Leaching and transport of PFAS from aqueous film-forming foam (AFFF) in the unsaturated soil at a firefighting training facility under cold climatic conditions

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ABSTRACT

The contaminant situation at a Norwegian firefighting training facility (FTF) was investigated 15 years after the use of perfluorooctanesulfonic acid (PFOS) based aqueous film forming foams (AFFF) products had ceased. Detailed mapping of the soil and groundwater at the FTF field site in 2016, revealed high concentrations of *per*- and polyfluoroalkyl substances (PFAS). PFOS accounted for 96% of the total PFAS concentration in the soil with concentrations ranging from < 0.3 μg/kg to 6500 μg/kg. The average concentration of PFOS in the groundwater down-gradient of the site was 22 μg/l (6.5–44.4 μg/l), accounting for 71% of the total PFAS concentration. To get a better understanding of the historic fate of AFFF used at the site, unsaturated column studies were performed with pristine soil with a similar texture and mineralogy as found at the FTF and the same PFOS containing AFFF used at the site. Transport and attenuation processes governing PFAS behavior were studied with focus on cold climate conditions and infiltration during snow melting, the main groundwater recharge process at the FTF. Low and high water infiltration rates of respectively 4.9 and 9.7 mm/day were applied for 14 and 7 weeks, thereby applying the same amount of water, but changing the aqueous saturation of the soil columns. The low infiltration rate represented 2 years of snow melting, while the high infiltration rate can be considered to mimic the extra water added in the areas with intensive firefighting training. In the low infiltration experiment PFOS was not detected in the column leachate over the complete 14 weeks. With high infiltration PFOS was detected after 14 days and concentrations increased from 20 ng/l to 2200 ng/l at the end of the experiment (49 days). Soil was extracted from the columns in 5 cm layers and showed PFOS concentrations in the range < 0.21–1700 μg/kg in the low infiltration column. A clear maximum was observed at a soil depth of 30 cm. No PFOS was detected below 60 cm depth. In the high infiltration column PFOS concentration ranged from 7.4 to 1000 μg/kg, with highest concentrations found at 22–32 cm depth. In this case PFOS was detected down to the deepest sample (~90 cm).

Based on the field study, retardation factors for the average vertical transport of PFOS in the unsaturated zone were estimated to be 33–42 and 16–21 for the areas with a low and high AFFF impact, respectively. The estimated retardation factors for the column experiments were much lower at 6.5 and 5.8 for low and high infiltration, respectively. This study showed that PFOS is strongly attenuated in the unsaturated zone and mobility is dependent on infiltration rate. The results also suggest that the attenuation rate increases with time.

1. Introduction

Aqueous film-forming foam (AFFF) containing *per*- and polyfluoroalkyl substances (PFAS) has been used extensively since the first development by 3M, Ansul and National Foam Companies in the mid 1960's (Place and Field, 2012). AFFF has surface-tension lowering properties and spreads rapidly across the surface of hydrocarbon fuels,

cooling the liquid fuel by forming a water film beneath the foam, resulting in superior firefighting capabilities (Schaefer et al., 2008). The use of AFFF has resulted in PFAS and especially perfluorooctanesulfonic acid (PFOS) contamination of soil, groundwater, surface waters and biota worldwide (Ahrens et al., 2015; Anderson et al., 2016; Filipovic et al., 2015; Houtz et al., 2013).

There is a growing concern for negative consequences for the

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environment and human health with regards to use and exposure to PFAS (Blume et al., 2015; Cousins et al., 2016; KEMI, 2016; Lindstrom et al., 2011). The persistency, bioaccumulation and toxicity of long-chain PFASs define them as persistent organic pollutants (POPs) and those of most concern are listed as substances of very high concern (SVHC) in the European Chemical Agency's (ECHA) (Blume et al., 2015; Brendel et al., 2018).

Fate of PFAS released to the soil environment is primarily dependent on infiltration and sorption to the solid matrix. Partitioning coefficients for PFOS and other PFASs to soil and sediments have been thoroughly investigated and reported in literature (Gellrich et al., 2012; Hale et al., 2017; Hellsing et al., 2016; Higgins and Luthy, 2006; Johnson et al., 2007; Zareitalabad et al., 2013). Recent studies have shown that sorption to the air-water interface can be a major contributor to the retention of PFOS and PFOA under partially saturated conditions (Brusseau, 2018; Lyu et al., 2018).

The Norwegian Aviation Organization (Avinor) has made an inventory of PFAS contaminated soil, groundwater, surface water and biota at several firefighting training facilities (FTF) throughout Norway. The studies have shown widespread leaching of PFAS from the soil to nearby water courses and exposure of biota in both fresh water and marine environments (Avinor Miljøprosjekt DP2, 2012). This study focuses on one specific FTF where AFFF containing PFOS has been used extensively since the early 1990's until it was phased out in 2001 and replaced by fluorotelomer containing AFFF (KEMI, 2015). All use of PFAS containing firefighting foams was banned at the airport in 2011 (Avinor Miljøprosjekt DP2, 2012).

The aviation authorities have raised the question how much of the PFOS is still present in the source zone after 15 years without PFOS application. Can these residual levels form a long-term source, potentially contaminating the groundwater at the site?

To answer this question, extensive field studies at the FTF site were performed to map the present PFAS contamination situation in the unsaturated soil profile, and the groundwater. To reconstruct the initial release scenario of AFFF used at this site, unsaturated column studies were performed under environmentally relevant experimental conditions. This model study allowed to compare field observations with an unknown contamination history with well controlled exposure of a similar soil. The objective was to improve our understanding of the contamination history and potential transport and attenuation processes governing PFAS behavior at this site.

To our knowledge this is the first study where the PFAS contamination history at a FTF site has been studied in a controlled unsaturated column experiment using a complete AFFF mixture.

2. Materials and methods

2.1. Site description

The FTF investigated in this work has a total size of 25,000 m^2 and was established in 1989–1990. Extensive use of AFFF at the site started after the airport opened for civil aviation in 1998, but the area has previously been used for firefighting training activities. There are six firefighting training platforms at the FTF, each designed with membranes and a collection system for fuel, water and firefighting foam to protect the underlying groundwater. Most of the liquids used at the site have been collected and discharged to the local sewage system. Foam and water have also spread to the soil outside the training areas, due to wind and the increased spraying range of modern firefighting engines. Soil and groundwater sampling in 2008 revealed PFAS contaminated soil outside the collection areas and leaching to the groundwater 4 m below the surface (Norwegian Pollution Authority, 2008). The amount of AFFF spread outside the collection areas and the water infiltration rate has varied across the FTF. In some AFFF source zones, there has been extensive use of water during firefighting training activities, resulting in high infiltration, while in other parts of the FTF infiltration

has been limited to the yearly precipitation, which is dominated by snow melting.

2.2. Infiltration at the FTF site

The yearly precipitation in the area of the FTF is approximately 800 mm (Jørgensen and Østmo, 1990). The soil at the site is a uniform medium fine sand over the whole unsaturated zone down to the groundwater at 4 m depth, with an organic carbon content below 1%. The hydraulic conductivity (Ks) at the site ranges from 10^{-3} to 10^{-5} m/s (French et al., 2009). The water balance developed by Jørgensen and Østmo (1990) for the area, showed that 50% (400 mm) of the annual precipitation is lost due to evapotranspiration and close to 60% (240 mm) of groundwater recharge is occurring during a 3–5 week long snowmelt period in spring, while the remainder of the annual infiltration occurs during the autumn months. It was estimated that water infiltrates through the unsaturated zone during the snowmelt period with a mean vertical velocity of 20 cm/day at 20% saturation (Jørgensen and Østmo, 1990). French et al. (1999) showed that the mean vertical velocity of infiltrating water in the springtime snowmelt period was approximately 5.2 mm per mm infiltration at an estimated saturation level between 18.5 and 20.8%. Infiltration was lower during the autumn and the vertical pore water velocity in the soil was estimated to be 7.7 mm/mm. In the summer months precipitation was balanced by evapotranspiration with no net infiltration (French et al., 1999).

2.3. Site investigation

2.3.1. Soil

Soil and groundwater investigation was performed at the FTF site in 2016, and included soil sampling at 80 locations in the unsaturated zone around the firefighting training platforms. Trial pits were excavated for sampling of the soil profile. All equipment was rinsed with methanol before sampling each point. Samples were collected from 0 to 1 m, 1–2 m, 2–3 m and from 3 m and down to the groundwater level at 4 m. Soil was transferred into sampling bags (1 l polyamide) and stored at 4 °C in the laboratory before being shipped to a commercial laboratory for analysis. A total of 288 soil samples were analyzed for PFAS content.

2.3.2. Groundwater

Groundwater was sampled at 5 pumping wells installed as part of a pump and treat remediation system down gradient of the site to intercept the plume spreading from the FTF. A total of 19 sampling campaigns were performed during 2016. The samples were stored in HDPE bottles at 4 °C before being shipped to a commercial laboratory for PFAS analysis.

2.4. Column set up

Pristine soil from an area close to the FTF, with similar texture and mineralogy, was collected and used to construct columns for the infiltration experiment. Acrylic glass columns with an inner diameter of 14 cm were equipped with a water drain at the bottom, consisting of a coarse metal grid covered by a fine stainless steel filter to prevent the sand from washing out (Fig. 1).

The columns were packed with sand by adding layers of 6 cm, compacting each layer with a 900 g weight dropped four times until each column had a total length of approximately 1 m. The total weight of the sand added to each column was recorded. The sand was analyzed for grain size distribution, water content, total organic carbon (TOC) and background levels of PFAS. The columns were placed in a temperature controlled room at 10 °C. A water reservoir (2l) at the top of each column was used for infiltration. The reservoir emptied through 25 needles to ensure uniform distribution over the whole surface area of

Fig. 1. a)The coarse and fine filter at the bottom of the colums; b)The top water reservoir with 25 needles to distribute the infiltration; c)The column set up.

the column, imitating natural infiltration of rain droplets. The soil surface in the columns was approximately 30 cm below the outlet of the needles. The flow through the columns was gravity driven to simulate field conditions (French et al., 1999). Leachate was collected and sampled at the bottom of the columns in plastic buckets.

2.5. Column infiltration rates

The experiments were performed at low and high infiltration rates of 4.9 mm and 9.7 mm per day for respectively 14 and 7 weeks. A total of 477 mm water was added in both the low and high infiltration experiment. The water was weighed and manually poured into the top reservoir and allowed to drip on the soil, 3 times per week.

2.6. Non-reactive tracer test

Non-reactive tracer (NRT) experiments were performed in the columns to estimate the unsaturated water transport through the columns at low and high infiltration rates by adding a solution of NaCl in the top reservoir during steady-state water infiltration at the stated rates. Leachate was sampled 3 times per week and analyzed for temperature, pH, and electrical conductivity (EC).

2.7. AFFF experiment

AFFF concentrate, containing PFOS as the main PFAS, from the same supplier as assumed to have been used historically at the site was used in the experiments. The AFFF concentrate was mixed with Milli-Q water in a 1:100 ratio and whipped to a stable foam, which was then applied to the soil surface of both columns. The 1:100 diluted AFFF concentrate was analyzed to determine the exact PFAS composition (see Appendix A, Table A1).

When applied, the AFFF was a dense foam (Fig. 2a). 24 h after the

first infiltration of water, the foam had disappeared from the surface, but foam bubbles could still be observed in the voids of the upper part of the soil columns (Fig. 2b).

2.8. Column leachate sampling

The infiltration period was 14 and 7 weeks respectively for the low and high infiltration experiments. Leachate samples were collected 3 times per week for the low infiltration experiment, and daily for the high infiltration experiment. For leachate volumes < 50 ml, samples were combined with the sample from the subsequent day to ensure sufficient sample size for analytical requirements. The samples were stored at 4 °C before shipping to a commercial laboratory for PFAS analysis.

2.9. Column soil sampling

After the infiltration experiments were completed, the soil was extracted from the columns in 5 cm intervals and analyzed for PFAS content. The water content was measured in the soil samples after the experiment to quantify and confirm the unsaturated conditions in the soil columns. The samples were stored at 4 °C before shipping to a commercial laboratory for analysis.

2.10. Chemical analysis

The list of target PFAS analyzed varied between the field study and the laboratory experiments. The field samples were analyzed for 12 PFAS compounds for soil and groundwater. In the column study 30 PFAS compounds were analyzed in soil and 23 PFAS compounds were analyzed in leachate and the AFFF foam. For a complete overview of the compounds analyzed, see Appendix A and C. Soil and leachate analyses were carried out at the accredited laboratory Eurofins GfA Lab Service

Fig. 2. a) The AFFF foam added to the soil columns. b) Foams bubbles visible in the soil after 24 h of water infiltration.

GmbH (in Germany), using method DIN 38414-S14,(DIN 38414-14, 2011) based on acetonitrile extraction followed by analysis using liquid chromatography coupled with mass spectrometry (LC/MS-MS) for soil samples and DIN 38407-F42, (DIN 38407-42, 2011) and quantification using LC/MS-MS for leachate samples.

2.11. Quality control and assurance

Following the NRT experiments, the infiltration reservoirs were thoroughly cleaned, and all needles changed. No material that could influence PFAS sorption behavior (glass, metal) was used when handling samples from the AFFF experiment. HDPE bottles were used to store samples until analysis. Reference samples of the soil used in the columns and water used for infiltration were analyzed to determine the background levels of PFAS. PFAS analysis was carried out at an accredited laboratory (Deutsche Akkreditierungsstelle GmbH, following DIN EN ISO/IEC 17025:2005). Internal isotopically labelled standards were added to all soil and leachate 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 varied from 0.2–1 µg/kg for the respective PFAS in soil and was 0.3 ng/l for each PFAS in leachate.

3. Results and discussion

3.1. PFAS concentration in soil at the FTF field site

PFAS concentrations found in the soil samples at the FTF varied from < 0.3 to 6500 µg/kg. PFOS accounts for 96% of the Σ 12-PFAS analyzed in the soil samples and is therefore focused on in the following discussion of source zones with high and low AFFF impact.

3.1.1. Source zones with low AFFF impact

In source zones with low impact of AFFF, the soil sampled from 0 to 1 m depth revealed concentrations of PFOS in the range of 100 to 900 µg/kg. The soil samples from 1 to 2 m depth contained much lower concentrations of PFOS from < 0.3–70 µg/kg. The PFOS concentrations at sampling locations with low AFFF impact are given in Fig. 3a. The soil was not analyzed below 2 m due to low PFOS concentrations in the soil samples from 1 to 2 m.

3.1.2. Source zones with high AFFF impact

The PFOS concentrations in the unsaturated soil in source zones with high impact of AFFF are shown in Fig. 3b. Concentrations of PFOS in the soil from 0 to 1 m were in the range of 500 to 3000 µg/kg. The

soil from 1 to 2 m revealed the highest PFOS concentrations, ranging from 1000 to 6500 µg/kg. From 2 to 3 m the PFOS concentrations ranged from 1000 to 3500 µg/kg, with a further reduction 1000 to 1200 µg/kg at 3–4 m depth (not all samples analyzed). Differences between the low and high impacted areas might indicate that the attenuation and transport processes of PFOS differ at these sites.

The concentration of PFOS in the soil profiles are in the same order of magnitude as reported previously at FTF sites in Sweden, Australia and the US (Baduel et al., 2017; Filipovic et al., 2015; McGuire et al., 2014). In France Dauchy et al. (2019) took 44 soil cores at a FTF belonging to an abandoned refinery, where activities probably ceased in 1984. The median concentration at the highest impacted areas was respectively 8701 and 12,112 µg/kg (Σ 34-PFAS). The highest concentrations were found in the top 1 m and 50% to > 99% of the PFAS content was identified as fluorotelemers, dominated by 6:2 fluorotelomer sulphonamide alkylbetaine (6,2 FTAB). Perfluorosulfonates represented < 1 to 46% of the PFAS in the soil, dominated by PFOS. PFOS precursors were in general below < 1% of the quantified PFAS.

3.2. PFAS concentration in the groundwater at the FTF field site

The groundwater concentration varies between the 5 pumping wells (Fig. 4a). The yearly average concentration for PFOS in the wells was 22 µg/l (6.5–44.4µg/l), accounting for 71% of the Σ 12-PFAS concentration (31.9 µg/l). The average concentrations of PFHxS and 6:2 FTS were 2.9 µg/l and 2.7 µg/l, respectively, accounting for 9 and 8% of the Σ 12-PFAS concentration in the groundwater. Other PFAS detected in the groundwater represented approximately 12% of Σ 12-PFAS quantified in the groundwater (Fig. 4b). The PFAS concentration in the groundwater varies during the year, as can be seen by the high standard deviations in the yearly average concentrations presented in Fig. 4b.

Filipovic et al. (2015) reported similar values at a Swedish airport, PFOS up to 42 µg/l was reported, representing > 80% of the 4 PFAS analyzed. At a US military site a maximum of 78 µg/l PFOS was found as well as PFHxS (360 µg/l), PFHxA (350 µg/l), 6:2 FTS (220 µg/l), PFOA (220 µg/l), PFBS (150 µg/l), and 120 µg/l PFPeA (Backe et al., 2013). Other studies have reported considerably lower groundwater concentrations. Dauchy et al. (2019) found PFAS concentrations in the range of 4 to 8277 ng/l, where perfluorosulfonates represented 16 to 100% of Σ 34-PFAS dominated by PFHxS and PFOS. The remainder were perfluorocarboxylic acids (C4-C8), 6:2 FTS and 6:2 FTAB.

3.3. Results from non-reactive tracer column study

The non-reactive tracer tests (NRT) in the unsaturated column study showed the first breakthrough of the tracer after 28 and 18 days, at low
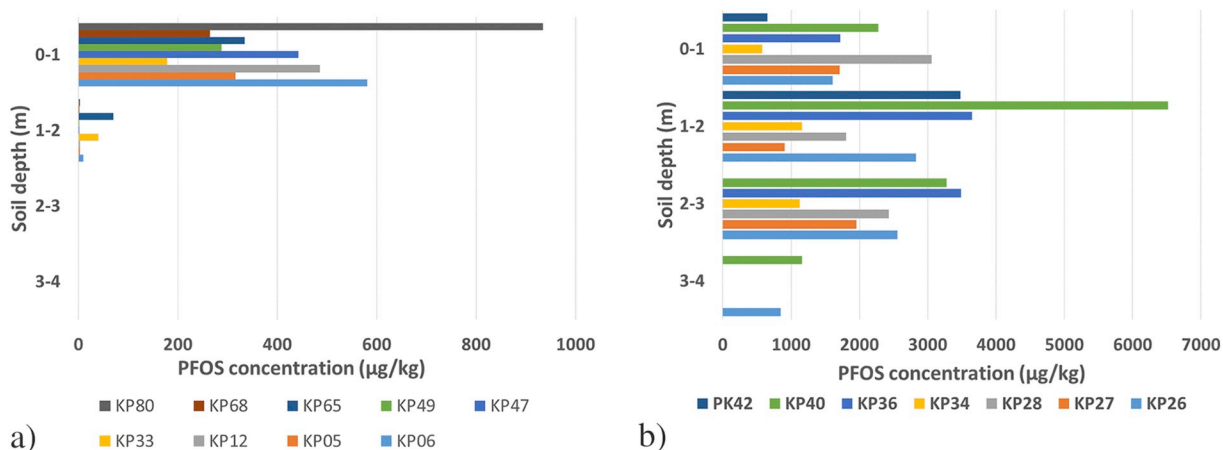

Fig. 3. a) PFOS concentrations in 9 soil profiles in source zones with low AFFF impact b) PFOS concentrations in 7 soil profiles in source zones with high AFFF impact.
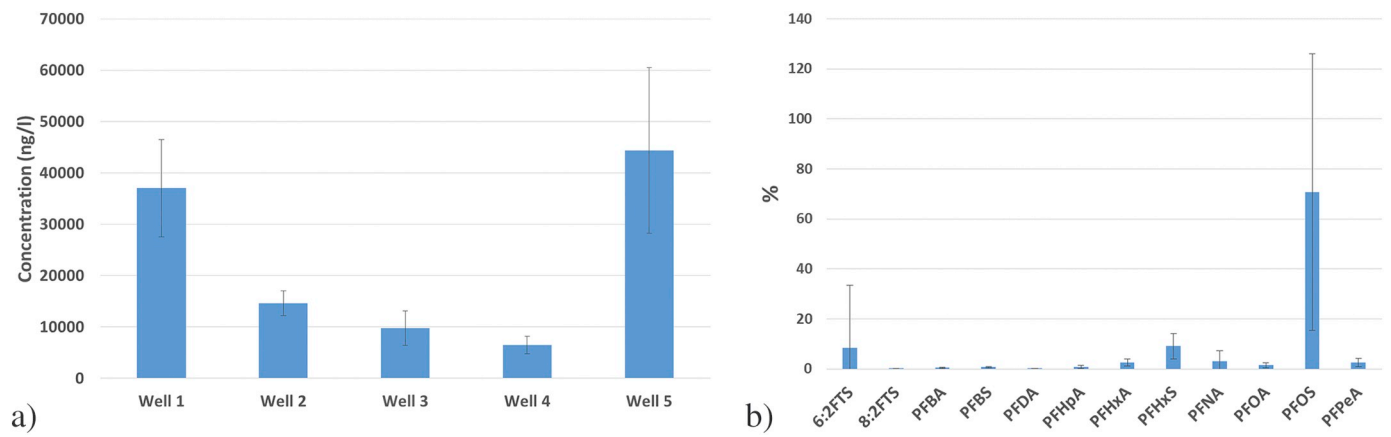
Fig. 4. a) Average PFOS concentrations sampled 19 times in each of the 5 pumping wells downgradient the FTF during 2016. b) Relative distribution of PFAS in groundwater sampled in 2016.
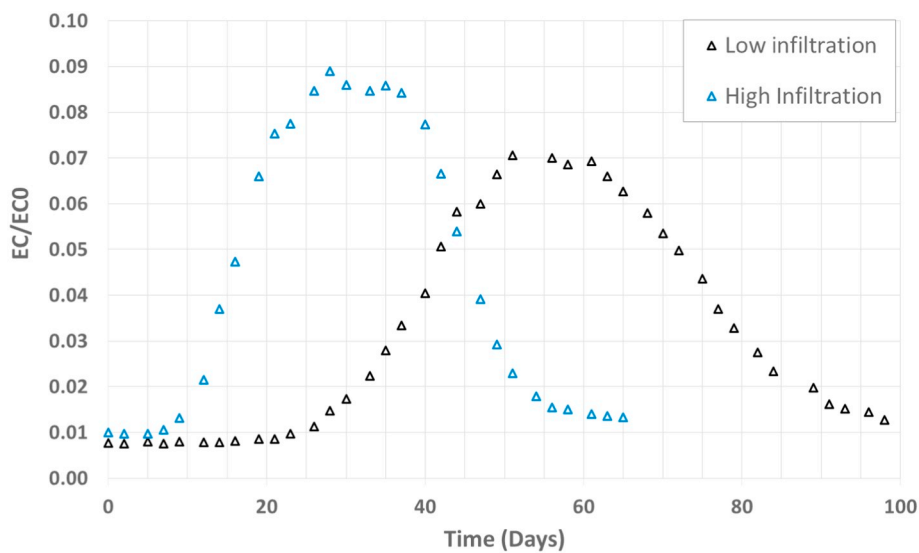
Fig. 5. Relative electrical conductivity (EC/EC$_0$) in leachate from the column studies at low and high infiltration rates.
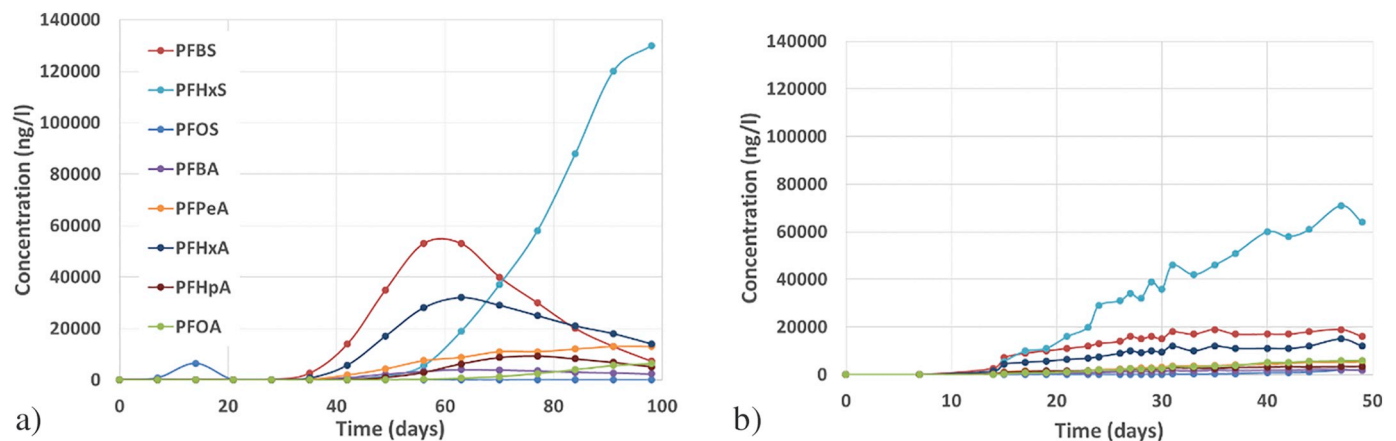
Fig. 6. PFAS concentrations in column leachate for low (a) and high (b) infiltration rates.

and high infiltration rates, respectively. A maximum electrical conductivity was measured in the leachate after 56 and 28 days, as shown in Fig. 5. The results showed average water flow velocities during the low and high infiltration experiments of 1.7 cm/day and 3.2 cm/day respectively. The observed pore water velocities in the columns are in the same order of magnitude as estimated for snow melting by French

et al. (1999). This shows that the hydrological behavior of our re-packed columns is representative of the field conditions at the site.

3.4. Concentration of PFAS in the AFFF foam applied

PFOS is the most dominant constituent of the Σ 23-PFAS determined

in the AFFF used in the column experiments, with a concentration of 100 mg/l after dilution, accounting for 90% of the Σ 23-PFAS amount applied. The other PFASs detected in amounts larger than 0.5% of the Σ 23-PFAS amount in the diluted concentrate are PFHxS contributing with 6.3%, PFBS with 1.2%, PFHpS with 1.4%, PFHxA with 0.7% and PFOA with 0.8% of the Σ 23-PFAS amount in the diluted AFFF. The PFAS concentrations measured in the diluted AFFF concentrate (1:100 with Q-water) are presented in appendix A.

3.5. Concentration of PFAS in the column leachate

3.5.1. Low infiltration experiment

The concentrations of PFAS detected in the column leachate for the low infiltration experiment are shown in Fig. 6a. PFBS shows a first breakthrough after 14 days, reaches a maximum concentration of 53 µg/l after 56–63 days and decreases thereafter. It seems to move through the column without retardation, at a rate similar to that observed for the NRT. Subsequently a breakthrough and decrease of PFHxA, PFPeA and PFHpA are seen. PFHxS, by definition a long-chain PFAS (Buck et al., 2011), is not attenuated to the same degree as other long-chained PFASs in the column soil. PFHxS is showing a breakthrough after 35 days and reaches a maximum concentration of 130 µg/l at the end of the experiment at 98 days (PFOS, PFHxS, PFHpS and PFOA were detected surprisingly in the 7 and 14 days samples). 100% of the initial amount of PFBS added to the column has been detected in the leachate at the end of the experiment while 45% of PFHxS and 29% of PFHpS added has leached through the column. PFOS was not detected over 15 ng/l during the experiment (disregarding the early detection most likely due to error). 6% of the Σ 23-PFAS in the AFFF applied had leached through the column at the end of the experiment, but only 0.006% of the PFOS in the applied AFFF.

3.5.2. High infiltration experiment

PFAS concentrations detected in the leachate for the high infiltration rate experiment are shown in Fig. 6b. The first detection of any PFAS was after 7 days but the onset of a breakthrough can be observed from 14 days and onwards. PFBS, PFHxA and PFHxS seem to breakthrough simultaneously. However, PFHxS is the most dominant compound in the column leachate thereafter, similar to the low infiltration experiment. The maximum PFHxS concentration in the leachate is 71 µg/l after 47 days (approx. 50% lower than in the low infiltration column). PFBS and PFHxA reach an apparently stable concentration at 17 and 15 µg/l respectively. Considerably lower than observed in the low infiltration column. PFOS concentrations are difficult to observe in Fig. 6b, but PFOS was detected in the leachate after 21 days at a concentration of 20 ng/l, which continued to increase to 2200 ng/l at the end of the experiment. This is a major difference between the two experiments since PFOS was not detected above 15 ng/l in the low infiltration experiment. 87% of the total PFBS amount had leached through the column at the high infiltration rate in contrast to 100% at low infiltration. PFHxS leaching was comparable in both experiments at 47% of the added amount. In contrast only 2% of PFHpS leached out at high infiltration compared to 29% at low infiltration. The total amount of PFOS leached in the low infiltration experiment was 0.006% while it was 0.05% in the high infiltration experiment. For the sum detected PFAS in the leachate the amount leached was slightly lower for the low infiltration experiment with 5.89% compared to 5.93% in the high infiltration experiment of Σ 23-PFAS added.

3.6. Distribution of PFAS in the soil columns

3.6.1. Low infiltration experiment

PFAS analysis of the soil in the column at the end of the experiment showed that short-chain PFAS were less attenuated than long-chained PFAS, as presented in Figs. 7a and 8a. PFOS was the most retarded PFAS at low infiltration with concentrations ranging from 0.21–1700 µg/kg

in the soil and a maximum concentration detected at 30 cm depth in the soil columns. Previous column studies have not analyzed remaining PFAS concentrations in the soil matrix. However, similar retention patterns have been reported, where short chain PFAS are less retarded in the soil than long chained PFAS (Vierke et al., 2014; Gellrich et al., 2012).

3.6.2. High infiltration experiment

The PFAS analysis of the soil in the column at the end of the high infiltration experiment showed, similar to the low infiltration experiment, that short-chain PFAS were attenuated less than long-chained PFAS (Figs. 7b and 8b). The PFOS concentration in the soil ranged from 7.4 to 1000 µg/kg with the highest concentrations observed in a zone at 22–32 cm depth. The highest PFOS concentration was approximately 60% of the maximum concentration in the low infiltration experiment. This might be a consequence of a reduced pore water concentration at the high infiltration rate, resulting in reduced sorption to the soil matrix. The presence of PFOS in the column leachate is indicative of reduced sorption and increased vertical transport.

3.7. Comparison of retardation factors in field and column experiments

The retardation factors for PFOS in the unsaturated zone at the FTF are estimated based on an assumed average vertical water velocity from snowmelt infiltration of 4.9–7.5 cm/day during 3 weeks (200-300 mm). In addition autumn infiltration of 160 mm precipitation with an infiltration rate of 7.7 mm/mm, results in an estimated yearly vertical pore water transport of (227–280 cm) (French et al., 1999). For the low impact areas, PFOS is mainly found at 1 m depth. Assuming PFOS use ended 15 years ago, this results in an average yearly vertical PFOS transport rate of 6.7 cm/year during the 15 years that have passed. From these data a retardation factor of 33–42 can be estimated. For the high impact areas, the yearly average vertical velocity might be higher due to the extra water added during firefighting activities. Assuming the same average vertical water velocity as for low impact areas vertical transport for PFOS down to 2 m is observed, resulting in a PFOS transport rate of 13.3 cm/year and a retardation factor of 16–21 for PFOS at high impact areas.

In the soil columns, the vertical distance travelled for the center of mass of PFOS in the soil columns was 25.5 and 27 cm, respectively for the low and high infiltration experiment. This results in mean PFOS transport rates of 0.26 cm/day and 0.55 cm/day, respectively. The NRT showed average vertical water velocities of 1.7 cm/day and 3.2 cm/day for low and high infiltration, respectively. This gives retardation factors of 6.5 and 5.8 at low and high infiltration rate, respectively. Based on these retardation factors and assuming a mean volumetric water content of 20%, apparent distribution coefficients (K_D) for PFOS of 4.0–5.1 l/kg and 1.9–2.5 l/kg can be estimated for low and high impacted areas at the FTF, respectively. Apparent K_D values of 0.8 and 0.7 l/kg for the column studies with low and high infiltration, respectively, can be estimated with the same volumetric water content as for the field estimates. These K_D values are in the range of values reported for PFOS in literature (Gellrich et al., 2012; Hale et al., 2017; Hellsing et al., 2016; Higgins and Luthy, 2006; Johnson et al., 2007; Zareitalabad et al., 2013) compiled in Appendix B. Zareitalabad et al. (2013) reports K_D values for the sorption of PFOS to various soils and sediments in the range of < 1 and 35.3 l/kg. For the sandy soil in our study the value reported for Ottowa sand of 2.8 l/kg could be expected (Johnson et al., 2007).

The lower retardation factors for PFOS in the column experiments compared to those estimated for field conditions at the FTF might be explained by the fact that the column infiltration was continuous in a relatively short time period compared to the field conditions, where infiltration will be intermittent. At the FTF there has only been net infiltration in the unsaturated zone during annual snowmelt and autumn precipitation. Long periods with stagnant pore water during
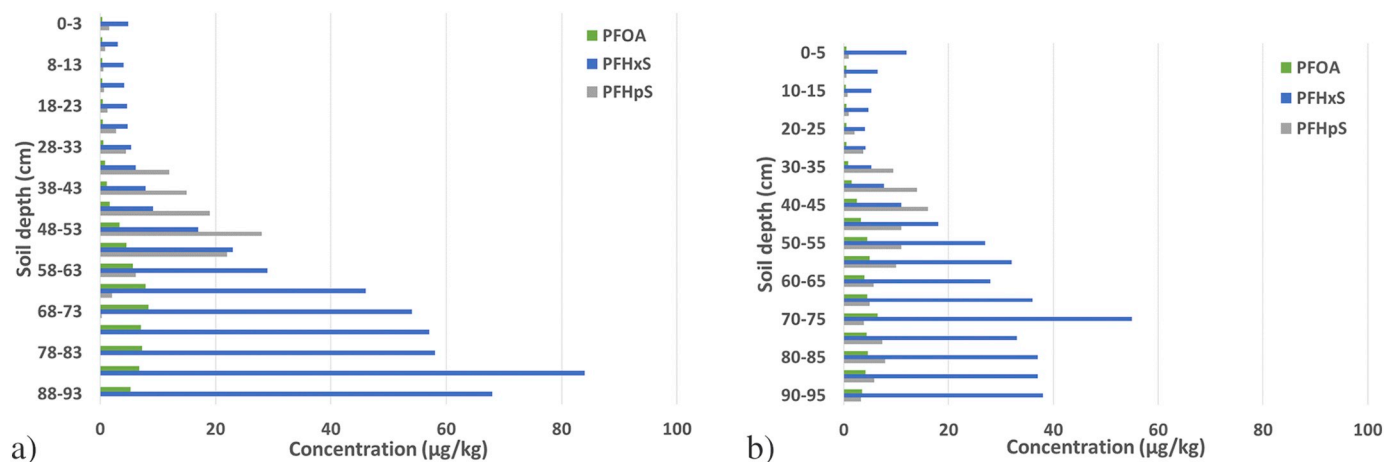
Fig. 7. PFOA, PFHxS and PFHpS concentration in the column soil for the low (a) and high (b) infiltration experiments.

summer and winter months are observed where the saturation is at field capacity and no groundwater recharge occurs (French et al., 1999). The annual changes in saturation level of the soil pores can have an effect on the attenuation processes for PFOS in the unsaturated soil under field conditions. Both sorption kinetics and the potential effect of disequilibria complicate the comparison of the field and column study results but were not studied in this work (Wei et al., 2017). Brusseau (2018) showed that retention processes other than sorption to the solid-phase can influence PFAS transport. Air-water interface adsorption alone accounted for > 50% of the total retention observed in these studies. The adsorption of PFOA to the air-water interface during transport in unsaturated porous media was further investigated by Lyu et al. (2018) and the results for the experiments showed that adsorption to the air-water interface was a significant source of retention contributing to approximately 50–75% of the total observed retention of PFOA. The potential contribution of this process to PFOS retention requires further attention.

4. Conclusion

The use of AFFF at the FTF in this study has resulted in PFAS and especially PFOS contamination of the soil. At high impacted areas soil contamination down to the groundwater level at 4 m below surface could be observed.

Unsaturated column studies with AFFF applied to pristine soil, to better understand the historic contamination history, showed that when exposed to low infiltration rates, PFOS was not detected in the column leachate, while exposed to high infiltration rates, PFOS was detected in increasing concentrations up to 2200 ng/l during the experimental period. Estimated retardation factors for PFOS in the field were 33–42 and 16–21 for low and high impacted areas compared to 6.5 and 5.8 for low and high infiltration column studies.

The leaching of PFAS from source zones in the unsaturated zone at this site can represent a long-term risk for contamination of the groundwater and transport to nearby surface water bodies.

Better insight in the retention processes in the unsaturated zone is essential to achieve a more accurate prediction of leaching rates and improve risk assessment and remediation design at PFAS contaminated sites.
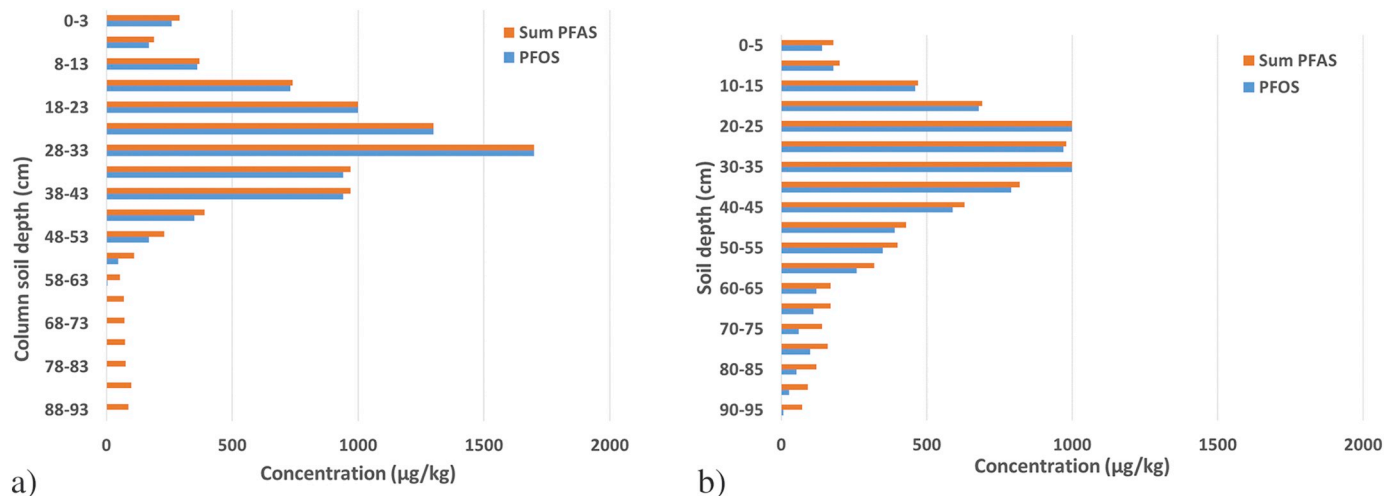
Fig. 8. PFOS and Σ 30-PFAS concentration in the column soil for low (a) and high (b) infiltration experiments.

Appendix

Appendix A

Table A1
PFAS concentrations in 1:100 diluted AFFF and a list of the analyzed compounds.

Compound	Foam conc. ng/l	LOQ ng/l	Relative amount	Field study soil and g.w.	Column study soil	Column study leachate
Perfluorobutane sulfonate (PFBS)	1,400,000	0.3	1.2%	x	x	x
Perfluorohexane sulfonate (PFHxS)	7,100,000	0.3	6.3%	x	x	x
Perfluoroheptane sulfonate (PFHpS)	1,600,000	0.3	1.4%		x	x
Perfluorooctane sulfonate (PFOS)	100,000,000	0.3	88.7%	x	x	x
Perfluorodecane sulfonate (PFDeS)	70,000	0.3	0.1%		x	x
Perfluorobutanoic acid (PFBA)	160,000	0.6	0.1%	x	x	x
Perfluoropentaonic acid (PFPeA)	230,000	0.3	0.2%	x	x	x
Perfluorohexaonic acid (PFHxA)	750,000	0.3	0.7%	x	x	x
Perfluoroheptaonic acid (PFHpA)	300,000	0.3	0.3%	x	x	x
Perfluorooctaonic acid (PFOA)	900,000	0.3	0.8%	x	x	x
Perfluorononaonic acid (PFNA)	3100	0.3	< 0.1%	x	x	x
Perfluorodecaonic acid (PFDeA)	1500	0.3	< 0.1%	x	x	x
Perfluoroundecanoic acid (PFUnA)	470	0.3	< 0.1%		x	x
Perfluorododecanoic acid (PFDoA)	540	0.3	< 0.1%		x	x
Perfluorotridecanoic acid (PFTrDA)	120	1	< 0.1%		x	x
Perfluorotetradecanoic acid (PFTA)	160	0.3	< 0.1%		x	x
Perfluorohexadecanic acid (PFHxDA)	< 0.3	0.3	< 0.1%		x	x
Perfluorooctane sulfonamide (PFOSA)	72,000	0.3	0.1%		x	x
4:2 Fluorotelomer sulfonate (4:2 FTS)	480	0.3	< 0.1%		x	x
6:2 Fluorotelomer sulfonate (6:2 FTS)	61,000	0.3	0.1%	x	x	x
8:2 Fluorotelomer sulfonate (8:2 FTS)	66,000	0.3	0.1%	x	x	x
7H-Dodecafluoroheptanoic acid (HPFHpA)	< 10	0.3	0%		x	x
Perfluoro-3,7-dimethyl acid (PF-3,7-DMOA)	< 10	0.3	0%		x	x
Σ23 PFAS	110,000,000	0.3	100%			
N-ethylperfluoroctansulfonamid (EtFOSA)					x	
N-ethylperfluoroctansulfonamid-HAc (EtFOSAA)					x	
N-ethylperfluorooctane sulfonamide etanol (EtFOSE)					x	
N-metylperfluoroctan sulfonamide-HAc (MeFOSAA)					x	
N-metylperfluoroctansulfonamidetanol (MeFOSE)					x	
N-metylperfluoroktansulfonamid (MeFOSA)					x	
Perfluoroktansulfonamid-HAc (FOSAA)					x	

Appendix B

Table B1
Kd values for PFOS reported in literature.

Sample type	log Kd	log Koc	References
Natural peat	2.4 ± 0.3	2.9 ± 0.3	MSc Løland (2014)
Lavangvatnet lake sediments	2.2	3.8	MSc Løland (2014)
Sand	1.3 ± 0.1	3.4 ± 0.4	MSc Nordskog (2012)
Organic rich forest soil	2.4 ± 0.2	3.2 ± 0.3	MSc Nordskog (2012)
Natural peat	2.3	2.7	MSc Nordskog (2012)
Average PFOS (log 1 kg-1)	–	3	Zareitalabad et al. (2013)
Sediment 1	1.2	4.7	Chen et al. (2009)
Sediment 2	1.2	3	Ahrens et al. (2011)
Sediment 3	1.9	3.8	Ahrens et al. (2011)
Taihu Lake	–	2.9 ± 0.6	Yang et al. (2011)
Aquifer sediment Washington County ($t = 0$)	0.1	2.5	Ferrey et al. (2009)
Aquifer sediment Washington County ($t = 574d$)	− 0.7	2.8	Ferrey et al. (2009)
Oil-derived black carbon (diesel soot) at pH = 5.05	–	3.0–3.1	Chen et al. (2009)
Crude oil spiked to soil	–	4.2–4.4	Ferrey et al. (2009)
Paddy soil (0.91% Corg)	–	3.3	Chen et al. (2009)
Clay	18.3	2.8	3 M corp. Cited in Johnson et al. (2007)
Clay loam	9.72	2.6	4 M corp. Cited in Johnson et al. (2007)
Sandy loam	35.3	3.1	5 M corp. Cited in Johnson et al. (2007)
River sediment	7.42	2.8	6 M corp. Cited in Johnson et al. (2007)
Water treatment sludge	120	2.5	7 M corp. Cited in Johnson et al. (2007)
Otttawa sand standard	2.81	–	Johnson et al. (2007)
Kaolinite	5.31	2.4	Johnson et al. (2007)
Goethite	7.88	–	Johnson et al. (2007)
High iron sand sediment	8.9	–	Johnson et al. (2007)
Lake Michigan sediment	7.52	2.4–2.6	Johnson et al. (2007)
Five sediments with Corg of 0.56–9.66%	–	2.7	Higgins and Luthy (2006)

Appendix C

Table C1
PFAS concentrations in leachate samples from low infiltration column.

Day	PFBS	PFHxS	PFHpS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	FTS 4:2	FTS 6:2	FTS 8:2
	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l
0	0	10	0	12	0	0	0	0	0	0	0	0
7	0	72[a]	20[a]	850[a]	0	0	0	0	13[a]	0	0	0
14	0	55[a]	39[a]	6500[a]	0	0	0	0	11[a]	0	0	0
21	0	0	0	22	0	0	0	0	0	0	0	0
28	140	0	0	13	0	0	0	0	0	0	0	0
35	2600	33	0	0	110	210	710	12	0	0	0	0
42	14,000	230	0	0	880	1900	5800	160	16	0	0	0
49	35,000	1500	0	0	2200	4300	17,000	960	45	33	0	0
56	53,000	5500	0	0	3300	7500	28,000	2900	190	43	0	0
63	53,000	19,000	0	0	4000	8800	32,000	6300	660	11	0	0
70	40,000	37,000	0	13	3800	11,000	29,000	8700	1300	0	0	0
77	30,000	58,000	0	13	3500	11,000	25,000	9300	2500	0	72	0
84	20,000	88,000	0	10	3000	12,000	21,000	8200	4000	0	400	0
91	13,000	120,000	0	15	2700	13,000	18,000	6800	5600	0	960	0
98	7300	130,000	0	10	2300	13,000	14,000	5100	6400	0	1700	0

[a] Results are considered to be a consequence of sample contamination.

Table C2
PFAS concentrations in leachate samples from high infiltration column.

Day	PFBS	PFHxS	PFHpS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	FTS 4:2	FTS 6:2	FTS 8:2
	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l	ng/l
0	0	13	0	17	0	0	13	11	34	0	0	0
7	29	14	0.47	11	3.5	8.1	17	8.1	26	0	7.4	0
14	2700	1100	0	16	250	430	1500	290	110	11	0	0
15	7100	5400	0	14	740	1200	4400	920	380	31	16	0
17	9000	9900	0	14	830	1500	5200	1300	760	37	120	0
19	10,000	11,000	0	15	830	1400	5700	1500	840	38	160	0
21	11,000	16,000	0	20	840	1400	6400	1500	1200	45	340	0
23	12,000	20,000	0	27	840	1600	6900	1700	1500	40	610	0
24	13,000	29,000	12	56	980	2100	7500	1900	2000	34	1300	0
26	14,000	31,000	26	62	1200	2300	9000	2000	2000	39	1300	0
27	16,000	34,000	40	78	1300	2600	10,000	2300	2600	42	1700	0
28	15,000	32,000	28	74	1300	2800	9200	2200	2400	34	1700	0
29	16,000	39,000	60	110	1400	2900	10,000	2300	2700	33	2000	0
30	15,000	36,000	48	100	1400	3100	9500	2400	2700	27	1900	0
31	18,000	46,000	200	280	1700	3700	12,000	2900	3500	31	3100	0
33	17,000	42,000	140	230	1600	3700	10,000	2800	3300	24	2600	0
35	19,000	46,000	250	300	1800	3800	12,000	2700	3700	28	3200	0
37	17,000	51,000	270	410	1700	4100	11,000	3000	3900	20	3400	0
40	17,000	60,000	570	840	1800	4500	11,000	3200	5100	16	4700	0
42	17,000	58,000	610	900	1900	4700	11,000	3400	5100	20	4700	0
44	18,000	61,000	760	1100	2100	5200	12,000	3300	5600	19	4900	0
47	19,000	71,000	1300	1900	2100	5300	15,000	3400	5800	16	5300	0
49	16,000	64,000	1200	1900	1900	5400	12,000	3400	5900	16	5200	0
51	17,000	63,000	1300	1800	2300	5400	14,000	3600	5800	13	5100	0
56	18,000	63,000	1200	1600	2400	5900	15,000	3900	6100	14	5300	0

Table C3
PFAS concentrations in soil samples from low infiltration column.

Column height	PFBS	PFHxS	PFHpS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	FTS 4:2	FTS 6:2	FTS 8:2
cm	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg
00–05	4	38	3.2	7.4	0.63	1.9	3.8	1.5	3.5	0	3.9	0
05–10	3.6	37	5.8	27	0.63	2.1	3.6	1.4	4.1	0	3.8	0
10–15	3.2	37	7.9	52	0.6	2	3.3	1.5	4.6	0	3.4	0
15–20	3.6	33	7.3	99	0.67	2.7	3.7	1.7	4.4	0	1.5	0
20–25	3.7	55	3.8	60	0.71	2.6	4.3	2.2	6.5	0	1.2	0
25–30	2.3	36	4.9	110	0.61	2.6	3.2	1.5	4.5	0	1.1	0
30–35	1.1	28	5.7	120	0.43	2	2.2	0.87	3.9	0	0.99	0

Table C3 (*continued*)

Column height	PFBS	PFHxS	PFHpS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	FTS 4:2	FTS 6:2	FTS 8:2
cm	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg
35–40	0.76	32	10	260	0.42	2.3	2.3	0.79	4.9	0	2.3	0
40–45	0.5	27	11	350	0.39	2.2	1.8	0.61	4.5	0	3.2	0
45–50	0.26	18	11	390	0.3	1.7	1.3	0.38	3.2	0	3.3	0
50–55	0.21	11	16	590	0.25	1.5	1.1	0.27	2.5	0	3.1	0
55–60	0	7.7	14	790	0.24	1.1	0.78	0.2	1.5	0	2.8	0
60–65	0	5.2	9.4	1000	0	0.84	0.57	0	0.87	0	1.7	0
65–70	0	4.1	3.7	970	0	0.6	0.4	0	0.53	0	1.1	0
70–75	0	4	2	1000	0	0.52	0.36	0	0.49	0	0.9	0
75–80	0	4.7	0.95	680	0	0.53	0.34	0	0.46	0	0.52	0.47
80–85	0	5.2	0.66	460	0	0.39	0.31	0	0.4	0	0.32	0.47
85–90	0	6.4	0.52	180	0	0.33	0.34	0	0.46	0	0.23	1.3
90–94.5	0.53	12	0.93	140	0.22	0.41	0.83	0	0.52	0	0.4	3.4

Table C4
PFAS concentrations in soil samples from high infiltration column.

Column height	PFBS	PFHxS	PFHpS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	FTS 4:2	FTS 6:2	FTS 8:2
cm	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg
00–05	0	68	0	0.21	0.58	3.1	3.3	1.5	5.3	0	1	0
05–10	0	84	0	0.23	0.48	2.7	2.5	1.2	6.8	0	1.2	0
10–15	0	58	0	0.22	0.44	2.7	2.1	0.9	7.3	0	1.1	0
15–20	0	57	0	0.27	0.42	2.7	1.9	0.67	7.1	0	1.4	0
20–25	0	54	0.28	0.22	0.42	2.6	1.8	0.57	8.4	0	1.8	0
25–30	0	46	2.1	2.2	0.41	2.6	1.7	0.5	7.9	0	2.4	0
30–35	0	29	6.2	4	0.3	1.8	1.2	0.34	5.7	0	2.4	0
35–40	0	23	22	48	0.32	1.8	1.1	0.27	4.6	0	3.1	0
40–45	0	17	28	170	0.27	1.5	0.96	0	3.4	0	3.2	0
45–50	0	9.2	19	350	0.23	1.1	0.57	0	1.7	0	2.3	0
50–55	0	7.9	15	940	0.25	0.85	0.46	0	1.2	0	2.8	0
55–60	0	6.2	12	940	0	0.71	0.37	0	0.9	0	2.2	0
60–65	0	5.4	4.5	1700	0	0.48	0.31	0	0.54	0	1.4	0
65–70	0	4.8	2.8	1300	0	0.43	0.24	0	0.45	0	0.95	0
70–75	0	4.7	1.3	1000	0	0.43	0.26	0	0.41	0	0.83	0.25
75–80	0	4.2	0.65	730	0	0.38	0.24	0	0.32	0	0.58	0.42
80–85	0	4.1	0.51	360	0.2	0.34	0.26	0	0.34	0	0.42	0.54
85–90	0	3.1	0.84	170	0	0.27	0.23	0	0.36	0	0.37	1.1
90–93	0	4.9	1.6	260	0.31	0.34	0.58	0	0.36	0	0.55	2.7

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