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Fly ash-based waste for *ex-situ* landfill stabilization of per- and polyfluoroalkyl substance (PFAS)-contaminated soil

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ABSTRACT

In response to world-wide soil and groundwater contamination per- and polyfluoroalkyl substances (PFAS), stakeholders require immediate mitigation. Soil deposition in landfill is a common mitigation scheme, but PFAS losses occur via landfill leachate. These leaching losses can be reduced by strategically utilizing other deposited waste materials for *ex-situ* contaminant stabilization. This screening study tested activated carbon (AC) and eight types of wastes (compost, rubber granulate, bentonite clay, industrial sludge, incineration slag, incineration bottom ash (n=4), incineration fly ash-based air pollution control residue (FA-APC) (n=16)) in amending (adding 4%, 5%, 10% or 25% sorbent) field-contaminated (n=19) and PFAS-fortified (n=11) soils. A subset of FA-based residue types, all originating from grate-fire incineration (G-F-I) plants, achieved extraordinarily high removal of PFAS. The removal was up to 98% (25% addition) of the sum of six dominant PFAS for field-contaminated soil and >99% of the sum of 11 PFAS for fortified soil (10/25% addition) (>99.9% for PFOS). Calculated partitioning coefficient revealed significant trends between sorption strength and perfluorocarbon chain length (0.21-0.47 log units per CF_2-moiety), indicating high importance of hydrophobic sorption (R^2>0.98). However, with incremental G-F-I FA-APC addition this relationship disappeared, indicating an alternative sorption mechanism. The exceptional PFAS sorption by G-F-I FA-APC was not explained by G-F-I surface area, surface charge, soil mineral- and metal composition, or solution DOC, metal, or ion composition (H^+, Ca^{2+}, Mg^{2+}, Al^{3+} and Ba^{2+}). Although the mechanism remains unknown, this study showed that landfill sites can utilize G-F-I FA-APC for *ex-situ* stabilization at negative cost, thus preventing PFAS-containing leachate.

1. Introduction

Continuous unsolicited contamination of groundwater and surface water by per- and polyfluoroalkyl substances (PFAS) has impacted drinking water sources worldwide, e.g., in Japan [1], Germany [2], and Sweden [3]. This contamination of drinking water often derives from unregulated use of PFAS-containing aqueous film-forming foams (AFFFs) [4] at firefighting training sites [5,6]. Few soil treatment technologies for PFAS-contaminated soils have been experimentally evaluated [7] and, due to the unique physiochemical properties of PFAS [8], it is not evident which established technique would be most efficient [9]. Previous studies on *in-situ* remediation techniques have shown promising results for phytoremediation [10], electrodialysis treatment [11], and thermal desorption [12,13]. Other promising *in-situ* laboratory- and field-scale remediation techniques include soil stabilization using activated carbons [11,14–16] and stabilization/solidification using cementitious binders and activated carbon as additives gave high rates of PFAS removal from leachate (>99% for PFOS from soil leachate) [17,18], but it does not remove all PFAS i.e. short-chained anionic species. Moreover, long-term removal rates have not yet been determined, so stakeholders remain hesitant about using this remediation technique for *in-situ* treatment. Due to the current lack of established off-the-shelf technologies for remediation of PFAS-contaminated soil, high-security landfills are often considered the best option for fast and safe treatment of contamination risk areas. However, concerns have been raised about this solution, since such landfills have been identified as considerable point sources of PFAS release to surface waters [17–19]. Although there are established and novel treatments technologies for

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PFAS removal from wastewater [20–22] and landfill leachate [23], efficient full-scale treatment systems have not been identified. Cost-efficient *ex-situ* stabilization techniques for PFAS-contaminated soil in landfill are needed to reduce PFAS release to leachate and groundwater (due to uncontrolled leakage from landfill) and to protect environmental and human health.

This study investigated the potential of a novel approach using other highly available landfill waste materials for cost-efficient *ex-situ* stabilization of PFAS-contaminated soil. Hereby, landfill waste materials such as fly ash was combined with PFAS-contaminated soil to reduce the environmental impact of PFAS-contaminated soil during deposition at e.g. landfills. The specific objectives were to (i) screen for high-removal landfill waste sorbents ($n = 23$), by analyzing amended field-contaminated soil for a wide range of PFAS ($n = 23$). For (ii), multiple ash types ($n = 12$) were used to stabilize PFAS-fortified soil ($n = 11$). For a better understanding of specifically high removal grate-fire incinerated (G-F-I) fly ash-based air pollution control residue (FA-APC) sorption mechanisms, surface area (BET), surface charge of materials (z-potential), pH, conductivity, aqueous metal (including multiple ions species) solution chemistry ($n = 15$), dissolved organic matter (DOC), solid metal and mineral content ($n = 27$), and total organic carbon (TOC) were determined.

2. Materials and methods

2.1. Analytical standards

The target PFAS ($n = 23$) were: C_3-C_{17} perfluoroalkyl carboxylic acids (PFCAs) (perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTriDA), perfluorohexadecanoic acid (PFHxDA), perfluorooctadecanoic acid (PFOcDA)), C_4, C_6, C_8 and C_{10} perfluoroalkyl sulfonic acids (PFSAs) (perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS), perfluorooctane sulfonic acid (PFOS), perfluorodecane sulfonic acid (PFDS)), 6:2, 8:2, 10:2 fluorotelomer sulfonic acids (FTSAs) and associated C_8 perfluorooctane sulfonamides (FOSAs) (FOSA, methyl perfluorooctane sulfonamide (MeFOSA), ethyl perfluorooctane sulfonamide (EtFOSA)), and C_8 perfluorooctane sulfonamidoacetic acid (FOSAAs) (ethyl-perfluorooctanesulfonamidoacetic acid (EtFOSAA)). In addition, 13 isotopically labeled internal standards (ISs) were included ($^{13}C_4$-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-MeFOSA, D_5-EtFOSA, and D_5-EtFOSAA). Abbreviation, supplier, and purity of the native PFAS and ISs are listed in Table S1 in Supplementary Information (SI).

2.2. Sampling and soil characterization

Field-contaminated soil was sampled at Stockholm Arlanda Airport, Sweden, in June 2018. Soil organic carbon content at sampling, determined as loss of ignition (Swedish standard SS-EN 15169:2007), was 4.0% and soil texture, determined by sieving (Swedish standard SS 027123, Swedgeo), consisted of 35% dry weight (dw) gravel (> 4 mm), 68% dw sand (>0.075 mm), and 0.41% dw (<0.075 mm) clay and silt. An artificial soil sample consisting of air-dried industrial sand (50-70 mm mesh particle size, Sigma Aldrich, Germany), kaolinite clay (Halloysite nanoclay, Sigma Aldrich, USA), and 10% organic carbon from peat originating from ECONOVA (Sweden) 2017 [24] was also prepared. A 0.5 kg portion of this artificial soil mixture was fortified in a 1-L PP bottle with a mixture of 18 PFAS to a concentration of 0.6 mg kg^{-1}, in accordance with our previous analyses [11,25], freeze-dried for seven days, and stored for aging for 2.5 years at 4°C.

2.3. Experimental batch leaching test

All leaching experiments on the field-contaminated soil were conducted as batch leaching tests at a liquid solid (L/S) ratio of 10, using 90 g dw of material and 0.9 L of water in 2-L bottles kept in a overhead shaker (Heidolph-Reax 20) at 7 rpm for 7 days until assumed equilibrium (Higgins and Luthy, 2006). For the fortified soil, 1.0 g of solid material and 10 mL MilliQ water were shaken in 15-mL PP tubes (Corning, Merck) kept in an end-over-end-shaker (Reax 2, Heidolph) at 200 rpm for 7 days until assumed equilibrium.

Two experiments were performed. First, a wide variety screening of waste sorbents (Table S2) and an activated carbon (Filtrasorb 400, Calgon Carbon, USA) were assessed for the field-contaminated soil, with an inclusion rate of 4% ($n = 6$), 10% ($n = 14$) and 25% dry dw ($n = 2$) for each sorbent. The waste sorbents comprised compost, rubber granulate, bentonite clay, industrial sludge, incineration slag, incineration bottom ash ($n = 4$), and incineration FA-APC ($n = 16$) (for detailed information on all sorbents, see Table S2 in SI). BA is the particulate matter that goes through the grate of the incinerators. FA-ACP in this study is defined as a mixture of fly ash (particulate matter removed from the flue gas stream) and the subsequent addition of lime and urea for sulfate and nitrate oxides flue gas removal. For some FA-ACP electrostatic precipitators were used for separating fly ash from the flue gas.

Soil and sorbents were combined at the same time in all cases. Preliminary testing identified Enk FA-APC (originating from a grate-fired incineration (G-F-I) plant) as promising, so additional G-F-I FA-APC samples ($n = 3$) at an inclusion rate of 10% dw ($n = 1$) were assessed as amendments for the field-contaminated soil. Secondly, a selection of previously tested ash samples ($n = 9$), at inclusion rates of 5% dw, 10% dw, and 25% dw, was assessed for sorption of 11 PFAS ($n = 1$) from the fortified soil (Table S2).

2.4. PFAS analysis

Two methods were used for analysis of PFAS in leachate: an *in-house* method targeting 26 PFAS and a commercial laboratory (SGS (former Synlab), Sweden) method (ISO 21675:2019), targeting 11 PFAS, accredited for drinking water analysis by the Swedish Food Agency [19]. The commercial laboratory method was used for initial screening for possible sorbents at 4% addition rate to naturally contaminated soil ($n = 11$ in duplicates including non-amended reference). All other PFAS analyses were performed using our validated *in-house* method [25]. In short, for the *in-house* PFAS leachate analysis, 500 μL leachate samples from the batch experiments were transferred to Eppendorf tubes (Merck, Germany) and fortified with 50 μL internal standard solution and 450 μL methanol. The samples were shaken at 200 rpm for 60 min, then centrifuged at 3700 rpm for 15 min and filtered through 0.45 μm recycled cellulose syringe filters (MiniSart RC, Sartorius, Germany) [27] into 2-mL amber auto-injection vials (Eppendorf, Germany). For soil measurement, 3.0 g dw homogenized soil was extracted for 3.0 h at 200 rpm in an end-over-end-shaker (Reax 2, Heidolph) in a two-step extraction (20+10 mL, in accordance with Ahrens et al. (2009)) using 80:20 methanol (LiChrosolv, Merck, Germany):sodium hydroxide (1 M in Millipore water) solution and fortified with 100 μL internal standard solution. A 7.5 mL aliquot was then concentrated under nitrogen gas to 500 μL, fortified with 500 μL Millipore water, and filtered through 0.45 μm recycled cellulose syringe filters into 2-mL amber auto-injection vials.

The instrument used for analysis was a DIONEX UltiMate 3000 ultra-performance liquid chromatography system (Thermo Scientific, Waltham, MA, USA) coupled to a triple quadrupole mass spectrometer (MS/MS) (TSQ QUANTIVA, Thermo SCIENTIFIC, Waltham, MA, USA). The PFAS standards were chromatographically separated on a BEH C18 column (1.7 μm, 50 mm, Waters Corporation, Manchester, UK), using acetonitrile and Millipore water with 5 mM ammonium acetate eluent gradient for 12 min and a flow rate of 0.5 mL min^{-1} in negative electrospray mode. A nine-point calibration curve (range 0.01-100 ng mL^{-1}) with a threshold linear regression curve fit of $R^2 > 0.99$ was used for quantification.

2.5. Additional analyses and calculations

Specific surface area (BET) was measured using an ASAP 2010 instrument (Micrometrics). Conductivity and pH were measured using a PW 9527 pH meter (Labassco Hanna instruments, USA) and conductivity meter (Metrohm, Switzerland), respectively. Surface charge (z-potential) was determined using a Zetasizer NanoZS device (25̊C, HeNe red laser, wavelength 633 nm; ZEN3600, Malvern Instruments). DOC was measured with a Multi N/C 3100 device (Analytik Jena AG. Jena, Germany). In the initial screening experiment, dissolved metal and ion concentrations in leachate were measured using inductively coupled plasma optical emission spectroscopy (ICP-OES; Perkin Elmer Optima 2000 DV) after digestion using the ASTM methods D3683 and D3682. For the other samples, dissolved metal, ion, and TOC concentrations in leachate were measured at ALS, Sweden, according to SS-EN ISO 11885:2009, CSN ISO 10694. Total solid metal and mineral concentrations were measured by Eurofins, Sweden, using SS-EN ISO 17294-2:2016.

Removal of PFAS from the aqueous phase (PFAS removal [%]) was calculated as the ratio of stabilized PFAS concentration in leachate $c_{amended}$ to that in reference soil leachate c_{REF}:

$$Removal\ [\%] = \left(1 - \frac{c_{amended}}{c_{REF}}\right) \times 100 \tag{1}$$

Sorption strength was calculated as partitioning coefficient $K_d[Lkg^{-1}]$ based on the ratio between solid (s) (sum of soil and sorbents) and aqueous (aq) phase as:

$$K_d\left[L\ kg^{-1}\right] = \frac{c_s}{c_{aq}} \tag{2}$$

where $c_{aq}$is the leachate concentration and c_s is the solid concentration after equilibrium ($c_{s,REF} - c_s$), where $c_{s,REF}$ is the PFAS concentration from measured solid soil extraction.

2.6. Quality assurance and quality control

For the *in-house* PFAS measurements, the limit of detection (LOD) was set to the lowest calibration point that did not deviate by >30% from the average response factor (ARF) and had a signal to noise ratio >3. In the laboratory blanks (only Millipore water), no PFAS was detected in concentrations above the LOD and thus the limit of quantification (LOQ) was set to LOD, which ranged between 0.5 and 5 ng mL^{-1} (Table S3 in SI). Sample losses combined with matrix effects were assessed by calculating internal standard absolute recoveries, comparing areas in samples and in calibration curves. Absolute internal standard recoveries were on average 88%, 88%, 94%, 96%, 100%, 107%, 114%, and 119% for PFHxA, FOSA, PFDA, PFOA, PFOS, PFHxS, PFNA, and PFUnDA, respectively. When screening novel sorbents for PFAS removal, PFAS were analyzed in leachate from duplicate experimental samples, while for ash amendment of fortified soil single samples were analyzed. For the latter, many of the ash types ($n = 11$) were tested at three different inclusion levels (5%, 10%, and 25%), allowing for good statistical resolution. Triplicate ($n = 3$) samples were used for analysis of initial soil concentration (liquid-solid extraction) and laboratory blanks. For duplicate leachate samples, relative variation between replicates was calculated as replicate difference divided by replicate average and was on average 12% ± 12% for the individual PFAS (highest for FOSA and PFDA, with on average 25%). Ash analysis (BET, metal and mineral content ((*aq.*) and (*s.*)), z-potential, TOC, DOC, pH, and EC) was performed for screening purposes on single samples and for a limited number of ash types and batch experiment samples. Hence, the results of some complementary analyses should be interpreted as indicative.

3. Results and discussion

3.1. Field-contaminated soil

3.1.1. Sorption and leaching of PFAS from field soil amended with FA-APC waste and other sorbents

Based on solid extraction measurements ($n = 3$), the field-contaminated soil contained 2160 μg $\sum$PFAS kg^{-1} dw soil, including 19 of the 26 target PFAS analyzed (PFOcDA, PFHxDA, PFDS, EtFOSA, EtFOSE, EtFOSAA, and MeFOSAA were not detected). PFOS comprised the largest fraction in total PFAS (61%), followed by FOSA (16%), 6:2 FTSA (7%), and FOSAA (5%). In the soil reference leachate (not treated), PFOS dominated, comprising 64% (60 μg L^{-1}) of total PFAS, followed by 6:2 FTSA (18%; 17 μg L^{-1}), PFPeA (3.6%; 3.4 μg L^{-1}), PFHxS (3.5%; 3.3 μg L^{-1}), FOSA (2.3%; 2.2 μg L^{-1}), PFOA (2.3%; 2.2 μg L^{-1}), 8:2 FTSA (2.0%; 1.8 μg L^{-1}), PFHxA (1.5%; 1.4 μg L^{-1}), and PFHpA (0.87%; 0.80 μg L^{-1}).

In the wider waste material screening of the waste sorbents (at 4%, $n = 10$) and additional FA-APC samples (at 10%, $n = 17$, at 25%, $n = 2$), only PFOS, 6:2 FTSA, PFHxS, FOSA, PFOA, and PFHpA ($\sum$PFAS$_6$) were consistently detected above LOD in the leachate samples (Fig. 1). Screening of the multiple sorbents revealed large variation in $\sum_6$PFAS sorption (stabilization/removal) ranging between -8% (4% OW BA) and >99% (4% AC) relative to the reference field-contaminated soil, where negative values mean that more PFAS was detected in leachate from amended soil compared with the reference. This can be explained by general low sorption potential together with increased pH, which is known to increase negative net surface charge on the sorbent and thus increase repulsion and reduce sorption [28]. The high removal of PFAS from leachate when using 4% AC is in agreement with previous findings of 3-4% AC soil amendment resulting in >99% PFOS removal [15,14], and of soil stabilization/solidification with 10% cementitious mixture and 0.2% AC reducing PFOS leaching by 99.9% [29]. The good sorption effect of AC has been attributed to its large surface area and it is already used for full-scale PFAS removal from drinking water [29].

Removal rate of $\sum$PFAS$_6$ by ash (4%, 10%, and 25% inclusion rate) ranged between -37% (4% Varta FA-APC) and 98% (25% Enk aged FA-APC). Although individual PFAS showed high variability in removal efficiency depending on the sorbent material, removal of individual

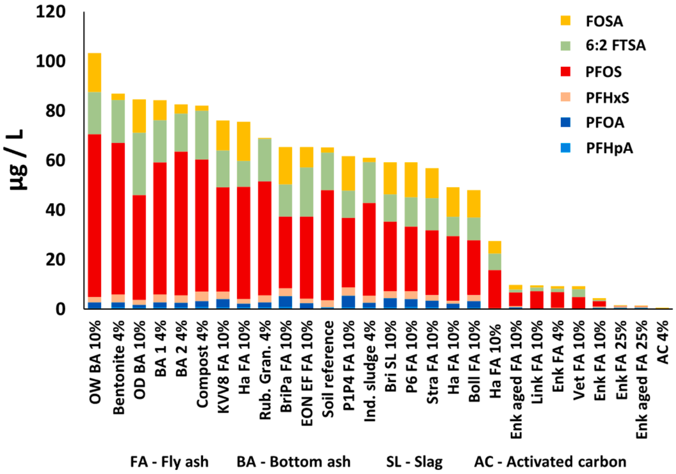

Fig. 1. Per- and polyfluoroalkyl (PFAS) (n = 6) mean concentration in batch experiment leachate (aq) from field-contaminated soil from Airport Arlanda, Sweden, amended with 4% by dry weight (soil reference 0% inclusion) of a wide variety of waste sorbent materials (organic wastes, clays, fly ash-based air pollution control residue (FA) and bottom (BA) ash, slags, activated carbon (AC)) or with 10-25% of selected ash sorbents.

PFAS correlated well with average removal of others (R^2 = 0.88; p<0.01, Figs. S1 and S2 in SI), with some materials achieving generally good sorption of many PFAS. A removal rate of 98% is high compared with that of other readily available waste materials used for soil stabilization (Zhang et al., 2019; Sörengård et al., 2020b; Militao et al., 2021). For example, PFOS removal from leachate of 31% and 24% has been reported when using 3% compost and montmorillonite (bentonite), respectively [14], while using 4% mixed wood or 4% paper mill waste biochar as an amendment has been found to have no effect on $\sum$PFAS retention in soil [15]). To the best of our knowledge, only one previous study has tested ash for PFOS stabilization, using maize straw-derived ash with high sorption capacity (log K_d 3.5-3.8) in a pure water/-sorbent system [31], which is one order of magnitude higher than in this study (log K_d = 2.6 for both 25% Enk aged FA-APC and 25% Enk FA-APC).

3.1.2. *Perfluorocarbon chain length, functional groups, and pH*

The general sorption behavior of $\sum_{17}$PFAS, as estimated by the median partitioning coefficient log K_d, was explained by perfluorocarbon chain length and functional groups (Fig. S3 in SI). Median log K_d value increased by 0.21 ($p<0.05$), 0.33 (p=0.070), and 0.42 ($p<0.001$) log units per –CF_2 moiety for PFCAs, PFSAs, and FTSAs, respectively. For different functional groups with the same perfluorocarbon chain length (C_8), the median log K_d value increased in the order: PFCA (0.13) < PFSA (0.69) < FOSAs (1.0) = FTSAs (1.0) < FOSAA (1.4). Such PFAS sorption behavior has been observed previously [26,30,32]. This indicates that hydrophobic sorption and electrostatic sorption are both important sorption mechanisms for PFAS [34]. However, the standard deviation when comparing all sorbents was high (n =) (Fig. S3 in SI), indicating varying sorption behavior for individual PFAS with different materials.

Ambient pH is known to be a critical variable for PFAS sorption strength [26,33,34], but only FOSA showed a significant correlation with pH in this study (Fig. 2). Thus, particularly low sorption of FOSA was observed for ash samples under basic conditions (pH between 9.1 and 12.5). This can be explained by the pK_a value of FOSA (6.2-6.5) [35], which results in FOSA being present mainly in its protonated state and thus being more water-soluble, as previously described by Sorengard et al. (2019). However, FOSA showed higher log K_d for sorption to AC and Enk FA-APC (aged and non-aged) compared with the other sorbents tested, indicating the presence of another sorption mechanism.

For AC, this is attributable to its large surface area [36], with e.g., Calgon carbon Filtrasorb 400 having surface area of around 1050 $m^2\,g^{-1}$ [37]. For Enk FA-APC, although the sample size was small (n = 7), sorption of PFOS (4.2 $m^2\,g^{-1}$) did not differ from that of other bottom or FA-APC types (range 2.7-45 $m^2 g^{-1}$) (Fig. S4 in SI).

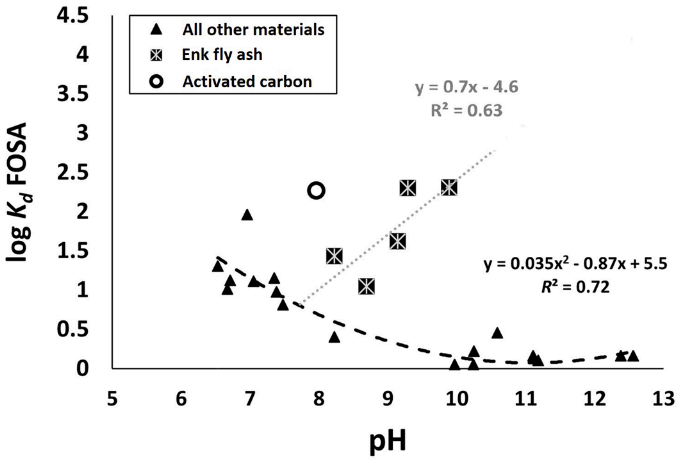

Fig. 2. Measured pH in relationship to sorption strength (log K_d) of perfluorooctane sulfonamidoacetic acid (FOSA) in batch tests in which per- and polyfluoroalkyl (PFAS)-contaminated field soil from Arlanda Airport, Sweden, was amended with 4%, 10%, or 25% of different sorbents.

3.2. *Fortified soil*

3.2.1. *Sorption and leaching of PFAS from PFAS-fortified soil amended with FA-APC waste*

In the in-depth study of different FA-APC types as sorbents for PFAS stabilization, three additional ashes (Ha rost FA-APC, Vet FA-APC, and Link FA-APC), originating from the same type of incineration process as the promising Enk FA-APC, were introduced. All these FA-APC types originate from similar grate-fired incinerators (G-F-I), primarily burning recycled wood. Unlike in the screening experiment (see Section 3.1), the amended soil was a standardized fortified soil including a wide range of PFAS (n = 14) at comparable concentrations (600 μg kg^{-1} for individual PFAS) and three different ash inclusion levels were tested (5%, 10%, and 25%).

As found in the screening experiment, most ash types showed low PFAS-stabilizing performance (Fig.e 3). The best sorption capacity was consistently shown by the subset of G-F-I FA-APC (i.e. Link FA-APC, Vet FA-APC, and Ha rost FA-APC, all at 25% level), with high PFAS removal (89-99%) from leachate. Among these, Vet FA-APC showed similar sorption capacity to AC for PFOS (mean >99% and 99.4%, respectively). Enk FA-APC showed lower sorption capacity (mean 65%) for the 5-25% amended fortified soil compared with the 10% amended field-contaminated soil (mean 98%). This was still a reasonably good amendment effect, but it raises questions regarding inconsistencies in stabilization performance over time and/or with soil type.

3.2.2. *Perfluorocarbon chain length and functional groups*

Perfluorocarbon chain length showed no significant correlation ($p>0.05$) with mean PFAS removal rate for any of the sorbents tested (n = 35). However, on comparing compounds with different functional groups among PFAS with the same perfluorocarbon chain length, FTSAs and PFSAs showed high removal (mean 75 and 60% reduction, respectively) compared with PFCAs (mean 29% reduction) (Fig. S5 in SI). This indicates that the sorption mechanism is influenced by ionic strength, with electrostatic differences between sulfonic and carboxylic functional groups being well documented [28]. On the other hand, the G-F-I FA-APC types, which showed the highest sorption capacity for PFAS, showed no difference in sorption capacity on comparing the functional groups, but displayed a significant increasing correlation with increasing perfluorocarbon chain length for PFCAs (3.9% increase per -CF^2-moiety; R^2 = 0.97, $p<0.01$), indicating a hydrophobic sorption mechanism.

For the FA-APC-amended fortified soil (5%, 10%, and 25% inclusion, n = 35), the log K_d values for PFCAs$_{5\text{-}7}$ showed no change, but those for PFCAs$_{7\text{-}11}$ increased significantly, by 0.47 log units for each –CF_2 moiety (R^2 = 0.98, $p<0.001$) (Fig. 4). This is in agreement with previous findings and is mainly explained by combined hydrophobic and electrostatic sorption mechanisms [26,38,39]. However, for the high-performing G-F-I FA-APC types, the relationship between sorption strength (log K_d) and perfluorocarbon chain length for PFCAs was weaker (Fig. 4). For 5% G-F-I FA-APC addition the relationship was still significant (R^2 = 0.62, $p<0.001$), whereas for 10% G-F-I FA-APC addition the significance level was lower (R^2 = 0.23, $p<0.05$) and for 25% G-F-I FA-APC addition no significant relationship was observed. The comparatively low effect of perfluorocarbon chain length on PFCA sorption strength at 10% and 25% G-F-I FA-APC addition confirms that this type of ash employs a different sorption mechanism than the other materials tested.

3.3. *Chemistry of grate-fired incineration (G-F-I) ash*

The G-F-I ashes (n = 4) at 5%, 10%, and 25% inclusion level displayed on average around 63%, 48%, and 50% higher sum of 11 PFAS removal, respectively, than the other ashes (n = 8). Link FA-APC 25%

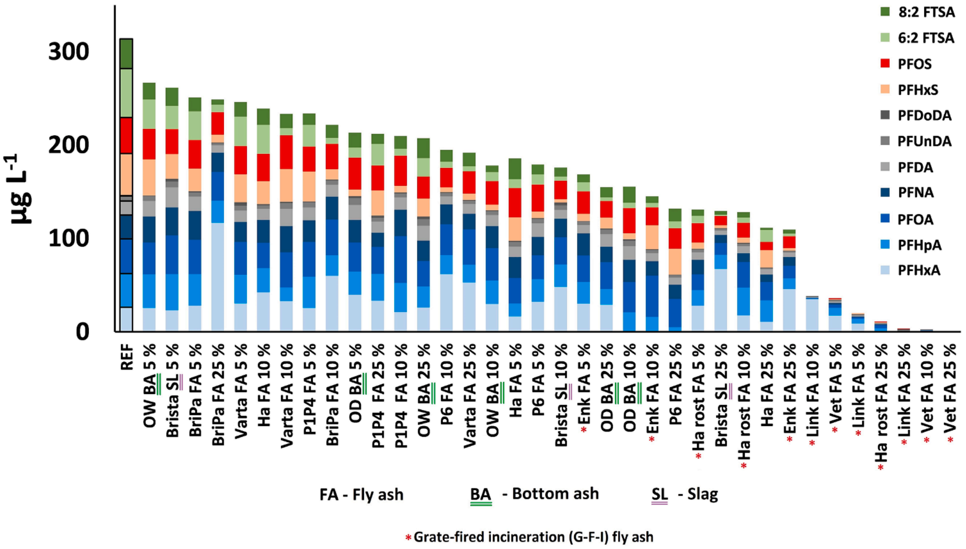

Fig. 3. Per- and polyfluoroalkyl (PFAS) concentration in batch experiment leachate (aq) from standardized PFAS-fortified soil amended with 5-25% (REF = 0% amendment) of a variety of incineration slag (SL), bottom (BA) and fly ash-based air pollution control residue (FA) sorbents.

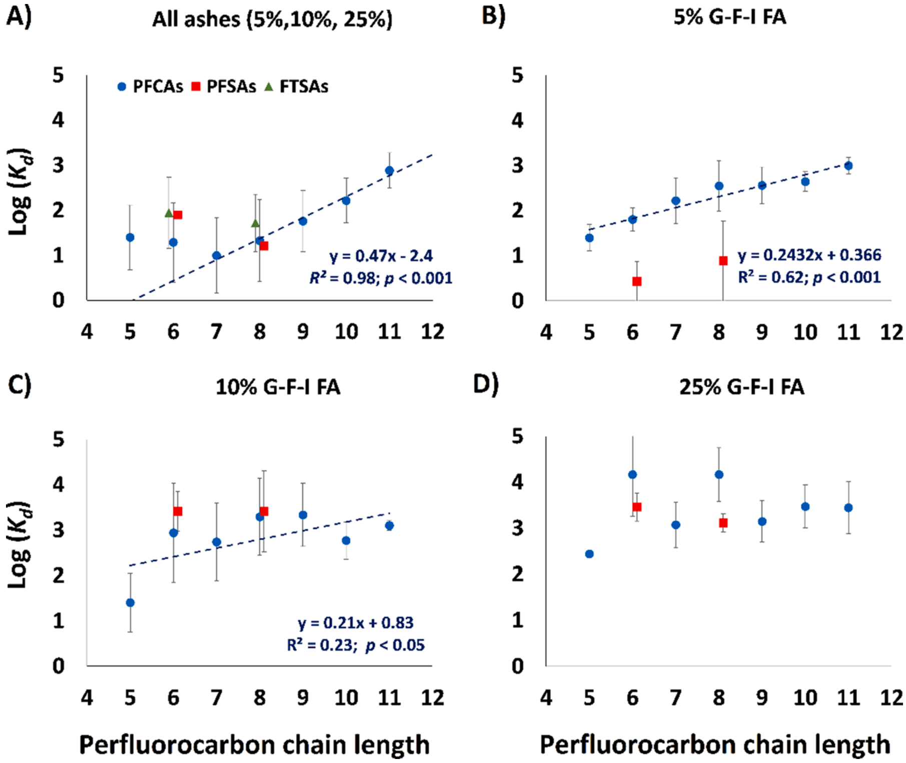

Fig. 4. Estimated per- and polyfluoroalkyl (PFAS) sorption strength (log K_d) as a function of perfluorocarbon chain length for batch experiment leachate from standardized PFAS-fortified soil amended with 5-25% of ash sorbent. Results for (A) all ash types and amendment levels (see Fig. 3) and (B-D) grate-fired incineration (G-F-I) fly ash-based air pollution control residues (FA) at 5%, 10% and 25% inclusion rate, respectively.

and Vet 10%/25% showed similar performance to 4% AC in average PFAS removal from leachate. Previous studies have shown that an important factor influencing PFAS sorption is the ionic composition of a solution, i.e., H^+, Ca^{2+} Mg^{2+} and Al^{3+} concentrations [38,40]. To assess this as a possible sorption mechanism for the different FA-APC types tested, we analyzed metal concentrations in solution. We detected metals in 15 out of 19 solutions tested, but only Ca^{2+} ($R^2 = 0.31$, $p<0.05$) and Ba^{2+} ($R^2 = 0.59$, $p<0.001$) showed a significant correlation with PFAS sorption for the field-contaminated soil ($n = 34$) (Fig. 5 and Fig. S6 in SI).

Solution concentrations of cationic divalent (Ca^{2+} and Mg^{2+}) and trivalent (Al^{3+}) ions have been shown previously to govern PFAS sorption [26,34,38]. Therefore, we performed an additional experiment to assess the role of divalent ion concentrations in PFAS sorption by adding Ba^{2+} ions (50-20000 mg $BaCl_2$ L^{-1} ($n = 9$)) to pure fortified soil and soil amended with the low-sorbing 10% Ha FA-APC (non-G-F-I ash) (Fig. S7 in SI). The results showed no correlation between divalent ion sorption effect and PFAS sorption, which indicates that cationic ions in solution (i.e., soil pore water) have little to no effect in ash-amended soil stabilization. For the different ash types tested on PFAs-fortified soil, we

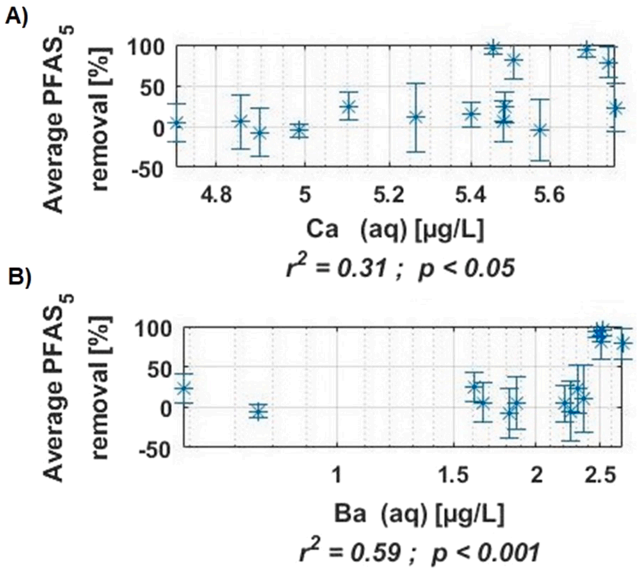

Fig. 5. Correlations between average of $\sum_5$PFAS (PFAS$_5$) removal [%] (PFHpA, PFOA, PFHxS, PFOS and 6:2 FTSA) from leachate in screening batch experiments using 10% fly ash-based air pollution control residue (FA-APC) amendment to PFAS field-contaminated soil and the FA-APCs inorganic compositions [μg / L] (log-scale).

found no significant correlations between ionic strength (conductivity [mS cm^{-1}]), pH, z-potential, or dissolved organic matter (DOC) and PFAS sorption strength (Fig. S8 in SI). Similarly, we found no relationship between solid metal concentration ($n = 27$), including the minerals P_2O_3, K_2O, Fe_2O_3, Al_2O_3 and CaO and PFAS sorption (Fig. S9 in SI). In contrast, previous studies have found that these parameters can influence PFAS sorption [36,32,41,42].

Ultimately, the results from the ash amendment experiments on PFAS-fortified soil showed that electrostatic and hydrophobic generally governed the sorption mechanisms for ash, as concluded in most other PFAS sorption studies [34]. However, no chemical variable relating to electrostatic (ions, pH, conductivity and z-potential) and hydrophobic sorption mechanisms of the materials (BET, solid species, DOC, and TOC) was identified as the main impact factor for PFAS sorption. Although some of the variables tested (pH and soil organic carbon) have been demonstrated to affect PFAS sorption in isolated laboratory tests [39], they show a limited degree of explanation for sorption in complex matrixes [43]. Thus, the mechanism behind the strong sorption of PFAS to G-F-I FA-APC sorbents still need to be identified. One possible explanation is that sorption of PFAS is governed by inner-sphere complexation, e.g., to alumina and iron oxides [44,45], but this must be studied in greater depth. The complex matrix present in ash, comprising a wide range of physiochemical properties unique to each incineration plant [46], could contain previously unstudied solid-phase constituents capable of inner-sphere complexation with PFAS. A previous study showed that modifying an inefficient PFAS industrial waste FA-APC sorbent with quaternary ammonium groups, and thus introducing net cationic surface charge and anion-exchange properties, can create a sorbent superior to AC materials [47]. It is therefore also possible that G-F-I FA-APC is modified during the incineration process to acquire similar properties, and that net cationic minerals with anion-exchange properties (e.g., hydrotalcites) can be formed from FA-APC [48]. These minerals, with double-layer hydroxides, can have higher PFAS sorption capacity than AC materials [49]. Although the sorption mechanisms were not identified for the G-F-I FA-APC types tested in the present study, all types displayed excellent sorption properties and are suitable for use in PFAS stabilization in soil. Further studies are required to identify the sorption mechanism associated with G-F-I ashes. Use of G-F-I FA-APC is a cost-efficient alternative to activated carbon, since around 200 000 tons of FA-APC are landfilled annually in Sweden which can have high economic and environmental value. The approach of *ex-situ* landfill stabilization with G-F-I FA-APC is patented (SE-543514 C2). Beside mixing of soil and amendment material, stabilization and solidification is an efficient alternative ex-situ treatment technique for PFAS-contaminated soil [25]. The technique combines the chemical immobilization of contaminants with considerably reducing the water permeability by using a strong cementitious binder . Some FA ACP can have good cementitious binding properties and are often used as a complimentary addition to Portland cement for economic and environmental reasons. Because of the excellent chemical PFAS stabilization properties of G-I-F FA-ACPs, the use of this material in S/S treatment context is promising and should be further investigated. Still, it shall be remembered that the FA-ACPs are hazardous materials containing (Fig. S9) and leaching (Fig. S6) heavy metals above threshold values for hazardous waste (NFS 2004:10, Swedish protection agency) and this is another aspect needed to account for when considering this approach.

4. Conclusions

Addition of 4% AC or 5-25% grate-fired incineration (G-F-I) fly ash-based air pollution control residues (FA-APC) to PFAS-contaminated soil reduced total PFAS concentration in leachate by 99% or 58-99%, respectively. However, not all ash types tested showed suitability for PFAS stabilization in soil. Most types showed interchangeable hydrophobic and electrostatic sorption mechanisms known from other PFAS sorption studies, but analysis of a subset of high-performing G-F-I FA-APC types indicated presence of another unidentified sorption mechanism. Challenges remain in using measured material and chemical properties to predict PFAS sorption performance for ash products in general and for G-F-I ashes in particular. Further studies should seek to identify the sorption mechanism associated with G-F-I ash, since low-cost, high-performance materials are needed for future up-scaling of stabilization work on PFAS-contaminated soils.Eqn 1-(2)

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Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: The study was conducted in collaboration with R&D division waste management company Ragn-Sells AB, Sweden, and the results were used for the patent SE-543514 C2. Co-author and developer Dr. Igor Travar works at this company and has been objective and scientific in this study. Remaining authors have no competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ceja.2022.100396.

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