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Plant uptake of per- and polyfluoroalkyl substances at a contaminated fire training facility to evaluate the phytoremediation potential of various plant species

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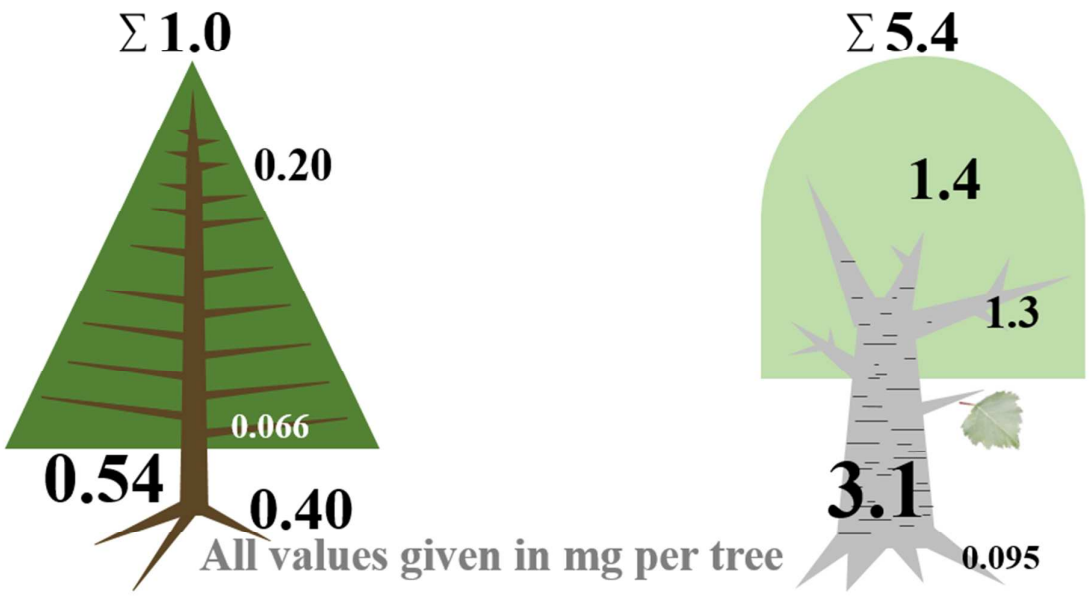


Abstract. Fire training facilities and other areas suffer from serious per- and polyfluoroalkyl substances (PFAS) contamination in soil, surface water and groundwater due to regular practices with PFASs-containing aqueous firefighting foams (AFFFs). Therefore, the uptake of 26 PFASs in plants and the contamination of soil and groundwater has been investigated at a fire training site at Stockholm Arlanda airport, Stockholm (Sweden) in 2016. Elevated $\sum_{26}$PFASs levels were detected in soil and groundwater ranging from 16-160 ng g^{-1} dry weight (dw) and 1,200-34,000 ng L^{-1}, respectively. Samples from different plant species and tissues (i.e. roots, trunk/cores, twigs, leaves/needles) of the local plant community were taken, namely silver birch (*Betula pendula*), Norway spruce (*Picea abies*), bird cherry (*Prunus padus*), mountain ash (*Sorbus aucuparia*), ground elder (*Aegopodium podagraria*), long beechfern (*Phegopteris connectilis*) and wild strawberry (*Fragaria vesca*). The plants showed a high variability of concentrations with highest $\sum_{26}$PFASs concentrations in vegetative compartments with up to 97 ng g^{-1} wet weight (ww) and 94 ng g^{-1} ww in birch leaves and spruce needles, respectively. Annual ground cover plants such as long beechfern and ground elder, and bushes like bird cherry showed concentrations up to 6.9, 23 and 21 ng g^{-1} ww, respectively. The bioconcentration factors (BCFs; plant/soil ratios) were highest in foliage, while the total tree burden of Σ_{26}PFASs per tree was up to 11 mg for birch and 1.8 mg for spruce. Considering a shelterwood system with mixed stands of silver birch and spruce in combination with regular harvest of leaves and birch sap and an understorey of ground elder, it is potentially feasible to remove 1.4 g of Σ_{26}PFASs per year and hectare from (heavily) contaminated sites. An alternative approach is the coppicing of birch trees in combination with an understorey of ground elder, potentially removing 0.65 g yr^{-1} ha^{-1} of Σ_{26}PFASs, while a simple meadow with ground elder can remove 0.55 g yr^{-1} ha^{-1} Σ_{26}PFASs.

Keywords: PFASs, phytoremediation, silver birch, Norway spruce, groundwater, airport

Introduction

Per- and polyfluoroalkyl substances (PFASs) are aliphatic substances (hydrophobic alkyl chain and hydrophilic end group), leading to their highly demanded properties as surfactants and the extensive use in e.g. textiles, cookware, paper packaging and aqueous firefighting foams (AFFFs)[1]. Their usefulness, in combination with their high persistence has led to their ubiquitous distribution even in remote environments[2]. Additionally, some PFASs (e.g. perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA)) have been proven potentially toxic, carcinogenic and bioaccumulative[3,4]. As a consequence, PFOS and perfluorooctanesulfonyl fluoride (PFOSF) have been listed in the Stockholm Convention on persistent organic pollutants (POPs) in 2010 (Stockholm Convention, SC-4/17, 2009). Furthermore, PFOS and PFOA have been restricted under the Toxic Substances Control Act (TSCA) by US EPA in 2012 (US EPA, 2009), while PFOA has been characterized as a substance of very high concern (SVHC) under the European REACH regulation (SVHC, article 57d). After legislative restriction of PFOS and PFOA, alternative PFASs[5] and short-chained PFASs (<7 CF_2) were used as replacements[6]. However, these are considered more mobile, particularly due to a higher water solubility (S_w) and lower sorption onto soil particles[5,7].

Typical point sources for PFASs are fire training facilities using PFAS-containing AFFF which have shown to have a long-term impact on the nearby environment[8] and runoff from fire training sites has previously led to drinking water contamination (up to 10,000 ng L^{-1}) near the air force in Brantafors in Kallinge (Ronneby municipality, Sweden in 2013)[9]. While the soil might even be in direct contact with the contaminated runoff, the groundwater accumulates PFASs over time due to leaching from contaminated soil[10]. To the current state of knowledge, there are no remediation techniques available for PFASs despite PFASs in groundwater posing a serious threat as they are migrating to other watercourses and thereby

contaminating the nearby environment[11]. Phytoremediation provides a sustainable, cost-efficient and passive *in situ* approach to remediate contaminated media such as soil and water, particularly due to the lack of established and commercially applicable remediation techniques for PFASs. Previous studies have shown PFASs uptake by plants, however, these studies were mainly limited to agricultural crops[12–14] and conducted under controlled conditions[15,16].

This study aims to assess the distribution of PFASs in local vegetation, surface soil and groundwater at Stockholm Arlanda airport, Sweden, to evaluate the feasibility of remediation using phytoextraction. The specific objectives were to i) determine the contamination of PFASs in vegetation, surface soil and groundwater at Stockholm Arlanda airport, Sweden, ii) assess the distribution of PFASs in various plant tissues including roots, stem, branches, leaves, needles, birch sap (xylem) and berries, iii) calculate the bioconcentration factor (BCF) and total tree burden for individual PFASs for various plant species, and iv) evaluate the groundwater and soil remediation potential on the basis of three remediation scenarios.

Experimental Section

Chemicals. The PFASs included C_3-C_{13}, C_{15}, C_{17} perfluoroalkyl carboxylates (PFCAs) (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA and PFOcDA), C_4, C_6, C_8, C_{10} perfluoroalkane sulfonates (PFSAs) (PFBS, PFHxS, PFOS, PFDS), methyl- and ethylperfluorooctanesulfonamides FOSAs (FOSA, MeFOSA, EtFOSA), methyl- and ethylperfluorooctanesulfonamidoethanol FOSEs (MeFOSE, EtFOSE), methyl- and ethylperfluorooctanesulfonamidoacetic acid FOSAAs (FOSAA, MeFOSAA, EtFOSAA) and 6:2 fluorotelomer sulfonate (FTSA). Additionally, $^{13}C_4$ PFBA, $^{13}C_2$ 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_3-*N*-MeFOSA, d_5-*N*-EtFOSA, d_7-*N*-MeFOSE, d_9-*N*-

EtFOSE, d_3-*N*-MeFOSAA and d_5-*N*-Et-FOSAA were used as mass-labelled internal standards (IS) (Table S1 and S2 in the Supporting Information (SI)). Full names as well as CAS registry numbers of the substances can be obtained from the Table S1 in the SI.

Historical Use of PFAS containing AFFF at the Fire Training Facility at Stockholm Arlanda airport. Stockholm Arlanda airport is Sweden's biggest airport located between Stockholm and Uppsala, Sweden. Its fire training site was established before 1987 with a runoff collection system installed in 1997. The runoff collection consists of a collection pan, three sedimentation ponds and a pre-treatment with activated carbon before release into the ordinary waste water stream. PFOS-based AFFF has been used from the 1980s until 2003, since then only PFOS-free AFFF was purchased (still containing >10% PFASs) while the remaining stocks of PFOS-based AFFF were used up. From 2011 onwards, PFASs-free AFFF was bought and applied[11].

Study Design and Sampling. Samples were taken twice, on the 22nd of March 2016 and the 30st of June 2016 at three locations south (<500 m) of the fire training site at Stockholm Arlanda airport (Figure 1). The first sampling campaign accounted for the birch sap collection, water-, soil- and some tree samples while the majority of plant samples was collected in June (see SI, Tables S3-S4, for detailed information). Besides representativeness, the locations were chosen according to the i) existence of a groundwater well, ii) proximity (<500 m) to the fire training site and within estimated groundwater flow direction, iii) availability of mature birch and spruce trees, and iv) accessibility for sampling. Sampling site 1 was furthest away from the fire training site with ~430 m, followed by site 3 with ~340 m and site 2 with ~250 m (see Figure S1).

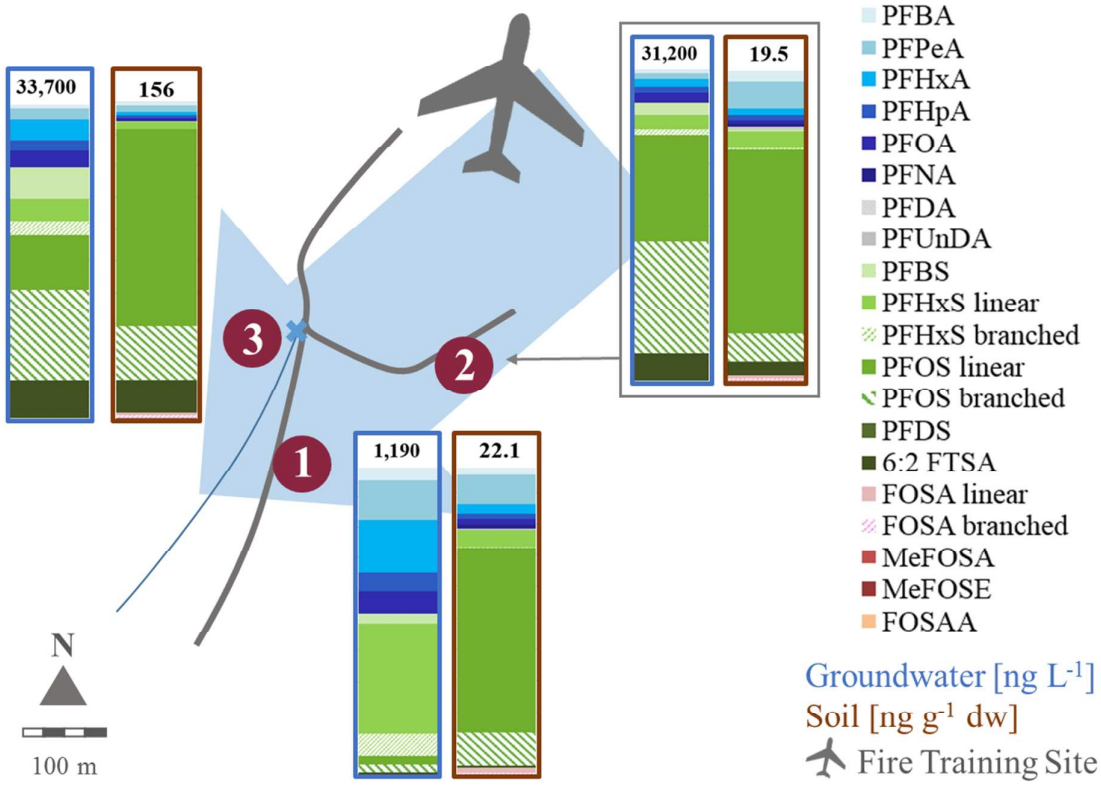


Figure 1 Schematic map of the sampling area around the fire training site (indicated with the airplane) showing the three sampling sites (1-3) with corresponding soil (brown) and groundwater (blue) composition profiles. The $\sum_{26}$PFAS concentration is given in bold at the top of each bar. Grey lines indicate the roads, the thin blue line represents the ditch where the surface water sample was collected (blue cross). The big pale blue arrow shows the groundwater flow direction.

One liter grab samples were collected in polypropylene (PP) bottles from the surface water in a ditch (n = 1) near the fire training facility and groundwater samples (n = 3) were collected at each site using a field peristaltic pump (12V DC, Eijkelkamp, The Netherlands) solely in March (Table S3 in the SI). The groundwater depth was ranging from 1.10-1.80 m. Composite soil samples were sampled with a steel shovel in 500 mL zip-lock bags at approximately 0-10 cm depth at all three sites close to the existing groundwater wells in

March (n = 3) and June (n = 3; Table S3 in the SI). In March, only birch and spruce were sampled, with leaves being collected from the ground, roots being dug up from approximately 10 cm depth and needles and spruce twigs being sampled directly from the tree (Table S4 in the SI). Birch twigs were solely collected in June. The trunks of the sampled trees had a diameter at breast height (DBH) of 19-35 cm and were considered mature trees. The trunks were sampled at chest height using an increment borer (diameter 1 cm, Suunto, Finland) that was drilled into the trunk according to a method described previously[17]. Small, food-grade plastic taps (Atkinson maple syrup supplies, Canada) were inserted into the resulting holes and a 1 L PP-bottle (Nalgene, USA) was placed directly underneath for collection of the birch sap from the xylem. As the effect of UV on PFASs degradation remains unknown, the bottles were covered with dark tape for UV protection. The bottles were left for ten days in March (March 22nd until 1st of April, n = 4) and then replaced with empty ones to collect another sap sample for approximately three months until the end of June (n = 4). In June, samples from silver birch (*Betula pendula*), bird cherry (*Prunus padus*), mountain ash (*Sorbus aucuparia*), ground elder (*Aegopodium podagraria*), long beechfern (*Phegopteris connectilis*) and wild strawberry (*Fragaria vesca*) were taken with focus on leaves and stems/twigs. Roots and fruits were sampled if feasible or applicable. All plants appeared to be in good condition and showed no abnormalities upon visual examination. In total, 44 plant samples were collected in 1-3 L zip-lock bags (for detailed information of the sampling and plant characteristics see Tables S3-S4 in the SI).

Sample Preparation and Analysis. Liquid samples, comprising surface- and groundwater as well as birch sap, were prepared as described by Ahrens et al. (2015)[11]. After filtration (1.2 μm glass fiber filter (GFF), GE Healthcare Life Sciences, Whatman, UK), 300 mL liquid sample were spiked with 100 μL of an internal standard (IS, concentration (c) = 4 ng mL^{-1}) mix and then processed with solid-phase extraction (SPE) using Oasis® WAX

cartridges (6cc, 500 mg, 60 μm, Waters Corporation, USA). The extracts were eluted in one vial using methanol and 0.1% ammonium hydroxide in methanol. Elutions were concentrated at room temperature under a nitrogen stream to 1 mL for analysis using high-performance liquid chromatography coupled with tandem mass spectrometry (HPLC-MS/MS) (for details on the instrumental analysis see elsewhere)[11].

The soil and plant material was extracted based on a method described previously[18]. While soil samples were freeze-dried for 7 days directly after sampling, plant samples were washed with MilliQ water first, then freeze-dried for 7 days and homogenized using a homogenizer (*Precellys Evolution*, Bertin, France), a mortar and pestle made of ceramics or a coffee blender with a grinder made of stainless steel (*GVX 2*, Krups, Germany). Soil and plant samples were then weighed to ~2 g each, spiked with 100 μL of an internal standard (IS, c = 4 ng mL^{-1}) mix and extracted with solid-liquid extraction using methanol. The extract was concentrated at room temperature under a nitrogen stream to 1 mL and then cleaned up with 25 mg ENVI-carb before analysis using HPLC-MS/MS. The blanks were extracted in the same way as the natural samples without the sample material. The branched isomers of perfluorohexanesulfonate (PFHxS), PFOS and perfluorooctanesulfonamide (FOSA) were semi-quantified using the corresponding linear standard (for detailed information of the extraction and analysis see Tables S5-S7 in the SI).

The dry matter content was determined for plant samples by weighing the fresh sample and weighing the freeze-dried sample after 7 days of drying. Aliquots of soil samples were weighed fresh and after drying in an oven at 105°C for 12 h before determination of the dry weight (dw)[19]. Additionally, aliquot soil samples were burned at 550°C in a muffle oven for 12 h to determine the organic matter content.

Quality Assurance and Quality Control. Blanks and duplicates were analysed with each batch of samples to track contamination and analyte recovery. There were separate

blanks for the water (n = 2) and sap samples (n = 2), the soil (n = 5) and plant samples (n = 5), the homogenizer (n = 1) and the coffee grinder (n = 2). The blanks were used to determine background noise and for calculation of the method detection limit (MDL). The method detection limit was calculated for individual PFASs as the sum of the blank's mean (mean is always referring to the arithmetic mean) concentration and the product of the blanks' standard deviation times three. The blank concentrations of all matrices were below minimum detected sample concentrations and ranging from non-detected to 0.19 ng L^{-1} per individual substance (Table S5 in the SI). All samples were prepared and analysed in duplicates for better accuracy, leading to 5 duplicates for the water samples, 8 duplicates for the sap samples, 6 duplicates for soil and 45 duplicates for the plant samples. The relative standard deviation of the duplicates was in the range of 2-43% for water, 10-48% for birch sap, 2-171% for soil and 8-95% for the plants (Table S6 in the SI). For details of the blank levels, MDLs, reproducibility and recovery see Tables S5-S7 in the SI.

Results and Discussion

PFASs Contamination of groundwater and soil at Stockholm Arlanda airport.

Thirteen out of the 26 analysed PFASs could be detected in the ground- and surface water samples at Stockholm Arlanda airport (Tables S8-S10 in the SI). The $\sum_{26}$PFASs concentrations (including the linear and branched isomers of PFHxS, PFOS and FOSA) in the three groundwater samples were ranging from 1,200 to 34,000 ng L^{-1} and were increasing with proximity (lack of dilution) to the training site (Figure 1). The $\sum_{26}$PFASs surface water concentration in the ditch was 650 ng L^{-1} (2016), which is 6-times lower than measured in 2011 (~4,000 ng L^{-1})[11], indicating a lower recent contamination due to the shift to PFASs-free AFFF and the introduction of the runoff collection system during the 1990's. Furthermore, the composition profiles based on the $\sum_{26}$PFASs concentration from the two studies (2011[11] and

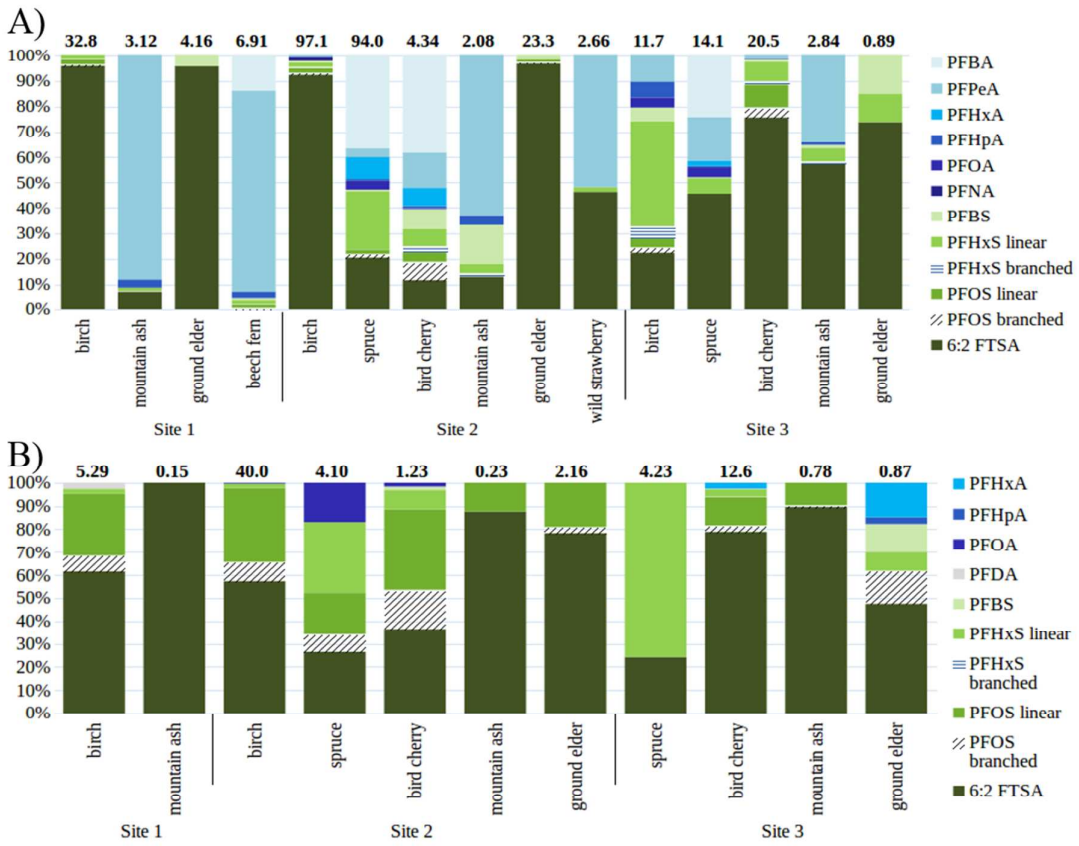
2016 (this study), respectively) were quite similar with PFOS (59% and 48%) prevailing, followed by PFHxS;(24% and 11%), perfluorohexanoate (PFHxA; 7% and 4%) and PFOA (5% and 2%, respectively). Additionally, the PFOS/$PFOA_{surface\ water}$ ratio was greater than 1 in both studies (6.5 ± 4, mean ± standard deviation, and 31 from 2011 and 2016, respectively), indicating a local source of PFOS originating from the use of AFFFs[8] (for the ratio, the sum of linear and branched PFOS isomers was used). The PFOS/$PFOA_{groundwater}$ ratios of 24 (site 2) and 8.7 (site 3) from this study support this finding. However, the groundwater at the sampling site furthest away from the training site (site 1) had a ratio <1 (0.73), possibly implying no recent influence from the training site. All water samples were highly contaminated in comparison to background levels in European river surface water ranging from <MDL to 200 ng L^{-1} for individual PFASs[20] and 10 to 100 ng L^{-1} for Σ_{26}PFASs[21]. Moreover, the groundwater concentrations (1.2-34 µg L^{-1}) were also exceeding the median concentrations found on AFFF-impacted sites in the USA (0.2-4.2 µg L^{-1})[22], which might be due to the fact that these sites were only exposed to AFFF during a one-time spilling event.

Seventeen out of 26 substances were detected in the soil (Tables S11-S13 in the SI). The soil samples showed a similar trend regarding proximity, with the Σ_{26}PFASs concentrations decreasing with distance from 160 ng g^{-1} dw (site 3) to 16 ng g^{-1} dw (site 1) (Figure 1). Similar to the water samples, PFOS was the dominant substance with up to 97 ng g^{-1} dw at site 3. The median soil concentrations for individual PFAS were in the range of <MDL (e.g. C_{11}-C_{17} PFCAs) to 19 (sum linear and branched PFOS isomers) ng g^{-1}dw, and are comparable to median individual PFAS concentrations ranging between 0.67 (PFTriDA) and 53 (PFOS) ng g^{-1} at other AFFF-impacted sites (i.e. AFFF lagoons and hangar-related storage tanks)[22].

Species Specific Accumulation of PFASs. Contaminant uptake is a passive process dependent on a plant's water- and nutrient demand. Accordingly, Dodgen et al. (2015)

observed a direct correlation between evapotranspiration and uptake of organic contaminants[23]. Most probably, contaminant uptake is only limited by the toxicity threshold at which the contaminant burden in the plant leads to inhibited development or death of the plant. Ten out of 26 PFASs were detected in plants (Tables S14-S40 in the SI). Foliage and twig samples were taken from all investigated species and are therefore suitable for comparing different species. Contaminant uptake is highly species-dependent (Figure 2), an observation previously made by Krippner et al. (2014)[24] and probably also varies with season, with highest Σ_{26}PFASs concentrations in foliage of birch (12-97 ng g^{-1} ww) > spruce (14-94 ng g^{-1} ww) > bird cherry (4.3-21 ng g^{-1} ww) > ground elder (0.89-23 ng g^{-1} ww) > mountain ash (2.1-3.1 ng g^{-1} ww). The concentrations detected in long beechfern (6.91 ng g^{-1} ww Σ_{26}PFASs) require further investigation as there was only one sample collected which might not be representative. The composition profiles of the leaves showed a high proportion of 6:2 fluorotelomersulfonate (6:2 FTSA; on average, 50% for Σ_{26}PFASs), which is used as a replacement for PFSAs[5]. Furthermore, the samples have high fractions of the short-chained PFCAs, in particular perfluoropentanoate (PFPeA; on average, 24% for Σ_{26}PFASs) and perfluorobutanoate (PFBA; 7.5%), which is most likely due to their high water solubility and therefore increased mobility of short-chained PFCAs[25].

The Σ_{26}PFASs concentrations in the twigs were substantially lower than those found in the foliage, however, the order of concentration level is similar, with highest Σ_{26}PFASs concentrations in birch (5.3-40 ng g^{-1} ww), followed by spruce (4.1-4.2 ng g^{-1} ww) and bird cherry (1.2-13 ng g^{-1} ww), ground elder (0.87-2.2 ng g^{-1} ww) and mountain ash (0.15-0.78 ng g^{-1} ww). Dominant PFASs in twigs were 6:2 FTSA (on average, 55% of the Σ_{26}PFASs), followed by PFHxS (12%) and PFOS (14%).



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248 **Figure 2 Foliage and Twigs.** A) Foliage and B) twigs/stems composition profiles of the
249 investigated species in percent. $\sum_{26}$PFASs concentrations [ng g^{-1} ww] are given in bold at the
250 top of each bar. The species are sorted in the following order: trees, bushes, ground cover
251 species. For the birches, only the concentrations from June 2016 are included.

252

253 **Plant Tissue Distribution.** The most comprehensive sampling was conducted with the
254 tree species birch (*Betula pendula*) and spruce (*Picea abies*) where samples were taken from
255 "roots to shoots" (Figure 3). For birch, the Σ_{26}PFASs concentrations were highest in leaves
256 (12-97 ng g^{-1} ww), followed by twigs (5.3 and 40 ng g^{-1} ww), trunk/core (0.37-31 ng g^{-1} ww)
257 and roots (2.6-6.2 ng g^{-1} ww). Similarly, Σ_{26}PFASs concentrations in spruce followed the
258 following trend: leaves > twigs > roots (41 ng g^{-1} ww) > trunk/core (1.3-19 ng g^{-1} ww). To the
259 best of our knowledge, there are no other studies available that focused on uptake of PFASs in

trees, however, PFASs uptake in aquatic plants[26], selected edible vegetables[12,13,27] and grass species[13,15] has previously been investigated. PFASs can be translocated within the plant from the roots to the leaves where the water is transpired and consequently leads to an accumulation of the contaminants in leaves[13,14]. A translocation of PFOS and PFOA to the leaves has been shown previously in spring wheat, oats, potatoes, maize and perennial ryegrass in an indoor experiment with spiked soil[13]. Lechner and Knapp (2011)[16] noted that >80% of PFASs were accumulated in the vegetative compartments of carrots, cucumbers and potatoes when grown in soil spiked with contaminated sewage sludge. This trend was not observed in the two tree species where a median fraction of 35% of PFASs was allocated in the foliage and twigs. However, there is a tendency of decreasing concentrations from leaves to roots in the birch trees with Σ_{26}PFASs concentrations starting as low as 2.6 ng g^{-1} ww in the roots and going up as high as 97 ng g^{-1} ww in the leaves. In contrast, the sampled spruce contained high Σ_{26}PFASs concentrations in the root (41 ng g^{-1} ww), not confirming this pattern.

Tree core samples were collected and the heartwood ("core without bark") was analysed separately from the sapwood with bark ("core with bark") of the tree core (Figure 3). Time trend analysis was not possible due to the low quantity of the tree core samples but would be interesting to explore in the future. However, both species showed a distinctive difference in concentration with threefold higher Σ_{26}PFASs concentrations in the heartwood in comparison to the sapwood, leading to 0.37 vs. 0.93 ng g^{-1} ww (site 1), 6.6 vs. 20 ng g^{-1} ww (site 3) and 11 vs. 31 ng g^{-1} ww (site 2) Σ_{26}PFASs concentrations for the birch cores and 1.3 vs. 4.3 ng g^{-1} ww (site 3) for the spruce core. Together with the PFOS/$PFOA_{groundwater}$ ratios (see above), this trend might be reflecting the decreased recontamination since the 1990's with the introduction of runoff collection system and switch to PFOS-free AFFF in 2011 and the dilution effects that were observed when comparing the surface water samples from 2016 (this

study) and 2011[11]. Regardless of the tissue, both species predominantly accumulated 6:2 FTSA (on average, 41% and 33% of the Σ_{26}PFASs for all tissue samples for birch and spruce, respectively) and PFOS (40% and 23%), while the rather water soluble PFCAs were mainly found in the needles (48% Σ_{26}PFASs) and leaves (20%) at site 3.

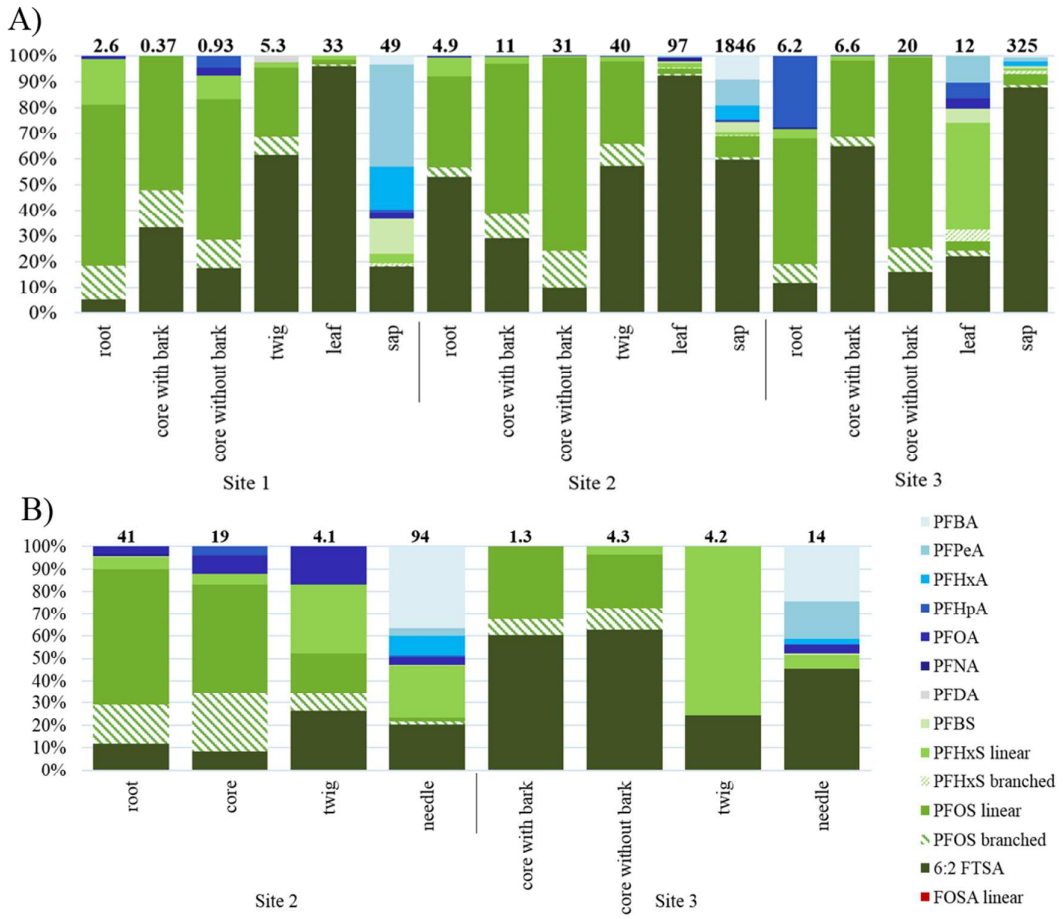

Figure 3 Birch and Spruce. A) Birch and B) spruce PFASs composition profiles in percent. The Σ_{26}PFASs plant concentrations [ng g^{-1} ww] or Σ_{26}PFASs sap concentrations [ng L^{-1}] are indicated in bold at the top of each bar. All tissue and sap samples were sampled in March 2016 except for birch leaves and twigs which were sampled in June 2016.

Bioconcentration Factors and Total Tree Burden. The BCFs represent the plant's ability to accumulate contaminants and remediate the soil or groundwater. Here, BCFs related

297 to the soil concentrations (BCF_s) and groundwater concentrations (BCF_{gw}) were calculated for
298 individual PFASs as follows:

299 $$BCF_s = c_{plant}/c_{soil} \text{ or } BCF_{gw} = c_{tree}/c_{groundwater} \quad (1)$$

300 where c_{plant} and c_{tree} are the individual PFAS concentrations in the plant [ng kg^{-1} ww], c_{soil} is
301 the individual PFASs concentration in the soil [ng kg^{-1} ww] and $c_{groundwater}$ is the individual
302 PFASs concentration in groundwater [ng L^{-1}].

303 Foliage had the highest BCF_s of all tissues ranging from 0-14,000 and accumulated the
304 most PFASs (8 out of 26; Tables S41-S47 in the SI), with birch sap showing BCF_s values up
305 to 41 for 6:2 FTSA. The highest mean BCF_ss were found for 6:2 FTSA (472; n = 52), PFOS
306 (28; n = 36), PFHxS (10; n = 42) and PFOA (5; n = 24) which might correspond to the AFFF
307 composition used at Stockholm Arlanda airport, whereas other studies usually used spiked soil
308 with equal amounts of PFASs[13,28]. From the 26 analysed PFASs studied, PFOS and PFOA
309 have been mostly studied and allow for comparisons with other studies[13,15]. For PFOA, the
310 mean BCF_s (± standard deviation) yielded 18 ±15 for spruce, followed by birch (1.2 ± 1.5)
311 and bird cherry (0.25 ± 0.043), taking into account all investigated compartments of the
312 species, namely foliage, twigs, trunk and the roots (if available; based on ng kg^{-1} ww) (Tables
313 S43-S49 in the SI). This is in the same range as the grass-soil-accumulation factors (GSAF;
314 based on ng g^{-1} dw concentrations) for grass from agricultural fields fertilized with
315 contaminated sewage sludge and mean BCF_s for PFOA of 0.25 ± 0.23 for grass, 0.25 ± 0.08
316 for corn straw, 2.0 ± 1.9 for oat straw and 4.0 ± 1.9 for wheat straw[13,15]. There was no BCF or
317 GSAF available for the species investigated within this study, however, the calculated BCF_s
318 for PFOA for bird cherry and birch fit well within the previous observations for grass, corn
319 straw, oat straw and wheat straw[13,15], whereas the BCF_s for spruce are exceeding these
320 observations. For PFOS, the mean BCF_ss in this study were 1.5 ± 2.4 for spruce, followed by
321 0.99 ± 2.4 for birch and 0.12 ± 0.048 for bird cherry. These results are comparable to Yoo et

al. (2011) with BCFs for PFOS of 0.77 ± 0.55 for wheat straw, 0.16 ± 0.04 for corn straw and 0.07 ± 0.04 for grass[15]. Other PFASs and their uptake behavior have been studied previously, confirming the patterns observed in this study with high PFAS concentrations in the foliage, particularly for short-chained PFASs, while longer-chained PFASs preferably accumulate in the roots[12,24,29,30]. It is important to note that plants tended to accumulate higher amounts of PFASs with increasing PFAS spiking concentrations under greenhouse conditions[12,13,30], while this is a field-based study with PFAS contaminated soil under altered conditions.

While bushes and ground cover species most likely solely access the contaminants in the soil (BCF_s), the trees can additionally take up contaminants from shallow groundwater[31]. Based on the assumption that mature trees access the shallow groundwater (<2 m depth) the following BCFs for the trees refer to the groundwater concentrations (BCF_{gw}). The highest BCF_{gw} (1.4) was calculated for PFOS in birch tissues, followed by a BCF_{gw} of 1.1 for PFBS in spruce. However, the BCF_{gw} were notably lower than the BCF_s (see above) with a median BCF_{gw} ranging from 0.020 (PFBS) to 0.22 (PFOS linear) for birch and from 0.023 (PFHpA) to 0.6 (PFBA) for spruce.

Despite being subject to fluctuations, the PFSAs showed a slight correlation trend of decreasing BCF_s in birches with increasing perfluorocarbon chain length (Figure S2 in the SI), however, no trend was observed for PFCAs. This is in contrast to other studies showing a decreasing PFASs accumulation with increasing perfluorocarbon chain length[28,32–34]. The lack of a trend for PFCAs in this study is most probably due to high variability of the data, attributed to the influence of field conditions with contaminated soil. In contrast, most other studies were performed using spiked non-aged soil which results in a different availability of PFASs[32–34]. Ultimately, individual PFASs were often detected in plants but not in the respective soil which indicate that the uptake of PFASs might not be solely via the soil and more studies about the uptake mechanisms of PFASs are needed.

347 The total tree burden is estimated using biomass proportions determined by Johansson
348 (2007 and 2014)[35,36] and Minerbi and Cescatti (2015)[37], assuming an average height and wood
349 density of 19 m and 530 kg m^{-3} for birches and 15 m and 470 kg m^{-3} for spruces, respectively
350 (Table S48 in the SI). Considering the DBHs of the trees, this leads to a total biomass of 410
351 kg for a birch tree and 93 kg for a spruce tree. The total tree burden was calculated with the
352 following formula:

353 $$\Sigma PFASs_{tree\ burden} = \Sigma PFASs_{root} + \Sigma PFASs_{trunk} + \Sigma PFASs_{twig} + \Sigma PFASs_{foliage} \quad (2)$$

354 $$\Sigma PFASs_{tissue} = c_{mean\ PFASs\ in\ tissue} * \text{tissue fraction of total tree biomass} * \text{tree biomass} \quad (3)$$

355 where $PFASs_{tree\ burden}$ is the total tree burden for all PFASs [mg absolute], $c_{mean\ PFASs\ in\ tissue}$ is
356 the mean concentration of Σ_{26}PFASs in that tissue (e.g. foliage, twigs) [mg kg^{-1} ww],
357 multiplied with the tissue fraction of total tree biomass [%] and the total tree biomass [kg].
358 The mean total tree burden for $\sum_{26}$PFASs was approximately 13 times higher for birches (1.5-
359 11 mg) in comparison to spruces (0.26 and 1.8 mg), which can be attributed to the higher
360 density of birch wood. Additionally, the birch is a deciduous tree, needing to replace the
361 foliage every autumn which leads to increased nutrient (and contaminant) turnover in
362 comparison to spruce trees which keep their needles for 3-7 years before replacing them.
363 The tissue distribution was generally similar in both species with the highest PFASs burden in
364 the trunk (14-88% and 53% for birches and spruces, respectively), followed by the foliage (8-
365 63% and 20%), twigs (20% and 4-26%) and roots (1-4% and 23%). The high PFASs burden
366 in the trunk is mainly due to the weight and volume of the trunk, which greatly exceeds that of
367 twigs or leaves, but not so much due to the mean PFASs concentration (in the trunk). In
368 contrast to birch, which was accumulating PFASs mainly in the above-ground biomass (96-
369 99%), the amount of PFASs in spruce roots was approximately 7 times higher (23% of the
370 total $\sum_{26}$PFASs) compared to birch roots (1-4%). However, since roots are not easily
371 accessible for removal, species with high root accumulation are less suitable for

372 phytoremediation. There was a general trend of increasing total tree burden for PFASs with
373 increasing groundwater and soil concentrations with highest tree burden at site 2, followed by
374 site 3 and 1. This implies that the PFAS uptake is depending on availability until a toxicity
375 threshold is reached which was not observed with concentrations present in this study. This is
376 in accordance with previous studies that reported increasing accumulation with increasing
377 amounts of PFASs for potatoes, carrots and cucumbers, earthworms, wheat, oat, spring wheat
378 and maize[13,16,28]. However, a laboratory study described visible abnormalities in oat, spring
379 wheat, maize and potatoes at soil PFASs concentrations of ~10 mg kg^{-1} [13], while no effect was
380 observed in this study in the field with soil concentrations of 0.16 mg kg^{-1} dw for $\sum_{26}$PFASs.
381 **Phytoremediation Potential.** All investigated plant species showed uptake of PFASs
382 with a maximum in the foliage and twigs. As contaminant concentrations in the plants were
383 increasing with increasing soil and groundwater concentrations without the plants showing
384 visible abnormalities, the toxicity-threshold was most probably not yet reached at the sites.
385 Accordingly, the plants are most likely able to accumulate even higher amounts of PFASs if
386 available which has been shown in previous studies[12,13,30]. While bushes and ground cover
387 species most probably only access contaminants in the soil, trees allow for deeper penetration
388 of the soil, consequently enabling groundwater exploitation. As a consequence, the
389 accessibility of the contaminants could be increased if ground cover species and bushes are
390 irrigated with contaminated groundwater[38–40], thereby using the fast biomass production of
391 ground cover species without resigning from higher extraction rates.
392 In the following, three remediation scenarios were developed to evaluate their
393 performance (Table 1) according to the PFAS extraction efficiency, while an economic
394 analysis was not an aim of this study. The first scenario comprises a shelterwood of mixed
395 silver birch (~66%) and Norway spruce (~33%) stands[36] (Table S51 and S54 in the SI). It very
396 much resembles the current vegetation structure at the sampling sites at Stockholm Arlanda

397 airport and is common in Nordic countries[36]. Sheltered spruces grow faster than unsheltered
398 ones which makes these stands highly profitable[41] with a total production of 350 m^3 ha^{-1}
399 within 45 years for both species combined[36]. The extractable amounts of PFASs were
400 calculated as follows:

401 $\Sigma PFASs_{tree\ biomass}$ = total biomass production * share of tree species * {$c_{mean\ PFASs\ in\ tissue}$ *
402 Tissue fraction of total tree biomass} (4)

403 where extractable PFASs are $\Sigma_{26}PFASs$ [g ha^{-1} yr^{-1}], total biomass production [m^3 ha^{-1} yr^{-1}],
404 share of tree species [birch: 66%, spruce: 33%], $c_{mean\ PFASs\ in\ tissue}$ [g kg^{-1} ww] and the tissue
405 fraction of the total tree biomass [%]. The product in {brackets} has to be calculated for
406 foliage, twigs and trunk separately.
407 The amount of extractable PFASs can be extended, if combined with birch sap extraction:

408 $\Sigma PFASs_{sap}$ = Number of birch trees * area of trunk cross section * sap flow * $c_{mean\ PFASs\ in}$
409 $_{sap}$ * 61 days (5)

410 where $\Sigma PFASs_{sap}$ is the extractable amount of PFASs from birch sap (ng ha^{-1} yr^{-1}), number of
411 birch trees is the average number of birch trees (106) per hectare according to Johansson
412 (2014)[36], area of trunk cross section is the cross section area per tree at chest height (401 cm^2),
413 sap flow is given per tree and day (0.25 L cm^{-2} d^{-1})[31] leading to a daily sap flow of ~11,000 L
414 ha^{-1}, $c_{mean\ PFASs\ in\ sap}$ is the mean concentration of PFASs in birch sap (ng L^{-1}) and the factor of
415 61 days is representing 2 months during spring time when the sap is usually flowing. Specific
416 values can be obtained from the Table S50 in the SI.
417 The amount of extractable PFASs can be extended further, if ground cover species are grown
418 underneath and regularly mowed:

419 $\Sigma PFASs_{meadow}$ = (1 ha – area covered with trees) * number of plants * weight of plants *
420 share of tissue * $c_{mean\ PFASs\ in\ tissue}$ (6)

421 where the number of plants is assuming one plant every 10 cm, weight of plants [g], share of
422 tissue [stem: 50%, foliage: 50%] and $c_{\text{mean PFASs in tissue}}$ [g kg^{-1} ww] is the concentration of
423 PFASs in ground elder stems or foliage.
424 If this management system is combined with yearly collection of birch sap in the springtime,
425 removal of birch leaves in autumn and a meadow consisting of ground elder which is mowed
426 regularly, then approximately 1.4 g of ΣPFASs can be extracted per year and hectare. Algreen
427 et al. (2014) has proposed an alternative approach to calculate PFASs extraction from soil by
428 trees, taking into consideration the decreasing soil concentrations over time[42]. Based on their
429 equations and the recommended threshold values for PFOS in soils by the Swedish
430 Geotechnical Institute (i.e. 0.003 mg kg^{-1} dw for sensitive land use and 0.02 mg kg^{-1} dw for
431 non-sensitive land use)[43], the necessary time for remediating the soil at Stockholm Arlanda
432 airport has been calculated (Table S54 in the SI). Accordingly, the extraction of PFOS by
433 birches and spruces would need 160,000 years and 48,000 years, respectively, to achieve the
434 target value for sensitive land use or 58,000 years and 18,000 years, respectively, for the non-
435 sensitive land use. This is in a similar range as needed for soil remediation of nickel (180,000
436 or 20,000 years)[43]. Assuming the same toxicity of the other 25 analysed PFASs as for PFOS,
437 this would result in a recommended threshold value for $\sum_{26}$PFASs of 0.078 mg kg^{-1} dw for
438 sensitive land use and 0.52 mg kg^{-1} dw for non-sensitive land use. Accordingly, the extraction
439 of $\sum_{26}$PFASs by birches and spruces would need <45 years due to the great extraction
440 efficiency, in particular for PFBA, PFNA, PFHxS and 6:2 FTSA. Algreen et al. (2014)
441 consider the decreasing soil concentrations over time, however this approach has also some
442 limitations. Firstly, the uptake of PFASs by birches is underestimated since the equations are
443 based on the tree BCF for above-ground biomass that includes the tree weight, however, the
444 tree weight for birches is more than 4 times higher in comparison to spruces of a similar size,

445 which results in a better remediation efficiency of spruces. Secondly, the ageing of the soil is
446 not taken into consideration. Lastly, the remediation of groundwater is not considered.
447 The second scenario is the coppicing method, which can be applied with birches[44] (Table S50
448 in the SI). This method leaves the stumps of the trees but harvests the shooting twigs and
449 leaves in rotating cycles of 3-5 years. This way 5 tons of biomass (comprising twigs and
450 leaves) can be produced per hectare and year [45], leading to 0.65 g of extractable Σ_{26}PFASs
451 each year for birches.
452 The third scenario is the maintenance of a meadow consisting of species with high
453 accumulation potential (Table S51 in the SI). According to our results, the long beechfern or
454 ground elder are reasonable choices, extracting (if mowed regularly) approximately 0.55 g of
455 Σ_{26}PFASs each year per hectare, respectively. While uptake thresholds for the investigated
456 plant species and the potential of the long beechfern remain unclear, the first scenario with a
457 shelterwood showed to be the remediation scenario with the highest PFASs removal per year.
458 The shelterwood method has the advantage that it yields the highest extraction rate of all
459 calculated scenarios while using the natural site conditions and extracting PFASs via three
460 different plant sources (trees, sap, meadow) and remediating both, soil and groundwater, with
461 this method. We can assume that there exist even more efficient plant species than those
462 investigated in this study and therefore recommend looking into the PFASs uptake of common
463 reed grass (*Phragmites australis*), willows (*Salix*) and poplars (*Populus*). Although
464 phytoremediation is a slow, long term approach, it is very cost-efficient in large scale
465 application due to the low maintenance cost and low capital cost[46]. The high biomass
466 production makes it economically attractive if sold as biofuel and might even cover some of
467 the remediation costs. While the wood cannot be used for construction etc. due to the
468 contamination, it is a good quality fuelwood with PFASs being completely destroyed in
469 combustion furnaces at 600-950°C[47,48] without posing a risk for humans or the environment.

Ultimately, phytoremediation is not just a passive, low cost and low maintenance remediation technique, it also remains, to the best of the author's knowledge, the cheapest and most sustainable option available for PFASs contaminated sites.

Table 1. Management scenarios for three different phytoremediation approaches based on the study's findings and forest mensuration data in mg ha^{-1} yr^{-1} for $\sum_{26}$PFASs (Table S50 in the SI).

Management scenario	1) Mixed shelterwood	2) Coppicing	3) Meadow
Tree	509	153	-[a]
Sap	368	-[a]	-[a]
Meadow	540	540	548
$\sum_{26}$PFASs extracted per scenario	1417	693	548

[a] Not included in the management system.

Associated Content

Supporting information. Map and aerial photo of the fire training site at Stockholm Arlanda airport, BCF graph for birch/soil ratios, list of target analytes, CAS registry number, acronyms, supplier and purity, table with most important characteristics of analyzed PFASs, table summarizing data about the sampling locations, table with sampled species, blank concentrations, relative standard deviations (RSDs) for analyzed duplicates, recoveries for individual PFASs in different matrices, results for water, soil and plant samples (mean for duplicates), Bioconcentration factors sorted by plant species including short statistical overview, table with tree biomass estimations, calculations for management scenarios 1-3, Calculation of Scenario 1 with Algreen et al. (2014) equations.

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References

(1) Buck, R. C.; Franklin, J.; Berger, U.; Conder, J. M.; Cousins, I. T.; de Voogt, P.; Jensen, A. A.; Kannan, K.; Mabury, S. A.; van Leeuwen, S. P. J. Perfluoroalkyl and Polyfluoroalkyl Substances in the Environment: Terminology, Classification, and Origins. *Integr. Environ. Assess. Manag.* **2011**, *7* (4), 513–541.

(2) Martin, J. W.; Smithwick, M. M.; Braune, B. M.; Hoekstra, P. F.; Muir, D. C. G.; Mabury, S. A. Identification of Long-Chain Perfluorinated Acids in Biota from the Canadian Arctic. *Environ. Sci. Technol.* **2004**, *38* (2), 373–380.

(3) Itoh, S.; Araki, A.; Mitsui, T.; Miyashita, C.; Goudarzi, H.; Sasaki, S.; Cho, K.; Nakazawa, H.; Iwasaki, Y.; Shinohara, N.; Nonomura, K.; Kishi, R. Association of Perfluoroalkyl Substances Exposure in Utero with Reproductive Hormone Levels in Cord Blood in the Hokkaido Study on Environment and Children's Health. *Environ. Int.* **2016**, *94*, 51–59.

(4) Webster, G. M.; Rauch, S. A.; Ste Marie, N.; Mattman, A.; Lanphear, B. P.; Venners, S. A. Cross-Sectional Associations of Serum Perfluoroalkyl Acids and Thyroid Hormones in US Adults: Variation According to TPOAb and Iodine Status (NHANES 2007-2008). *Environ. Health Perspect.* **2016**, *124* (7), 935–942.

(5) Wang, Z.; Cousins, I. T.; Scheringer, M.; Hungerbuehler, K. Fluorinated Alternatives to Long-Chain Perfluoroalkyl Carboxylic Acids (PFCAs), Perfluoroalkane Sulfonic Acids (PFSAs) and Their Potential Precursors. *Environ. Int.* **2013**, *60*, 242–248.

(6) Moeller, A.; Ahrens, L.; Surm, R.; Westerveld, J.; van der Wielen, F.; Ebinghaus, R.; de Voogt, P. Distribution and Sources of Polyfluoroalkyl Substances (PFAS) in the River Rhine Watershed. *Environ. Pollut.* **2010**, *158* (10), 3243–3250.

(7) Collins, C. D.; Finnegan, E. Modeling the Plant Uptake of Organic Chemicals, Including the Soil-Air-Plant Pathway. *Environ. Sci. Technol.* **2010**, *44* (3), 998–1003.

(8) Awad, E.; Zhang, X.; Bhavsar, S. P.; Petro, S.; Crozier, P. W.; Reiner, E. J.; Fletcher, R.; Tittlemier, S. A.; Braekevelt, E. Long-Term Environmental Fate of Perfluorinated Compounds after Accidental Release at Toronto Airport. *Environ. Sci. Technol.* **2011**, *45* (19), 8081–8089.

(9) Jakobsson, K.; Kronholm Diab, K.; Lindh, C.; Persson, B.; Jönsson, B. Exponering för perfluorerade ämnen (PFAS) i dricksvatten i Ronneby kommun. Region Skåne 2014.

(10) Gellrich, V.; Stahl, T.; Knepper, T. P. Behavior of Perfluorinated Compounds in Soils during Leaching Experiments. *Chemosphere* **2012**, *87* (9), 1052–1056.

(11) Ahrens, L.; Norström, K.; Viktor, T.; Cousins, A. P.; Josefsson, S. Stockholm Arlanda Airport as a Source of per- and Polyfluoroalkyl Substances to Water, Sediment and Fish. *Chemosphere* **2015**, *129*, 33–38.

(12) Felizeter, S.; McLachlan, M. S.; de Voogt, P. Uptake of Perfluorinated Alkyl Acids by Hydroponically Grown Lettuce (*Lactuca sativa*). *Environ. Sci. Technol.* **2012**, *46* (21), 11735–11743.

(13) Stahl, T.; Heyn, J.; Thiele, H.; Huether, J.; Failing, K.; Georgii, S.; Brunn, H. Carryover of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate (PFOS) from Soil to

Plants. *Arch. Environ. Contam. Toxicol.* **2009**, *57* (2), 289–298.
(14) Stahl, T.; Riebe, R. A.; Falk, S.; Failing, K.; Brunn, H. Long-Term Lysimeter Experiment To Investigate the Leaching of Perfluoroalkyl Substances (PFASs) and the Carry-over from Soil to Plants: Results of a Pilot Study. *J. Agric. Food Chem.* **2013**, *61* (8), 1784–1793.
(15) Yoo, H.; Washington, J. W.; Jenkins, T. M.; Ellington, J. J. Quantitative Determination of Perfluorochemicals and Fluorotelomer Alcohols in Plants from Biosolid-Amended Fields Using LC/MS/MS and GC/MS. *Environ. Sci. Technol.* **2011**, *45* (19), 7985–7990.
(16) Lechner, M.; Knapp, H. Carryover of Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate (PFOS) from Soil to Plant and Distribution to the Different Plant Compartments Studied in Cultures of Carrots (*Daucus carota Ssp sativus*), Potatoes (*Solanum tuberosum*), and Cucumbers (*Cucumis sativus*). *J. Agric. Food Chem.* **2011**, *59* (20), 11011–11018.
(17) Larsen, M.; Burken, J.; Machackova, J.; Karlson, U. G.; Trapp, S. Using Tree Core Samples to Monitor Natural Attenuation and Plume Distribution after a PCE Spill. *Environ. Sci. Technol.* **2008**, *42* (5), 1711–1717.
(18) Ahrens, L.; Felizeter, S.; Sturm, R.; Xie, Z.; Ebinghaus, R. Polyfluorinated Compounds in Waste Water Treatment Plant Effluents and Surface Waters along the River Elbe, Germany. *Mar. Pollut. Bull.* **2009**, *58* (9), 1326–1333.
(19) International Organization for Standardization. ISO 11465:1993 Soil Quality - Determination of Dry Matter and Water Content on a Mass Basis - Gravimetric Method. ISO 1993.
(20) Mclachlan, M. S.; Holmstrom, K. E.; Reth, M.; Berger, U. Riverine Discharge of Perfluorinated Carboxylates from the European Continent. *Environ. Sci. Technol.* **2007**, *41* (21), 7260–7265.
(21) Ahrens, L. Polyfluoroalkyl Compounds in the Aquatic Environment: A Review of Their Occurrence and Fate. *J. Environ. Monit.* **2011**, *13* (1), 20–31.
(22) Anderson, R. H.; Long, G. C.; Porter, R. C.; Anderson, J. K. Occurrence of Select Perfluoroalkyl Substances at U.S. Air Force Aqueous Film-Forming Foam Release Sites Other than Fire-Training Areas: Field-Validation of Critical Fate and Transport Properties. *Chemosphere* **2016**, *150*, 678–685.
(23) Dodgen, L. K.; Ueda, A.; Wu, X.; Parker, D. R.; Gan, J. Effect of Transpiration on Plant Accumulation and Translocation of PPCP/EDCs. *Environ. Pollut.* **2015**, *198*, 144–153.
(24) Krippner, J.; Brunn, H.; Falk, S.; Georgii, S.; Schubert, S.; Stahl, T. Effects of Chain Length and pH on the Uptake and Distribution of Perfluoroalkyl Substances in Maize (*Zea mays*). *Chemosphere* **2014**, *94*, 85–90.
(25) Rayne, S.; Forest, K. Perfluoroalkyl Sulfonic and Carboxylic Acids: A Critical Review of Physicochemical Properties, Levels and Patterns in Waters and Wastewaters, and Treatment Methods. *J. Environ. Sci. Health Part -ToxicHazardous Subst. Environ. Eng.* **2009**, *44* (12), 1145–1199.
(26) Chen, Y.-C.; Lo, S.-L.; Lee, Y.-C. Distribution and Fate of Perfluorinated Compounds (PFCs) in a Pilot Constructed Wetland. *Desalination Water Treat.* **2012**, *37* (1–3), 178–184.
(27) Bizkarguenaga, E.; Zabaleta, I.; Mijangos, L.; Iparraguirre, A.; Fernandez, L. A.; Prieto, A.; Zuloaga, O. Uptake of Perfluorooctanoic Acid, Perfluorooctane Sulfonate and Perfluorooctane Sulfonamide by Carrot and Lettuce from Compost Amended Soil. *Sci. Total Environ.* **2016**, *571*, 444–451.
(28) Zhao, S.; Fang, S.; Zhu, L.; Liu, L.; Liu, Z.; Zhang, Y. Mutual Impacts of Wheat (*Triticum aestivum L.*) and Earthworms (*Eisenia fetida*) on the Bioavailability of

Perfluoroalkyl Substances (PFASs) in Soil. *Environ. Pollut.* **2014**, *184*, 495–501.
(29) Felizeter, S.; McLachlan, M. S.; De Voogt, P. Root Uptake and Trans Location of Perfluorinated Alkyl Acids by Three Hydroponically Grown Crops. *J. Agric. Food Chem.* **2014**, *62* (15), 3334–3342.
(30) Krippner, J.; Falk, S.; Brunn, H.; Georgii, S.; Schubert, S.; Stahl, T. Accumulation Potentials of Perfluoroalkyl Carboxylic Acids (PFCAs) and Perfluoroalkyl Sulfonic Acids (PFSAs) in Maize (*Zea mays*). *J. Agric. Food Chem.* **2015**, *63* (14), 3646–3653.
(31) Lewis, J.; Qvarfort, U.; Sjostrom, J. Betula Pendula: A Promising Candidate for Phytoremediation of TCE in Northern Climates. *Int. J. Phytoremediation* **2015**, *17* (1), 9–15.
(32) Navarro, I.; de la Torre, A.; Sanz, P.; Pro, J.; Carbonell, G.; de los Angeles Martinez, M. Bioaccumulation of Emerging Organic Compounds (Perfluoroalkyl Substances and Halogenated Flame Retardants) by Earthworm in Biosolid Amended Soils. *Environ. Res.* **2016**, *149*, 32–39.
(33) Sijm, D.; Kraaij, R.; Belfroid, A. Bioavailability in Soil or Sediment: Exposure of Different Organisms and Approaches to Study It. *Environ. Pollut.* **2000**, *108* (1), 113–119.
(34) Hatzinger, P. B.; Alexander, M. Effect of Aging of Chemicals in Soil on Their Biodegradability and Extractability. *Environ. Sci. Technol.* **1995**, *29* (2), 537–545.
(35) Johansson, T. Biomass Production and Allometric above- and below-Ground Relations for Young Birch Stands Planted at Four Spacings on Abandoned Farmland. *Forestry* **2007**, *80* (1), 41–52.
(36) Johansson, Tord. *Growth and Yield in Shelter Stands of Silver Birch and Norway Spruce - Tillväxt Och Produktion Hos Skärmbestand Med Björk Och Gran*; Rapport 068; Swedish University of Agricultural Sciences, Department of Energy and Technology: Uppsala, 2014.
(37) Minerbi, S.; Cescatti, A. Tree Volume and Biomass Equations for *Picea abies* and *Larix decidua* in South Tyrol. *For. Obs.* **2015**, *7*, 5–34.
(38) Wu, X.; Conkle, J. L.; Ernst, F.; Gan, J. Treated Wastewater Irrigation: Uptake of Pharmaceutical and Personal Care Products by Common Vegetables under Field Conditions. *Environ. Sci. Technol.* **2014**, *48* (19), 11286–11293.
(39) Hurtado, C.; Dominguez, C.; Perez-Babace, L.; Canameras, N.; Comas, J.; Bayona, J. M. Estimate of Uptake and Translocation of Emerging Organic Contaminants from Irrigation Water Concentration in Lettuce Grown under Controlled Conditions. *J. Hazard. Mater.* **2016**, *305*, 139–148.
(40) Pietrini, F.; Di Baccio, D.; Iori, V.; Veliksar, S.; Lemanova, N.; Juskaite, L.; Maruska, A.; Zacchini, M. Investigation Onmetal Tolerance and Phytoremoval Activity in the Poplar Hybrid clone “Monviso” under Cu-Spiked Water: Potential Use for Wastewater Treatment. *Sci. Total Environ.* **2017**, *592*, 412–418.
(41) Valkonen, S.; Valsta, L. Productivity and Economics of Mixed Two-Storied Spruce and Birch Stands in Southern Finland Simulated with Empirical Models. *For. Ecol. Manag.* **2001**, *140* (2–3), 133–149.
(42) Algreen, M.; Trapp, S.; Rein, A. Phytoscreening and Phytoextraction of Heavy Metals at Danish Polluted Sites Using Willow and Poplar Trees. *Environ. Sci. Pollut. Res.* **2014**, *21* (15), 8992–9001.
(43) Riktvärden PFAS - SGI http://www.swedgeo.se/sv/om-sgi/vart-samhallsuppdrag/sarskilda-utredningar/riktvarden-pfas/ (accessed Feb 8, 2017).
(44) Cameron, A. D. Managing Birch Woodlands for the Production of Quality Timber. *Forestry* **1996**, *69* (4), 357–371.
(45) Jordbruksverket. Handbok för Salixodlare. Jordbruksverket.

(46) Schnoor, J. L. Phytoremediation. GWRTAC 1997.
(47) Tsang, W.; Burgess, D. R.; Babushok, V. On the Incinerability of Highly Fluorinated Organic Compounds. *Combust. Sci. Technol.* **1998**, *139* (1–6), 385–402.
(48) Vecitis, C. D.; Park, H.; Cheng, J.; Mader, B. T.; Hoffmann, M. R. Treatment Technologies for Aqueous Perfluorooctanesulfonate (PFOS) and Perfluorooctanoate (PFOA). *Front. Environ. Sci. Eng. China* **2009**, *3* (2), 129–151.