{solid}}$ ) were the input values to the model. The predicted output from the 1-D model were the PFOS concentrations in the soil fraction ( $C_{\text{PFOS,soil d.w.}}$ ) ( $\mu\text{g}/\text{kg}_{\text{solid}}$ ) and the porewater (pw) ( $C_{\text{PFOS,pw}}$ ) ( $\mu\text{g}/\text{L}_{\text{pw}}$ ) during the saturated soil washing period is derived by Eqs. (1) and (2). The mass transfer coefficient of soil depletion (k) (1/day) is given by Eq. (3). The soil to porewater partition coefficient or sorption coefficient (**Kd**) (L/kg) for PFAS controls the distribution of PFAS between the soil fraction and the porewater and the relationship is given by Eq. (4). A more detailed description of the derivation of the 1-D model is given in the supporting information.

$$C_{\text{PFOS,soil}}(t) = C_{\text{PFOS,soil,0}}e^{-kt} \tag{1}$$

**Table 1**  
Site specific data for field testing of soil washing.

Soil washing sites	Test area ( $\text{m}^2$ )	Soil depth (m)	PFOS concentration in soil before washing ( $\mu\text{g}/\text{kg}$ ) (n)	PFOS mass in soil before soil washing ( $\text{g}$ ) <sup>a</sup> (n)	Irrigation rate of water added (m/day)	Test duration (days)	Liquid/Solid ratio (L/kg)
Excavated site A	532	0.90	2743 $\pm$ 372 (7)	1707 $\pm$ 232 (7)	0.07	26	1.6
Excavated site B	478	0.80	751 $\pm$ 74 (3)	373 $\pm$ 37 (3)	0.16	117	18.3
Excavated site C	500	1.00	1096 $\pm$ 694 (2)	712 $\pm$ 451 (2)	0.17	117	15.3
<i>In situ</i> site D	360	3	1515 $\pm$ 358 (7)	2749 $\pm$ 650 (7)	0.32	80	5.0
<i>In situ</i> site E	614	3	1291 $\pm$ 366 (10)	3995 $\pm$ 1131 (10)	0.23	120	5.5
<i>In situ</i> site F	761	3	1405 $\pm$ 326 (9)	5389 $\pm$ 1252 (9)	0.17	163	5.5

<sup>a</sup> PFOS in the soil (g) is calculated using the PFOS concentration ( $\mu\text{g}/\text{kg}$ ) in the soil and the mass (kg) of the soil for each location values  $\pm$  standard error of the mean (SEM). The mass of the soil was estimated using the soil depth (m), the area ( $\text{m}^2$ ) and the bulk density (kg/L).

$$C_{PFOS,pw}(t) = C_{PFOS,pw,0}e^{-kt} \tag{2}$$

$$k = \frac{\text{Added water} * IF}{h_{soil}(Kd * \rho_b + n)} \tag{3}$$

$$Kd = \frac{C_{PFOS,soil} d.w.}{C_{PFOS,pw}} \tag{4}$$

### 2.7. Quantification of fraction PFOS removed

The mass (kg) of PFOS in the soil  $M_{PFOS,soil}$ , is determined by the average PFOS concentration measured in the soil,  $C_{PFOS,soil}$  (µg/kg), and total mass of soil  $M_{soil}$ :

$$M_{PFOS,soil} = C_{PFOS,soil} * M_{soil} \tag{5}$$

The number of samples used to determine  $M_{PFOS,soil}$ , and the resulting variation, was different at the six test sites (details in the SI). The PFOS fraction removed was calculated using the mass of PFOS in the soil initially,  $M_{PFOS,soil,initial}$  and after washing,  $M_{PFOS,soil,final}$ :

$$\text{Fraction Removed} = \frac{M_{PFOS,soil,initial} - M_{PFOS,soil,final}}{M_{PFOS,soil,initial}} \tag{6a}$$

In cases where there was very little difference between  $M_{PFOS,soil,initial}$  and the mass of  $M_{PFOS,soil,final}$ , the fraction removed was based on the average PFOS concentration in the wash water,  $C_{PFOS,wash}$ , and the volume of water used for washing,  $L$ :

$$\text{Fraction Removed} = \frac{C_{PFOS,wash} * L}{M_{PFOS,soil,initial}} \tag{6b}$$

It is noted that Eq. (6a) was favoured when possible, as the remaining concentration in soil was considered the remediation target by the local authorities, specifically set at 150 µg/kg dw.

To compare the different treatments and soils we define a normalized “liquid-to-solid removal ratio”,  $L/S_{removal}$ , which is the liquid of wash water required to remove PFOS from a given mass of soil, which is here calculated as the liquid to solid ratio applied at the test site divided by the fraction removed:

$$\text{normalized } L/S_{removal} = \frac{L/S}{\text{Fraction removed}} \tag{7}$$

The normalized  $L/S_{removal}$  is approximately the  $L/S$  ratio needed to flush the PFAS out, assuming the sorption of PFAS to the soil remains constant and the soil porewater remains saturated.

### 2.8. Chemical analysis

Soil and water analyses were carried out at the accredited laboratory Eurofins GfA Lab Service GmbH (Germany), using method DIN 38414-S14 based on acetonitrile extraction followed by analysis using liquid

chromatography coupled with mass spectrometry (LC/MS-MS) for soil samples and DIN 38407-42 and quantification using LC/MS-MS for eluate. Soil samples were analyzed for  $\sum$  12 PFAS, while water samples were analyzed for  $\sum$  23 PFAS. A complete list of the PFAS analyzed in soil and water is given in SI.

### 2.9. Quality control and assurance

No materials that could influence PFAS sorption behavior (glass, metal) were used when handling soil and water samples. PE bottles were used to store samples at 4 °C until analysis. Internal isotopically labelled standards were added to all soil and eluate samples prior to PFAS analysis. PFAS identification was based on retention time and molecule or fragment ions and quantification was carried out by comparison with the internal isotopically labelled standards. Analytical detection limits were 0.2–1 µg/kg TS for PFAS in soil and 0.2–1 ng/L for PFAS in water samples and 0.21 µg/kg TS for PFOS soil and 0.2 ng/L for PFOS in water samples.

### 2.10. Estimation of cost for in situ soil washing as PFAS remediation

The cost assessment of using *in situ* soil washing to remediate the PFAS contaminated soil considered: 1) the installation of 2 pumping extraction wells for groundwater equipped with winter isolation and operation costs; 2) either that PFAS contaminated groundwater is pumped to an existing (no added investment cost for AC treatment), or 3) otherwise to a mobile AC treatment plant from 2 pumping wells with a total capacity of 1.5 L/s (requiring additional operational costs). The cost of incinerating the AC filters is not included in the cost estimates. It is also assumed the water used to wash can be taken from groundwater, The AC treated groundwater is re-infiltrating at the soil surface of the soil washing area with no extra cost for added water. It is further assumed that 95% of the water infiltrates the soil. Tables S17 and S18 show the costs estimated for the most efficient site in this study with and without existing AC treatment.

## 3. Results and discussion

### 3.1. Results from field test of excavated and in situ soil washing

Results from the two washing methods for the three excavated soil sites and the three *in situ* sites are presented in the SI (Tables S2–S8) and summarized in Table 2, which presents the amount of PFOS before and after treatment, the fraction removed, as well the required  $L/S$  ratio for PFOS removal at each site (normalized  $L/S_{removal}$ ).

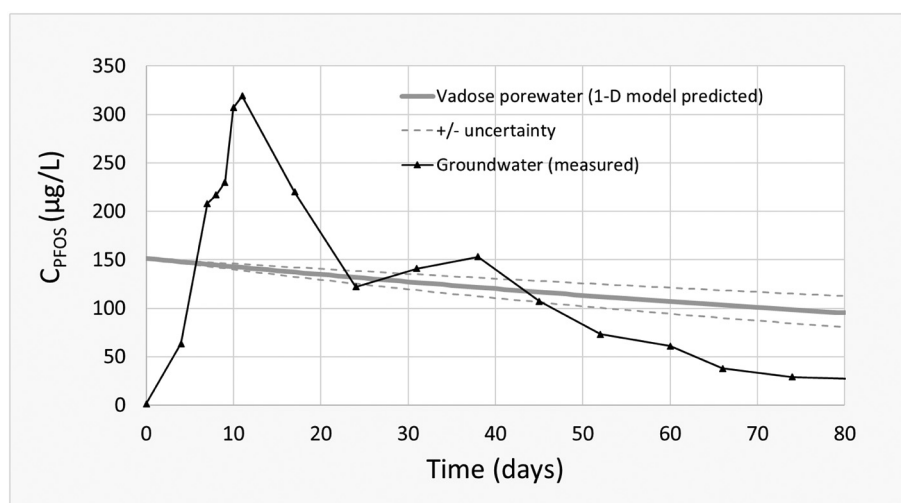
The fractions removed, as calculated using Eqs. (6a) and (6b), are between 11 and 73%, with *in situ* site D being the most effective during the test trial. The average soil concentrations, and therefore fraction removed based on soil concentrations (e.g. 6a), have large standard deviations, due to uneven/heterogeneous soil distribution of PFOS in the soil both before and after washing. The fraction removed based on water concentrations (Eq. (6b)) is a more accurate measurement for the sites A, B and C where all the wash water was collected and sampled,

**Table 2**  
Results from field test of *in situ* soil washing.

Test sites for soil washing	PFOS in soil before soil washing (g) (n)	PFOS in soil after soil washing (g) (n)	PFOS removed from soil (g)	PFOS removed (%)	$L/S$ ratio (L/kg)	Normalized $L/S_{removal}$ (L/kg)
Excavated site A	1707 ± 232 (7)	1736 ± 429 (8)	180 <sup>a</sup>	11 <sup>a</sup>	1.6	14.8
Excavated site B	373 ± 37 (3)	401 ± 220 (5)	170 <sup>a</sup>	46 <sup>a</sup>	18.3	40.3
Excavated site C	712 ± 451 (2)	272 ± 104 (5)	440 ± 463	62 ± 76 <sup>b</sup>	15.3	24.8
<i>In situ</i> site D	2749 ± 650 (7)	734 ± 171 (9)	2014 ± 672	73 ± 30 <sup>b</sup>	5.0	6.8
<i>In situ</i> site E	3995 ± 1131 (10)	1533 ± 278 (24)	2463 ± 1165	62 ± 34 <sup>b</sup>	5.5	8.9
<i>In situ</i> site F	5389 ± 1252 (9)	4131 ± 1042 (10)	1258 ± 1629	23 ± 30 <sup>b</sup>	5.5	23.6

<sup>a</sup> Estimate from average effluent water samples and the amount of water given in SI, Tables S3a and S3b (see Eq. (6b)).

<sup>b</sup> Estimated from the before and after PFOS concentration in the soil, and the amount of soil given in SI (see Eq. (6a)) values ± standard error of the mean (SEM).



**Fig. 2.** Relationship between measured groundwater PFOS concentration (black line) and predicted vadose zone PFOS porewater concentration (grey line) at site D. The dashed line shows the predicted uncertainty of the model.

and was generally consistent with mass loss observed in the soil concentrations. Therefore, the comparisons here between sites need to be viewed with the consideration of the heterogeneity of PFOS distribution in the field sites, in that PFOS was substantially removed as evident based all the collected wash water and soil concentrations, but the heterogeneity in PFOS distribution in the field site was similar before and after washing (Table 2).

Site D requires the lowest normalized L/S (6.82) to essentially remove PFOS. In terms of the amount of water added per weight of contaminated soil, the *in situ* soil washing (normalized L/S ratio 6.9–23.6) appeared to perform better than local washing of excavated soil (L/S ratio 14.7–40.3). As presented in Fig. 2 for Site D, there was a notable chromatographic-like peak in PFOS concentration (319,000 ng/L) 11 days after soil flushing started, which subsequently decreased in a skewed “chromatographic tail” due to potential variations in the lengths of porewater flow paths, variations of PFOS distribution, and variations of sorption properties within the test area, as could potentially be described with a convective-dispersive equation. The soil washing operation ceased before all PFOS was removed (when 73% had been removed). It is clear from Fig. 2 that more PFOS could have been washed out if the experimental period went on for longer. However, it is also evident that since most PFOS was washed out after 120 days followed by skewed chromatographic tail (Fig. 2), the L/S removal ratio would increase if the washing experiment had continued; hence, the normalized L/S ratios in Table 2 are likely lower than required for 100% removal. The lowest fractions removed were observed for local site A (11%) and the *in situ* site F (23%). Site A has a higher average PFOS concentration after soil washing, than before, attributed to the limited number of samples taken after washing and the heterogeneity of PFOS distribution.

It was hypothesized that water infiltration during could be modelled by assuming the washed soil would reach near saturated conditions. Assuming PFOS sorption in all sites were similar, it is likely that this hypothesis was mostly met for the set up used for the *in situ* soil washing, particularly in the case of sites D and E, which gave a more effective PFOS removal than seen in the excavated soil. The excavated soil is uncompacted and might have many macropores that gives an uneven water distribution, causing preferential flowpaths. Preferential flow can bypass the diffusion pathways, thereby reducing the desorption of sorbed PFAS which may be one the reasons for relatively less removal of PFAS in *ex-situ* than *in situ* soil. Therefore, exploration of ways to ensure even infiltration of water at specific rates to obtain a high and even moisture content approaching fully saturated soil in the test setups is recommended; furthermore, sampling density horizontally and vertically and reduction of analytical biases may also impact the quantification of treatment results.

### 3.2. Predicted results for soil washing using a 1-D model

The expected outcome of the soil washing field tests were predicted in advance of obtaining measurements, by using a 1-D first order rate model and the design of the actual field trials. The model input parameters are given in Table S15 and detailed results for each test site including statistical parameters are shown in the supporting information Figs. S1 to S6. The, non-calibrated, predicted concentration after soil washing and the PFOS amount removed are shown in Table 3 together with the calculated PFOS fraction removed. A comparison of the measured amount and fraction of PFOS removed under field conditions is presented in Fig. 3, along with the predicted amount removed.

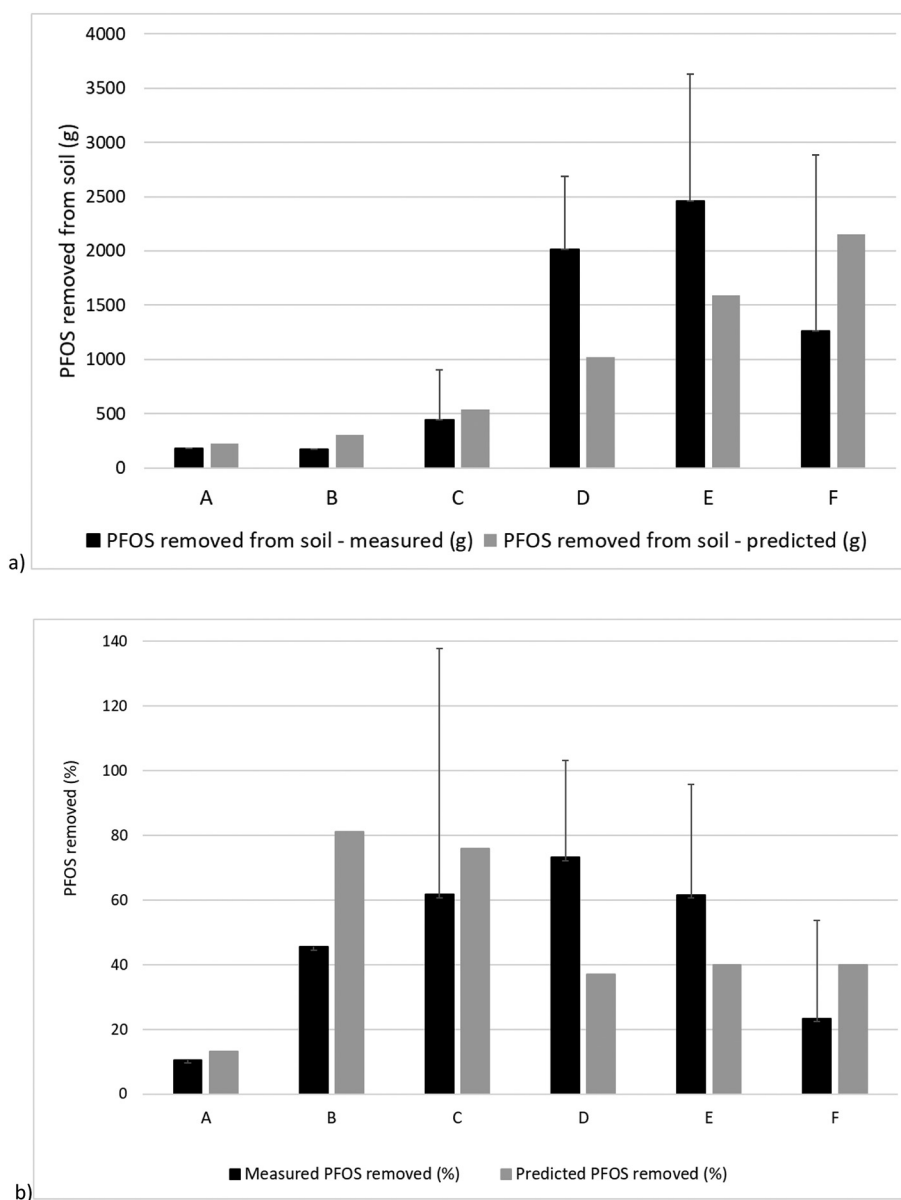
**Table 3**  
Predicted results using the 1-D model for the soil washing tests sites.

Test sites for soil washing	Measured PFOS conc. in soil after soil washing (µg/kg) (n)	Predicted PFOS conc. in soil after soil washing (µg/kg)	Measured PFOS amount removed (g)	Predicted PFOS amount removed (g)	Measured PFOS removed (%)	Predicted PFOS removed (%)
Excavated site A	2789 ± 690 (8)	2379 <sup>c</sup>	180 <sup>a</sup>	227 <sup>c</sup>	11 <sup>a</sup>	13
Excavated site B	807 ± 443 (5)	140 <sup>c</sup>	170 <sup>a</sup>	303 <sup>c</sup>	46 <sup>a</sup>	81
Excavated site C	419 ± 160 (5)	264 <sup>c</sup>	440 ± 463 <sup>b</sup>	541 <sup>c</sup>	62 ± 76 <sup>b</sup>	76
<i>In situ</i> site D	405 ± 94 (9)	952 <sup>c</sup>	2014 ± 672 <sup>b</sup>	1022 <sup>c</sup>	73 ± 30 <sup>b</sup>	37
<i>In situ</i> site E	495 ± 90 (24)	776 <sup>c</sup>	2463 ± 1165 <sup>b</sup>	1594 <sup>c</sup>	62 ± 34 <sup>b</sup>	40
<i>In situ</i> site F	1077 ± 272 (10)	843 <sup>c</sup>	1258 ± 1629 <sup>b</sup>	2156 <sup>c</sup>	23 ± 30 <sup>b</sup>	40

<sup>a</sup> Estimate from average effluent water samples and the amount of water given in SI (see Eq. (6b)).

<sup>b</sup> Estimated from the before and after PFOS concentration in the soil, and the amount of soil given in SI (see Eq. (6a)) values ± standard error of the mean (SEM).

<sup>c</sup> Model errors are logarithmically distributed and given in supporting information in Figs. S1–S6 and Table S16.



**Fig. 3.** a). The calculated amount of PFOS (g) removed from the soil during the test of soil washing compared to the amount removed predicted by the 1-D model. 3b) PFOS removed (%) in the field compared to the predicted results using the 1-D (error bars indicate standard error of the mean). Error for predicted values is given in Fig. S1–S6 and Table S16 in S.I.

Predictions of PFOS fractions removed were greater than observations for all three excavated sites (A, B, C) and one *in situ* site (F) but were lower than observed for the remaining two *in situ* sites (D and E). However, the model agreed with observations generally within a factor 2, with on average a factor of  $1.1 \pm 0.8$  for the average soil concentration after soil washing, and on average a factor of  $1.2 \pm 0.5$  for the predicted fraction removed. The correlation of the measured and the predicted PFOS concentration is shown in Fig. S7 and the correlation for the removed amount of PFOS (g) is shown in Fig. S8. The correlation is high with  $R^2$  of 0.76 for the concentrations and smaller for the PFOS amount removed with  $R^2$  of 0.49. If the largest uncertainty site F is removed the correlation for the amount of PFOS removed increases to an  $R^2$  of 0.94 (Fig. S9). The most likely explanations for these minor deviations is that the model assumes the PFOS contamination and infiltration of wash water to be homogeneously distributed over the entire volume of the test sites. As previously discussed, this may not be the case for sites A, B and C, and this hypothesis can be extended to site F due to observed ponding and uneven distribution of the water at the surface, as preferential flow paths and inhomogeneous PFOS distribution are possible.

Based on the good agreement between predictions of the non-calibrated 1-D model, it appears that this simplistic model is suitable for estimations of the expected performance of soil washing, particularly in cases where sorption ( $K_d$ ) is well characterized and homogeneous infiltration of water through the soil column can be assured.

### 3.3. Predicted long term effect of soil washing

For practical reasons, the field soil washing had a limited duration. It was therefore not possible to see if near complete removal of PFOS was achievable. The 1-D first order rate model was used to predict the potential duration of soil washing to reach different target goals for PFOS for the sites, using the same rate of water infiltration. The predicted duration of washing to achieve two possible target goals of 150  $\mu\text{g}/\text{kg}$  and 50  $\mu\text{g}/\text{kg}$  average PFOS concentration in the soil are given in Table 4. The model predictions show that the soil target of 150  $\mu\text{g}/\text{kg}$  will be reached within 112 to 712 days of soil washing dependent on the site, and the target of 50  $\mu\text{g}/\text{kg}$  after 189 to 1063 days of soil washing. The longer times for the *in situ* sites were due to higher initial concentrations of

**Table 4**

Predicted duration to achieve two possible target goals of 150 and 50 µg/kg of PFOS in soil at the different soil washing tests sites.

Test sites for soil washing	Soil depth (m)	PFOS concentration in soil before (µg/kg) (n)	Duration to achieve soil target goal of 150 µg/kg of PFOS (days)	Duration to achieve soil target goal of 50 µg/kg PFOS (days)
Local site A	0.9	2743 ± 372 (7)	531	732
Local site B	0.8	751 ± 74 (3)	112	189
Local site C	1	1096 ± 694 (2)	163	253
<i>In situ</i> site D	3	1515 ± 358 (7)	397	587
<i>In situ</i> site E	3	1291 ± 366 (10)	506	762
<i>In situ</i> site F	3	1405 ± 326 (9)	712	1063

PFOS being present as well as larger soil volumes relative to irrigation rate of water (Table 1). Thus, such a remediation option would require long term operation of water applications such as perforated hoses or sprinklers, pumps and water treatment facilities, but could ultimately lead to removal of PFAS in the timescales of a few years, without the need for disposal of contaminated soil at designated landfills for PFAS.

### 3.4. Estimated cost of *in situ* soil washing

The predicted costs for *in situ* soil washing are calculated for the test sites based on the cost of treating PFOS contaminated water in an existing AC filter at the site. The cost includes the operational cost (including AC filter incineration), and the installation and full time of operation of two groundwater extraction wells. For site D showing the highest PFOS fraction removed (73%), a cost of 25,000 USD/kg PFOS removed was estimated using an AC water treatment plant already on site (Table S17). If the installation of a mobile AC treatment plant is necessary, the cost will increase to 110,000 USD/kg PFOS removed for the same site. For the excavated soil washing sites the costs included are the treatment of PFAS contaminated water. The cost of tap water and installation of a soil washing platform facility with impermeable lining and piping system is not included. There is no need for installation of wells for the excavated soil treatment. For the sites considered in this study, containing between 0.4 and 5.4 kg of PFAS, the cost per site would vary from 10,000 USD to 600,000 USD, for site A and site F, respectively.

## 4. Environmental implications

*In situ* soil washing is a promising remediation method for PFOS contaminated soils with high permeability and hydraulic conductivity such as sand and gravels. However, it should be emphasized that this was not tested for soils with lower permeability, where this is not expected to perform as well. The better results for *in situ* treatment of undisturbed soil indicates that preferential flow paths that can occur in excavated soil can reduce the effect of soil washing. Uniform distribution of the wash water within the soil pores is important to get the best performance of *in situ* soil washing in the field. Other factors that could play a role in the efficiency of soil washing, which would be of interest to explore in future studies, is the utilization of surfactants (Bolan et al., 2021), and how changes in water chemistry, particularly ion content, on the washing removal performance. The hydrogeology of the site is of crucial importance, as it will determine the recovery and treatment potential of the infiltrated water. An incorrectly designed system could cause PFAS to be spread into the groundwater rather than towards the pumping wells during soil washing.

It is also of crucial importance to collect all the PFAS contaminated water from the soil washing for treatment in an efficient PFAS treatment system. If the soil washing is done at a site with shallow distances to groundwater, it is equally important that the PFAS contaminated groundwater is collected and treated in an efficient PFAS treatment system. This can be done for instance with a row of groundwater

wells downgradient as well as the infiltration of clean water as a barrier downstream. If there are very mobile PFAS on site, such as short-chain PFAS (e.g. perfluorobutylsulfonate, PFBS), the washing would be more efficient, but the use of AC filters would not be appropriate, due to their lack of efficiency; more expensive water treatments would need to be in place (e.g. reverse osmosis) (Liu et al., 2019).

*In situ* soil washing could be a feasible and sustainable remediation option for PFAS contaminated sandy soil in many situations, though in particular for sandy-to-gravelly soils and if there is an existing pump and treatment system already installed, for PFOS or other similar long-chain PFAS. However, no two sites are the same. The sustainability of different, local remediation options should always be compared at the planning stage, including the assessment of excavated soil washing, transport to dedicated landfills with PFAS remediating infrastructure, or continuous long-term pump-and-treat without washing. The risk of contamination to the surrounding environment during excavation and handling of contaminated soil should also be considered. Results from this study show that *in situ* soil washing is a feasible, straight-forward and potentially cost-effective and sustainable remediation approach for many AFFF impacted locations.

### CRedit authorship contribution statement

**Åse Høisæter:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Visualization, Project administration, Funding acquisition. **Hans Peter H. Arp:** Methodology, Software, Formal analysis, Writing – review & editing, Supervision, Funding acquisition. **Gøril Slinde:** Conceptualization, Investigation. **Heidi Knutsen:** Investigation, Writing – review & editing. **Sarah E. Hale:** Methodology, Investigation, Writing – review & editing, Funding acquisition. **Gijs D. Breedveld:** Validation, Writing – review & editing, Supervision. **Mona C. Hansen:** Conceptualization, Methodology, Investigation, Writing – original draft, Visualization.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.148763>.



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