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Laboratory-scale and pilot-scale stabilization and solidification (S/S) remediation of soil contaminated with per- and polyfluoroalkyl substances (PFASs)

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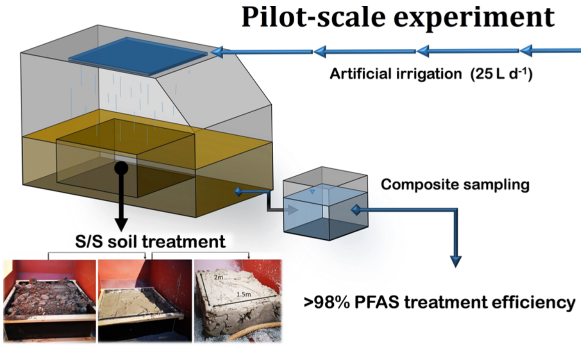
GRAPHICAL ABSTRACT



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ABSTRACT

Remediation of soil contaminated with per- and polyfluoroalkyl substances (PFAS) is critical due to the high persistence and mobility of these compounds. In this study, stabilization and solidification (S/S) treatment was evaluated at pilot-scale using 6 tons of soil contaminated with PFAS-containing aqueous film-forming foam. At pilot-scale, long-term PFAS removal over 6 years of precipitation (simulated using irrigation) in leachate from non-treated contaminated reference soil and S/S-treated soil with 15 % binder and 0.2 % GAC was compared. PFAS removal rate from leachate, corresponding to reduction in leaching potential after 6 years, was > 97 % for four dominant PFASs (perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), perfluorohexanesulfonic acid (PFHxS) and perfluorooctanesulfonic acid (PFOS)), but low (3%) for short-chain perfluoropentanoic acid (PFPeA). During the pilot-scale experiment, PFAS sorption strength (i.e., soil-water partitioning coefficient (K_d)) increased 2- to 40-fold for both reference and S/S-treated soil, to much higher levels than in laboratory-scale tests. However, PFAS behavior in pilot-scale and laboratory-scale tests was generally well-correlated ($p < 0.001$), which will help in future S/S recipe optimization. In addition, seven PFASs were tentatively identified using an automated suspect screening approach. Among these, perfluorohexanesulfonamide and 3:2 fluorotelomer alcohol were tentatively identified and the latter had low removal rates from leachate (< 12 %) in S/S treatment.

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1. Introduction

Contamination of soil and groundwater with per- and polyfluoroalkyl substances (PFASs) is impacting drinking water delivery systems globally, and is posing a risk to human health and the environment (Murakami et al., 2009; Li et al., 2018a; Gellrich et al., 2013; Gyllenhammar et al., 2015). PFAS-contaminated hotspot soils are a particular concern because, although use of PFAS-containing aqueous film-forming foams (AFFFs) has been regulated, these soil contaminants will continue to pollute groundwater and aquatic environments over a long time (Ahrens et al., 2015; Baduel et al., 2015a; Filipovic et al., 2015a). PFAS-contaminated hotspot soils are often the result of unregulated use of PFAS-containing AFFFs at municipal, airport, and military firefighting training facilities (Anderson et al., 2016; Barzen-Hanson et al., 2017; Mejia-Avendaño et al., 2017). Regulations on PFASs are relatively new and focus mainly on direct consumption of food and drinking water (Gobelius et al., 2018), although increasing attention is being given to pollutant source control from soil contamination (Ross et al., 2018a). Due to the unique physiochemical properties of PFASs such as hydrophobic and oleophobic surfactant properties, compared with other organic micropollutants (Rostvall et al., 2018), few viable remediation technologies for soils have been properly evaluated (Ross et al., 2018a; Mahinroosta and Senevirathna, 2020). Remediation techniques based on stabilization, where PFASs are chemically immobilized by adding sorbents (e.g., activated carbon), are receiving growing attention as soil mitigation methods (Shen et al., 2019). These techniques have been shown to reduce PFAS leaching considerably (> 99 % for perfluorooctasulfonate (PFOS)) (Kupryianchyk et al., 2016; Hale et al., 2017a; McGregor, 2018a). However, treatment with high quantities of activated carbon is costly and poor long-term binding efficiency has been reported for short-chain PFASs (McCleaf et al., 2017a). A promising alternative is stabilization and solidification (S/S) (Sörengård et al., 2019a), where a solidifying binder (e.g., cement, bentonite, lime) is added to the soil, combining chemical binding and physical hydrogeological protection that decreases contaminant transport by leaching. In a laboratory-scale study on S/S treatment of PFAS-contaminated soil showed that using 10 % of a binder mixture (CEM II/A-V 52.5 N Portland-fly ash and ground-granulated blast-furnace slag (GGBS)) and 0.2 % pulverized activated carbon (PAC) was a highly effective treatment technique (Sörengård et al., 2019a). The treated soil developed a monolithic structure with 4000 kPa unconfined compressive strength (UCS), indicating reliable long-term physical durability, and conservative leaching tests (crushed to < 2 mm) showed reduced leaching (by > 99.9 %) of most PFASs (except those with perfluorocarbon chain length < 4) (Sörengård et al., 2019a).

However, laboratory studies on S/S treatment of contaminated soils and sediments have been criticized for not truly representing field conditions (Ludwig et al., 2000a; Bozkurt et al., 2000a). For example, the scale is critical for S/S remediation since the ratio between exposed contaminated volume and the infiltrated water changes with scale and hence the rate of contaminant mass-transfer (Hills et al., 2015). Another important factor is that the chemical conditions such as soil aging can affect sorption strength, mass-transfer and kinetics, as it has been shown that field-scale stabilization with activated carbon achieved much higher PFAS removal rate from leachate (McGregor, 2018b) than the same treatment in the laboratory (Sorengard et al., 2019). Thus field-derived solid-liquid sorption coefficients (K_d) may be orders of magnitudes higher that laboratory-derived K_d values (Li et al., 2018b). Remediation methods based on stabilization have also been criticized for having limited longevity (Ross et al., 2018b), and questions remain about how well results obtained in laboratory-scale tests can be used to predict long-term durability and stability in the field (Sörengård et al., 2019a). However, long-term field performance is difficult to assess although an experimental study showed satisfactory treatment performance after 17 years for heavy metals (Wang et al., 2014) and modeling studies have shown that S/S treatment can delay the leaching process by over 100 (Ludwig et al., 2000b) to 1000 (Bozkurt et al., 2000b) years for metals and polyaromatic hydrocarbons (PAHs).

Although the S/S technique has gained recent vitality (Shen et al., 2019), only a few recent studies have been conducted under field conditions (Xia et al., 2019a, b) and the leaching behavior of organic compounds are often only assessed for total organic carbon or chemical oxygen demand (Wang et al., 2015(Xia et al., 2019a), or advancements of the hydration process of the binder (Pan et al., 2019). However, studies on S/S treatment for persistent organic pollutants are rare including PFASs (Sörengård et al., 2019a), 2-chloroaniline (Gallo et al., 2009), nitrobenzene (Liu et al., 2012), dioxide, furan (Bates et al., 2000a) and PAHs (Bates et al., 2000b; Ma et al., 2018a, b).

The aim of this study was to evaluate the efficacy of the S/S technique in reducing leaching of PFASs from contaminated soil in a pilot-scale controlled experiment under conditions close to reality. Specific objectives were to (i) evaluate the PFAS removal rate from leachate following S/S treatment of 3 tons of contaminated soil ($n = 2$) compared with non-treated reference contaminated soil ($n = 2$) in an experiment simulating 6 years of natural rainfall, (ii) assess the effect of S/S treatment on the leaching behavior of a set of well-known PFASs ($n = 18$), (iii) compare the PFAS removal rate in leachate of S/S treatment in pilot-scale, laboratory-scale batch and laboratory-scale monolith leaching tests, (iv) predict long-term PFAS leaching (> 6 years) in pilot-scale conditions, and (v) identify treatment effects on previously non-studied PFASs using an automated suspect screening approach (including > 3000 PFASs) based on liquid chromatography coupled to high-resolution mass spectrometry (LC-HRMS).

2. Method

2.1. Analytical standards

The target PFASs were: C_4-C_{11} perfluoroalkyl carboxylic acids (PFCAs) (PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA), C_4, C_6, and C_8 perfluoroalkyl sulfonic acids (PFSAs) (PFBS, PFHxS, PFOS), 6:2 and 8:2 fluorotelomer sulfonic acids (FTSAs), C_8-based perfluorooctane sulfonamides (FOSAs) (FOSA, EtFOSA), and C_8-based perfluorooctane sulfonamidoacetic acids (FOSAAs) (FOSAA, MeFOSAA, EtFOSAA). In addition, 12 isotopically labeled internal standards (ISs) were included ($^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_5$-PFNA, $^{13}C_2$-PFDA, $^{13}C_2$-PFUnDA, $^{13}C_2$-PFDoDA, $^{18}O_2$-PFHxS, $^{13}C_4$-PFOS, $^{13}C_8$-FOSA, D_5-EtFOSA, D_3-MeFOSAA, and D_5-EtFOSAA). Abbreviation, supplier, and purity of the native PFASs are listed in Table S1, and those of the ISs in Table S2, in Supporting Information (SI).

2.2. Soil characteristics

In June 2018, composite soil samples were collected at a site in Uppsala, Sweden, where the soil is contaminated with PFAS-containing AFFF. The site (not specified for confidentiality) has been identified as a possible local risk area for the municipal drinking water source and is therefore a candidate for remediation. The soil texture at the site is 30 % sand, 28 % silt, and 42 % clay (based on wet sieving in the Swedish standard method (SS027123)), and soil pH is 8.6 (liquid/solid ratio of 10 for 24 h; 691 pH Meter, Metrohm, Switzerland). Soil organic carbon content at sampling, measured using loss of ignition at 550 °C for 24 h, was 6% and hydraulic conductivity (Swedish standard method (SS027126), n = 2) was $< 7.3 \times 10^{-11}$.

2.3. Soil-specific optimization of binder and additive for S/S treatment

Binder optimization of the S/S recipe was performed in accordance with findings in a previous study on S/S additives for PFAS remediation (Sörengård et al., 2019a). In brief, separate samples ($n = 2$) comprising 500 g of soil were treated with binder mixtures including: Bascem (CEM

II/A-V 52.5 N Portland-fly ash, Cementa Sweden), Multicem (Cementa Sweden), ground granulated blast-furnace base slag (GGBS) (Merit 5000, Merox, Sweden), lime (QL, Nordkalk, Sweden), powdered activated carbon (PAC) (Pulsorb FG4, Chermviron Carbon, Sweden), and granulated activated carbon (GAC) (Filtrasorb 400, Chermviron Carbon, Sweden) (Table S3 in SI). For strength test evaluation, a standard method (ISO/TS 17892-7:2005, Swedish Standards Institute, 2005) was used, where the shear strength [kPa] was measured and UCS was calculated as two times the shear strength. Leaching tests were performed following the European standard batch leaching test procedure (EN 12457-1. CEN, 2002) (for details see Table S3, Fig. S1 and SI text).

2.4. *Laboratory-scale leaching tests*

At laboratory-scale, both a batch test and a monolith leaching test were performed. The batch test was performed on the soil samples used in S/S recipe optimization (see Section 2.3) and on pilot-scale S/S-treated samples ($n = 3$) cured for 12 months, following method EN 12457-1. CEN, 2002, as in a previous PFAS S/S study (Sorengard et al., 2019). In brief, 15 g dry weight (dw) portions of reference soil and crushed S/S material sieved to a 0.1–2.0 mm fraction were mixed with 30 mL Millipore water in 50 mL PP-tubes (Corning®, nonpyrogenic) with a liquid/solid ratio of 2. The soil suspensions were shaken for 0.1 h, 0.5 h, 1 h, 2 h, 4 h, 8 h, 16 h, 24 h, and 28 h at 200 rpm in an end-over-end shaker (Reax 2, Heidolph), and then separated through centrifugation at 3000 rpm for 15 min. The PFAS concentrations in the liquid phase were analyzed as described in Section 2.6.

The laboratory-scale monolith leaching test was performed, using a method described elsewhere (USEPA, 2013), on 12-month cured specimens from the S/S pilot-scale test, in order to compare the results of laboratory-scale and pilot-scale leaching tests. In brief, 5cm × 5cm (height x diameter) cured samples of S/S-treated soil (56 ± 7.6 g) from the pilot-scale treatment were placed in 1-L wide-mouth polypropylene containers filled with Millipore water. The convex side of the S/S-treated soil sample was placed towards the bottom and it was assumed that mass transfer of PFASs from this relatively small area was negligible. After 0.5, 1, 2, 9, 20, 29, and 39 days, S/S-treated soil was transferred to fresh 1-L wide-mouth polypropylene containers filled with Millipore water. The PFAS concentrations in the leachate from each container were measured as described in Section 2.6.

2.5. *Pilot-scale leaching experiment*

In the pilot-scale leaching experiment, two S/S treated soil samples and two non-treated reference soil samples were evaluated (Fig. 1). The soil originated from the same AFFF-contaminated field site as the soil used in laboratory tests, and was homogenized with an excavator before the experiment. For S/S treatment, a ratio of 15 % binder (Bascement, Cementa, Sweden) to 85 % soil was used, with 2% of the binder consisting of GAC (Filtrasorb 400, Chermviron Carbon, Sweden). Although PAC had better removal capacity (Fig. S1 in SI), GAC was used here because of its practical benefits when mixing large volumes of soil with activated carbon. Binder, additive and soil were mixed batch-wise in a 100-L forced-action mixer. Tap water was added until a workable mixture was reached, with a final water:binder ratio of approximately 1:1. Tap water was used because of practical and real-life conditions, and was assumed not to have a considerable influence on the S/S system. The blocks of S/S treated soil ($n = 2$) and untreated reference soil ($n = 2$) were placed in containers (12 m^3) fitted with a drainage system made from PVC piping and a permeable layer of gravel (0.3 m height) at the bottom. Using an excavator, soil was transferred into a wooden frame (1.4 m × 1.5 m × 0.6 m, width × length x height) inside each container. After 24 h of curing, inert sand was added to fill the space between the wooden frame and the container to the level of the S/S-treated soil surface, and finally the wooden frame was removed. A PVC roof was erected above each container to protect the experiment from rainfall. Under the roof, an irrigation system consisting of drip tubes (Gardena, Germany) with a flow rate of 30 L d^{-1} (equivalent to 15 mm irrigation d^{-1} applied for 15 min per day simulating rain events) was installed above each container. The flow rate was regulated with a digital water timer (C1060 plus, Gardena, Germany) connected to a four-way distributor (8194-20, Gardena, Germany). The leachate from each container was collected in a 1-m^3 PVC container connected to the bottom of the container with a U-welded metal tube. This prevented backflow and kept the water level constant at the height of half the S/S treated or reference soil, thereby simulating both subsurface unsaturated percolation and saturated transport. Leachate samples were collected at 2- to 3-week intervals for a total period of six months, simulating > 6 years of natural irrigation (average precipitation in Uppsala, Sweden, is 400 mm $year^{-1}$).

2.6. *PFAS target analysis*

Concentrations of the target PFASs in the soil were analyzed in triplicate ($n = 3$) using a method described and validated elsewhere (Gobelius et al., 2017). In brief, 3.0 g of freeze-dried (over 7 days) homogenized (in the field by an excavator and in the laboratory by a ceramic mortar) solid sample were fortified with 100 µL of IS mixture (c = 10 ng mL^{-1}) and extracted with solid-liquid extraction using 30 mL methanol solution comprising methanol and 1 M sodium hydroxide solution (80:20) (LiChrosolv, Merck, Germany). The extract was concentrated under a gentle stream of nitrogen gas to 0.5 mL, fortified with 0.5 mL Millipore water, and transferred to Eppendorf tubes (Eppendorf, Germany). The extract was then vortexed for 15 min, centrifuged at 15,000 rpm for 15 min, and filtered through a 0.45 µm cellulose syringe filter (Sartorius, Germany) into a 2.0 mL brown glass vial for analysis.

For analysis of liquid samples from the batch leaching tests (see Sections 2.3 and 2.4), 500 µL of the aqueous phase (c_w) were transferred to Eppendorf tubes and directly spiked with 50 µL of the IS mixture (c = 10 ng mL^{-1}) together with 450 µL HLPC grade methanol. The Eppendorf tubes were vortexed and centrifuged (15,000 rpm) and the contents were then filtered through recycled cellulose syringe filters (0.45 µm, Sartorius) into 2.0 mL brown glass vials for instrumental

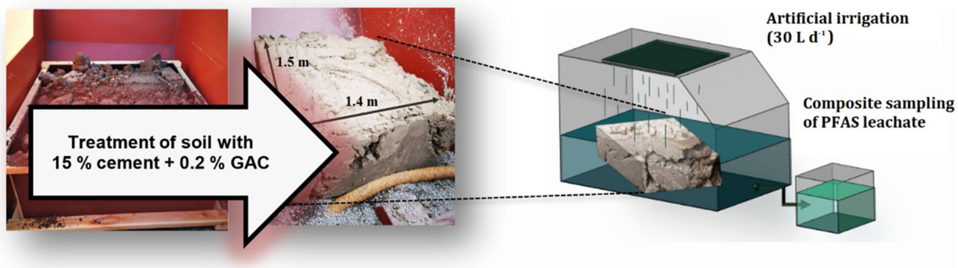

Fig. 1. (Left) Stabilization and solidification (S/S) treatment of soil (15 % binder and granulated activated carbon (GAC) at 0.2 % of the binder amount) and (right) experimental set-up used for SS-treated soil in the pilot-scale leaching experiment.

analysis.

Samples of soil leachate (250 mL) from the monolith leaching tests (see Section 2.4) and the pilot-scale leaching tests (see Section 2.5) were fortified with 100 µL of IS mixture (c = 10 ng mL^{-1}), filtered through a 0.7 µm GGF (Whatman) syringe filter, and concentrated on a solid-phase extraction cartridge (150 mg Oasis® WAX, 60 µm, Waters, Massachusetts, USA) at a rate of one drop per second. The samples were eluted with 8 mL 1 M ammonium hydroxide in methanol and 4 mL pure methanol. The extract was concentrated under a gentle stream of nitrogen gas to 0.5 mL, fortified with 0.5 mL Millipore water, and transferred to a 2.0 mL brown glass vial for instrumental analysis.

Analysis of the 18 target PFASs was performed using an ultra-high performance liquid chromatograph (TSQ Quantiva; Thermo Fisher, USA) coupled to a tandem mass spectrometer (UHPLC-MS/MS) (Quantiva TSQ; Thermo Fisher) using a BEH-C18 column (1.7 µm, 50 mm, Waters) and an injection volume of 10 µL. The eluent gradient was set to 12 min, and the mobile phases were Millipore water and acetonitrile plus 5 mM ammonium acetate (NH_4Ac). The isotope dilution method was used for quantification and for compensating for losses and matrix effects. An eight-point calibration curve ($0.01-100$ ng mL^{-1}) was used for quantification and the data were evaluated using TraceFinder™ software (Thermo Fisher).

2.7. PFASs suspect screening

The soil extracts ($n = 2$) and PFAS leachate from the last sampling point in the pilot-scale tests ($n = 2$) were subjected to PFASs suspect screening using an Acquity UHPLC system (Waters, Milford, MA, USA) coupled to a hybrid quadrupole-Orbitrap mass spectrometer Q-Exactive® (Thermo Fisher Scientific, San Jose, CA, USA) equipped with a heated-electrospray ionization source (HESI) in negative ionization mode. Chromatographic separation was achieved on an Acquity UPLC® BEH C18 (2.1 mm x 50 mm, 1.7 mm particle size, Waters UK). The injection volume of extract was 10 µL. A solvent gradient with 5 mM ammonium NH_4Ac in acetonitrile and an aqueous solution of 5 mM NH_4Ac was used. Acquisition was carried out in data-independent (DIA) mode.

The suspect screening strategy applied for tentative identification of PFASs was as described elsewhere (Sörengård et al., 2020a), based on the NORMAN Digital Samples Freezing Platform (DSFP) (Alygizakis et al., 2019). In brief, raw files were converted to mzML files and the data-independent acquisition chromatograms were separated into low- and high-collision energy chromatograms. The layer-separated mzML files and their meta-data (instrument conditions, sample information, matrix-specific data, and retention time index (RTI) for calibrant substances) were uploaded to DSFP. The platform has an integrated standard procedure for processing mzML files and for storing the files together with all meta-data for generation of data collection template (DCT) databases. Uploading of files in DSFP enabled retrospective screening of 3,425 PFASs (List S25 OECD PFAS LIFE APEX) in the soil samples, based on (i) mass accuracy, (ii) use of advanced chromatographic retention time prediction models (Aalizadeh et al., 2016, 2019), and (iii) inspection of MS/MS fragments (Gago-Ferrero et al., 2018).

2.8. Calculations

To evaluate the effect of S/S treatment on the leaching behavior of different PFASs, removal rate from leachate (E %) was assessed by comparing the individual PFAS concentrations in the aqueous phase (c_a [µg mL^{-1}]) of treated S/S soil (c_{aq}) to those in the aqueous phase of non-treated S/S soil ($c_{aq,ref}$), calculated as:

$$E\,\%= \left(1-\frac{C_{aq}}{C_{aq,ref}}\right)\times 100 \tag{1}$$

where E [%] > 0 indicates increased PFAS retention.

The solid-water partitioning coefficient (K_d) [L kg^{-1}] was calculated as:

$$K_d=\frac{C_s}{C_{aq}} \tag{2}$$

where c_s [µg g^{-1} soil dw] is the concentration of an individual PFAS in the soil solid phase.

A one-dimensional (1-D) model was used to predict future leaching from the S/S-treated soil and reference soil Hale et al. (2017b):

$$C_{aq}(t)=C_{aq,0}e^{-kt} \tag{3}$$

where t [years] is time and $c_{aq,0}$ is starting leaching concentration. The constant –k was assessed with the Trust-Region algorithm in MATLAB.

2.9. Quality control and quality assurance

Laboratory ($n = 3$) and field blank ($n = 2$) samples were included in all experiments. The PFAS concentrations in the blanks were lower than the instrument detection limit (IDL), and hence method detection limit (MDL) was set equal to IDL, which in turn was set to the lowest calibration point with a response factor not deviating by more than 30 % of the average response factor (ARF). The average IDL for all PFASs analyzed ranged between 0.05^{-1} ng mL and 1 ng mL^{-1}, and differed between experiments (Table S4 in SI).

Absolute method recovery for the analytes was calculated based on the loss of ISs during sample preparation and matrix effects, and was compared against the calibration curve. For PFASs detected over the MDL (PFPeA, PFHxA, PFOA, PFBS, PFHxS, PFOS, 6:2 FTSA, FOSA and

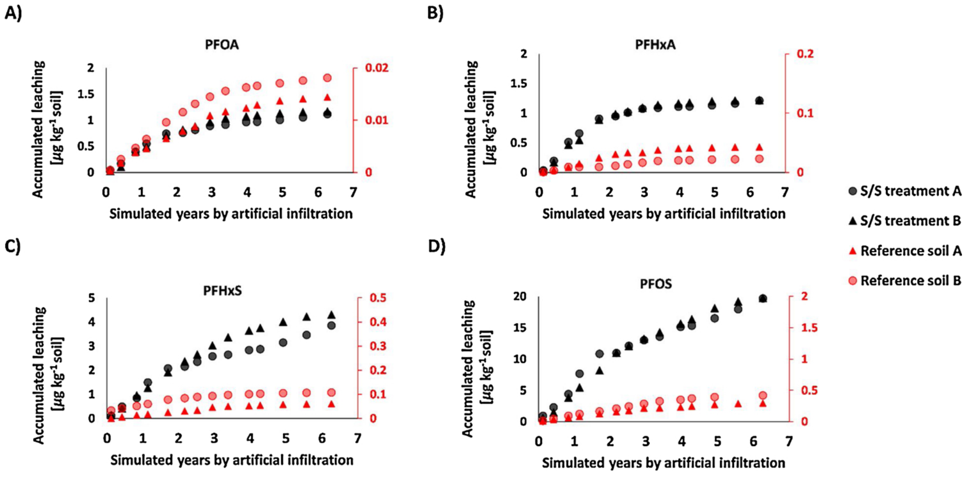

Fig. 2. Accumulated leaching (µg kg^{-1} soil dw) from the pilot-scale experiment during 6 years of simulated rain (15 mm d^{-1}) of: A) PFHxA, B) PFOA, C) PFHxS, and D) PFOS for non-treated reference soil (n = 2, black dots) and stabilization and solidification (S/S)-treated soil (n = 2, red dots). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

EtFOSA) in the experiments, the median recovery of the associated IS was 90 % (Table S5 in SI). Measurement (replicate) error for the triplicate soil extractions ($n = 10$) was on average 45 ± 26 % for the individual PFASs, due to the low concentrations for some PFASs, while it was lower for the PFASs found at higher concentrations (15 %, 11 %, and 28 % for PFHxS, PFOS, and PFOA, respectively). Average regression coefficient (R^2) of the calibration curves was > 0.99 in all cases.

3. Results and discussion

3.1. S/S soil treatment for PFASs in the pilot-scale experiment

After artificial irrigation of 15 mm day^{-1}, representing more than 6 years of precipitation, the concentrations of PFASs were significantly lower ($p < 0.05$) in leachate from the S/S-treated soil than in leachate from the reference soil (Figs. 2 and S2 in SI). In leachate samples from the reference soil (without S/S treatment), 10 of the 18 target PFASs (PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS PFOS, 6:2 FTSA, and FOSA) were detected in concentrations ranging between 0.0026 μg L^{-1} (PFNA) and 32 μg L^{-1} (PFOS). In leachate samples from the S/S-treated soil, seven of the 18 target PFASs (PFHxA, PFHpA, PFOA, PFBS, PFHxS PFOS, 6:2 FTSA) were detected, in concentrations ranging between < MDL and 0.52 μg L^{-1} (PFOS) (except for PFHxS in the first sampling point, with 1.1 μg L^{-1}). In both types of leachate (from reference soil and S/S-treated soil), the PFAS concentration decreased over time (Figs. 2 and S2 in SI). The ΣPFAS concentration in the reference soil leachate decreased from on average 32 μg L^{-1} at the first three sampling points (dominated by PFOS and PFHxS, representing 64 % and 14 % of ΣPFASs, respectively) to on average 9.8 μg L^{-1} at the last three sampling points (dominated by PFOS and PFHxS, representing 78 % and 15 % of ΣPFASs, respectively). In the S/S-treated soil leachate, the average ΣPFAS concentration decreased from on average 0.37 μg L^{-1} at the first three sampling points (dominated by PFOS and PFHxS, representing 50 % and 28 % of ΣPFASs, respectively) to on average 0.26 μg L^{-1} at the last three sampling points (dominated by PFOS and PFHxS, representing 38 % and 4.5 % of ΣPFASs, respectively).

A noteworthy observation was that the proportion of short-chain PFPeA in ΣPFAS increased from 0 to 14% at the first three sampling points to 50 % at the last three sampling points. At the same time, PFPeA removal rate from leachate decreased from > 92 % in the beginning of the experiment to < 3% at the end. In contrast, for other PFASs the removal rate from leachate increased or was stable over time (Fig. S3 in SI). Relatively higher leachability of shorter-chain PFASs has been observed previously in studies analyzing PFASs in drinking water sources (Gobelius et al., 2018). In previous studies, stabilization techniques applied in soil treatment (Sörengård et al., 2019a; Sorengard et al., 2019; Hale et al., 2017b) and water treatment (McCleaf et al., 2017a) have shown low removal rates from leachate for short-chain PFASs. This is in accordance with a previous S/S laboratory-scale PFAS study, where shorter chained PFASs (C < 6) were retained much less than the longer counterparts (Sörengård et al., 2019a) propably hydrophobic sorption mechanism was dominant (Du et al., 2014; Sorengard et al., 2019; Sörengård et al., 2020b; Campos Pereira et al., 2018). The importance of hydrophobic sorption was also observed in stabilization treatment of PAHs, for which the leaching was reduced for low molecular weight homologues e.g. nephtalene (Mulder et al., 2001). The difficulty of stabilizing short-chained PFASs has also been observed for GAC filters in wastewater and drinking water treatment plants (McCleaf et al., 2017b; Rostvall et al., 2018). Other remediation techniques, such as electrokinetic remediation (Sörengård et al., 2019b) and phytoremediation (Gobelius et al., 2017), have been shown to be more promising for removal of shorter-chain PFASs as they partitioning mainly to the aqueous phase. However, in most AFFF-contaminated soils, including that in the present study, short-chain PFASs (e.g., PFPeA) make only a small contribution to total PFAS contamination (Ahrens et al., 2015; Baduel et al., 2015a; Filipovic et al., 2015b). The S/S removal rate from leachate for the other main PFASs (i.e., PFHxA, PFOA, PFHxS, and PFOS) was > 97 % and considerably decreased leaching of ΣPFASs over time (Figs. 2, S3 in SI).

Guideline values in the aquatic environment and in drinking water have only been established for a few PFASs and differ between countries ^{43}. For example, PFOS concentration in leachate from untreated reference soil and S/S-treated soil at the last sampling point in this study was 120-fold and 1.4-fold higher, respectively, than the guideline value for PFOS in groundwater in vulnerable areas (Swedish guideline value 0.045 μg L^{-1}) (Gobelius et al., 2018). On the other hand, PFAS concentration in leachate from untreated and S/S-treated soil at the last sampling point was 84-fold and 1.8-fold higher than the Swedish drinking water guideline value, which is 0.090 μg L^{-1} for Σ_{11}PFASs (of which 10 were assessed in this study, i.e., all except perfluorobutanoic acid (PFBA)) (Gobelius et al., 2017).

3.2. Temporal changes in k_d values for PFASs

The K_d values for PFOS and PFOA in the reference soil ranged between 45 − 59 L kg^{-1} and 17 − 49 L kg^{-1}, respectively, in the first three simulated years. These values were within the same range or slightly higher than those reported in other laboratory-scale partitioning studies (Sörengård et al., 2019a; Sorengard et al., 2019; Hale et al., 2017b; Higgins and Luthy, 2006; Ahrens et al., 2011a). However, in the latter half of the experiment, the K_d values increased three-fold, to over 700 L kg^{-1}, for these two PFASs, without any signs of plateauing (Fig. 3). For the other PFASs, the K_d values increased by 2- to 40-fold in the reference soil, with values of 140, 770, 470, 4200, 5100, 8.3, 65, and 320 L kg^{-1} for PFPeA, PFHxA, PFHpA, PFBS, PFHxS, 6:2 FTSA, FOSA, and EtFOSAA, respectively, at the last sampling point. The increase was significant ($p < 0.05$) for all PFASs except EtFOSA in one experiment. These results of increasing sorption over time support findings in a previous study that PFAS partitioning in the field can be one order of magnitude higher than in laboratory batch tests (Li et al., 2018b). Increased K_d over time is known for other soil micropollutants (e.g., phenathrene and 4-nitrophenol) and is explained by soil aging (Hatzinger and Alexander, 1995). Thus, the more exchangeable fraction of PFASs leaches first, and leaching decreases over time the remaining fraction is more strongly sorbed or less available for mass transfer from soil pores. The increase in partitioning of PFASs to solids was even

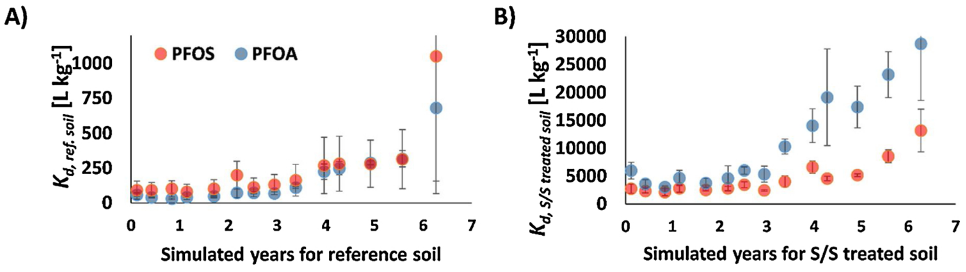

Fig. 3. Estimated average ($n = 2$) partitioning coefficient (K_d [L kg^{-1}]) in the pilot-scale experiment simulating over 7 years of precipitation (15 mm d^{-1}) for PFOA and PFOS in: A) untreated reference (ref.) soil and B) stabilization and solidification (S/S)-treated soil.

larger for the S/S-treated soil, where e.g., partitioning to the solid phase increased 5-fold for PFOS and 8-fold for PFOA. This behavior was observed for all PFASs, but was more prominent for longer-chain PFCAs and PFSAs.

The higher K_d values obtained in the pilot-scale tests compared with the laboratory batch tests have major implications for prediction of long-term environmental behavior. A 1-D leaching prediction model (Eq. (3)) was used to visualize the PFOS concentration in the S/S-treated and untreated soil over time (Fig. 4). A similar prediction model has been used for predicting transport of PFOS in activated carbon-treated soil (Hale et al., 2017b), showing significantly faster leaching of PFOS than in the present study. The difference in predicted leaching behavior of PFASs between this and the previous study (Hale et al., 2017b) may be explained by different assumed K_d values (see Fig. 3).

3.3. Comparing pilot-scale and laboratory-scale S/S soil treatment for PFASs

In this study, we compared leaching of PFASs in two standard laboratory-scale tests and in a pilot-scale experiment using the same reference soil. For the reference soil, there was a significant correlation in leaching behavior of the six main PFASs (PFHxA, PFHpA, PFOA, PFBS, PFHxS, and PFOS) between the laboratory-scale tests (over 26 h, Fig. S4 in SI) and the pilot-scale experiment for 0–5 simulated years (R^2 = 0.89, $p < 0.01$) and 3–6 simulated years (R^2 = 0.98, $p < 0.01$) (Fig. 5A). However, the PFAS leachate concentrations in the laboratory-scale tests were 1–2 orders of magnitude higher than those in the pilot-scale experiment, while the variation was larger in the pilot-scale experiment.

For the S/S-treated soil, there was also a significant correlation in leaching behavior of the eight main PFASs (PFPeA, PFHxA, PFHpA, PFOA, PFBS, PFHxS, PFHxS, PFOS and 6:2 FTSA) between the laboratory-scale tests (over 39 days, see Fig. S5 in SI) and the pilot-scale experiment for 0–5 simulated years (R^2 = 0.78, p < 0.001) and 3–6 simulated years (R^2 = 0.98, $p < 0.01$) (Fig. 5B). The PFAS concentrations in the laboratory-scale monolith leaching test were of the same order of magnitude as in the pilot-scale leaching experiment during 0–3 simulated years, but 1–2 orders of magnitude lower than in the pilot-scale experiment during 3–6 simulated years.

The laboratory-scale tests revealed differences in PFAS removal with respect to i) additive, ii) binder selection, and iii) binder:soil ratio (Fig. S1 in SI). The largest impact was from use of activated carbon additives, while the effects of binder selection and binder:soil ratio were relatively small. There was no significant relationship between UCS and S/S treatment efficiency (p = 0.18, Fig. S6 in SI), which is in agreement with previous findings on S/S binders and hydrocarbon leaching (Wang et al., 2015). Overall, it can be concluded that laboratory-scale batch leaching and monolith leaching tests can both predict PFAS behavior at pilot scale. However, the PFAS concentrations differed widely between the laboratory-scale tests and the pilot-scale experiment, as observed previously (Li et al., 2018b). The discrepancy between laboratory-scale and pilot-scale results can be explained by different sorption behavior (i.e., K_d values) of PFASs over time (see Section 3.2) and different experimental conditions. Laboratory-scale testing is currently the most commonly used method for evaluating sorption of pollutants to solid media and PFAS sorption mechanisms (Johnson et al., 2007; Campos Pereira et al., 2018), environmental fate (Higgins and Luthy, 2006; Ahrens et al., 2011a; Xiao et al., 2017), leachate removal rates (Ochoa-Herrera and Sierra-Alvarez, 2008), and treatment technique development (Sörengård et al., 2019a). Thus, a better understanding of the sorption behavior of PFASs under realistic conditions is needed.

3.4. PFAS suspect screening

A suspect screening approach was used to assess presence of PFASs (see Section 2.7) not included in target analysis (see Section 2.6). After applying the criteria of mass accuracy, chromatographic retention time plausibility, and presence of MS/MS fragments, and following detailed expert manual inspection of the spectra, seven unique PFASs were tentatively identified in the reference soil contaminated with AFFF and in the leachate from the reference and S/S-treated soil (Fig. 6, for details of the name, molecular formula, structure, retention time, and fragments. see Table S6 in SI). Four of the seven PFASs (PFOS, PFHxS, PFOA, and PFHxA) were already identified in target compound analysis. The three additional PFASs tentatively detected were perfluorohexanesulfonamide (PFHxSA), 3:2 fluorotelomer alcohol (3:2 FTOH), and heptafluoropiperidin butane ($C_9H_{10}F_7NO$). Based on total intensity of the seven tentatively identified PFASs, PFOS and PFHxS dominated in the reference soil and in leachate from the reference soil and S/S-treated soil in the pilot-scale experiment.

Of the three PFASs not included in the target screening, PFHxSA represented a significant proportion of the total PFAS intensity (9% in reference soil, 3% in reference leachate, 5% in leachate from S/S-treated soil). Occurrence in soil of PFHxSA originating from AFFF has been observed previously (Baduel et al., 2015b; D'Agostino and Mabury, 2017; Barzen-Hanson et al., 2017; Kaboré et al., 2018; Martin et al., 2019; Liu et al., 2019), but the C_6-based PFHxSA have been much less studied than the C_8-based FOSA (Ahrens et al., 2015). The compound 3:2 FTOH has been previously included in suspect lists (Trier et al., 2011; Liu et al., 2019), but, to the best of our knowledge, has not been tentatively identified or confirmed in environmental samples. The corresponding FTOHs, 4:2 FTOH and 6:2 FTOH, have been studied extensively in air in e.g., the Arctic (Shoeib et al., 2006) and Europe (Jahnke et al., 2007), and detected in emissions from wastewater treatment plants (Ahrens et al., 2011b). $C_9H_{10}F_7NO$ is a piperidin, and, to our knowledge, this functional group of PFASs has not been identified previously in environmental samples. Thus, more studies are needed to confirm possible presence of these PFASs in environmental samples.

The removal rate for the seven identified PFASs, estimated based on the change in intensity, was > 88 % (on average 92 %) for most PFASs (5 out of 7), which is comparable to the value estimated by target screening. For the tentatively identified 3:2 FTOH and $C_9H_{10}F_7NO$, the average removal rates were lower (11 % and -13 %, respectively). The low removal rate of $C_9H_{10}F_7NO$ from leachate may be explained by its low intensity in all samples (Fig. 6), so the values have relatively high uncertainty. The low S/S leachate removal rate of 3:2 FTOH may be

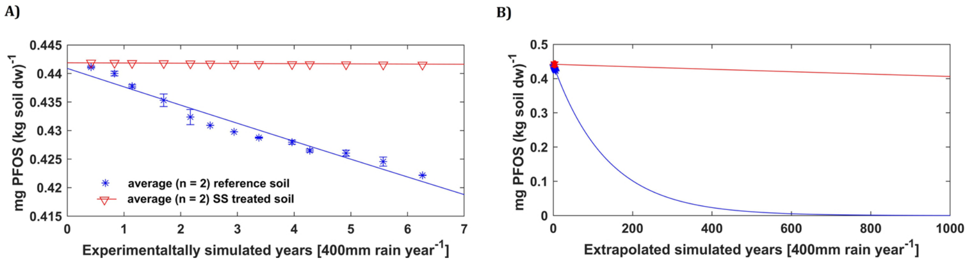

Fig. 4. Concentration of PFOS in soil [mg kg^{-1}] based on: A) artificial irrigation in a pilot-scale leaching experiment representing 6 years of precipitation and using a 1-D regression fit model (Eq. (3)) and B) extrapolation of predicted leaching over time from the stabilization and solidification (S/S)-treated soil (red) and the untreated reference soil (black). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

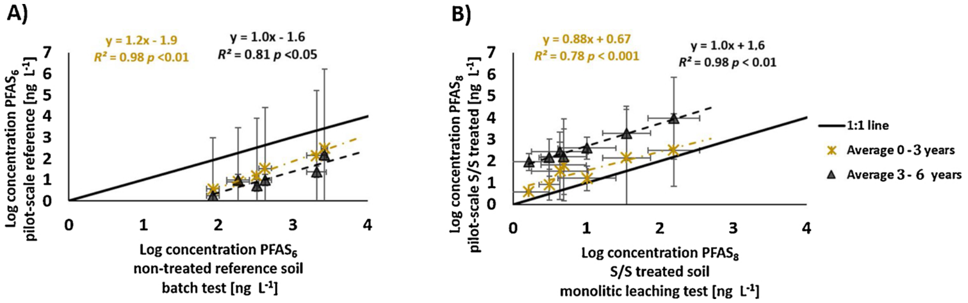

Fig. 5. Correlation between PFAS leaching in: A) the pilot-scale leaching experiment compared with the laboratory-scale leaching batch test for the untreated reference soil, and B) the pilot-scale leaching experiment compared with the laboratory-scale monolith leaching test for the stabilization and solidification (S/S)-treated soil. Individual PFAS concentrations shown are: A) PFHxA, PFHpA, PFOA, PFBS, PFHxS, and PFOS, and B) PFHxA, PFHpA, PFOA, PFBS, PFHxS, PFOS, PFPeA and 6:2 FTSA. Brown stars represent the correlation with 0-3 simulated years, black triangles represent the correlation with 3-6 simulated years in the pilot-scale experiment.

explained by its short perfluorocarbon chain length (C_3 perfluoroalkyl) (Sörengård et al., 2019a).

3.5. Technology limitations

Although S/S treatment has been shown to be a cost efficient and versatile remediation technique, some countries (e.g. Denmark and South Korea) have not recognize S/S treatment as a viable treatment since they required that contaminates are completely removed (Shen et al., 2019). There also remain challenges in determining target values for defining treatment success for regulators, for example guidelines for PFASs in soil depends on the soil usage in Australia and New Zealand (Heads of EPAs Australia and New Zealand (HEPA), 2018), or drinking water guideline values which vary between countries (e.g. 70 ng L^{-1} for PFOS and PFOA in USA and 90 ng L^{-1} for Σ_{11}PFASs in Sweden (Gobelius et al., 2018)). Yet another challenge is the uncertainty of very-long-term performance of S/S and other stabilization treatments, whereby modeling studies have assessed its performance over centuries (Ludwig et al., 2000a) and even millennia (Bozkurt et al., 2000b), but only a few studies (Wang et al., 2014) have been performed experimentally. Hence, the authors encourage such studies in order to make long-term assessments of this technique.

4. Conclusions

Treatment of PFAS-contaminated soil by resulted in high long-term removal from leachate of compounds tested in the pilot-scale experiment. Recipe optimization for S/S treatment showed that addition of activated carbon was crucial for efficient removal of PFASs, while type of binder and soil:binder ratio had smaller impacts on contaminant removal rate from leachate. The removal rate was generally high for PFASs with perfluorocarbon chain length > 6, over 98 % after 6 years simulated precipitation using irrigation. For S/S-treated soil, these high removal rates resulted in concentrations close to groundwater and drinking water guideline values for PFOS and other PFASs. PFAS sorption strength increased significantly over time, by a factor of 2–40. Laboratory-scale tests showed good correlations with the pilot-scale experiment, but sorption of PFASs in S/S-treated soil was 1–2 orders of magnitude stronger in pilot-scale than in laboratory-scale testing, demonstrating the importance of pilot-scale testing. In suspect screening, three PFASs were tentatively identified for the first time in environmental samples. Two of these (3:2 FTOH and $C_9H_{10}F_7NO$) had removal efficiency < 12 %, and should be considered in future analyses of AFFF-polluted environments.

CRediT authorship contribution statement

Mattias Sörengård: Conceptualization, Methodology, Data curation, Writing - original draft, Visualization, Investigation. **Dan Kleja Berggren:** Conceptualization, Methodology, Supervision, Writing - review & editing. **Lutz Ahrens:** Conceptualization, Methodology, Supervision, Writing - review & editing.

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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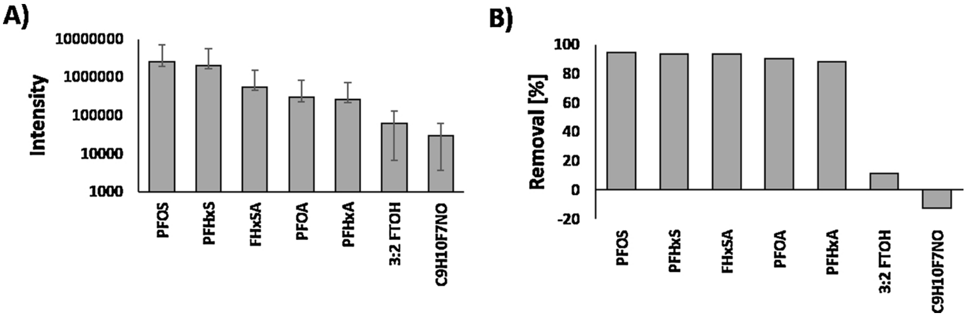

Fig. 6. A) Average intensity of different PFASs detected in the reference soil (n = 2) using suspect screening analysis, and B) leachate removal rate [%] based on intensity of the detected PFASs in the aqueous phase from the last sampling point in the pilot-scale experiment (n = 2).

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2020.123453.

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