Modified Clays as Effective Soil Amendment to Reduce Leaching of Per- and Polyfluoroalkyl Substances from Contaminated Soils

Chenyang Wang

Department of Civil Engineering and Applied Mechanics

McGill University, Montreal, Quebec, Canada

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Abstract

Repeated discharges of aqueous film-forming foams (AFFFs) in designated firefighting training areas over decades are closely linked to severe soil and groundwater contamination by per- and polyfluoroalkyl substances (PFASs). Increasingly stringent regulations require mitigation measures and treatment technologies that can effectively control the spread of PFASs or clean up impacted sites. A broad array of treatment technologies has been developed for treating contaminated water, while few have focused on effective soil treatment technologies that are available to contaminated site practitioners. The literature has documented that modified clays, produced by inserting organic cations (e.g. quaternary ammonium surfactant) in the exchange sites of extendable clays (e.g. smectite clays), can immobilize a range of organic contaminants in soil remediation. The goal of this study is to evaluate the performance of modified clays (bentonite-based materials) as a soil amendment in reducing the mobility or leachability of the dozens of PFASs detected in impacted soils, which may contribute to developing low-cost and easy-to-implement soil remediation technology.

PFAS profiles of four AFFF-contaminated soils and the soil leachates were first characterized using UHPLC coupled with high-resolution mass spectrometry to demonstrate the significant presence of anionic, cationic, neutral and zwitterionic PFAS. In the first stage, a soil leachability test modified based on the USEPA method 1311 Toxicity Characteristic Leaching Procedure (TCLP) was used to determine the effect of sorbent dosage, equilibration time, and potential microbial activities using a heavily contaminated soil. The significant decrease (95~99%) in aqueous concentrations of anionic PFAS from the soil leachate, including perfluorooctane sulfonate (PFOS), perfluorohexane sulfonate (PFHxS), and perfluorooctane carboxylate (PFOA), can be achieved in 1-4 days with a dosage as low as 0.5% (w/w). There was also a significant decrease of several cationic and neutral/zwitterionic PFAS (70~99%), but a higher dosage was necessary to achieve removal similar to the anionic PFASs. Meanwhile, the solution pH and ionic strength of extraction fluids did not show any detectable impact on PFAS leachability. In the second stage, a comparative assessment was conducted among the modified clay, granular activated carbon (GAC), and biochar. Both modified clays and GAC achieved significant PFAS decrease in aqueous concentration whereas negligible PFASs decrease was found in biochar-amended soil leachates. Modified clays performed better than GAC in the uptake of anionic and neutral/ zwitterionic PFASs, while GAC was the most effective sorbent in preventing the leaching of cationic PFASs. In the third stage, the performance of modified clays on the cationic PFAS was improved by adding a natural bentonite clay, which can be attributed to the strong affinity of natural montmorillonite minerals for cationic PFASs. The findings suggest that the combined uses of natural and modified bentonite clay minerals present a viable low-cost and effective solution in soil remediation of PFASs.

Résumé

Les rejets répétés de mousses filmogènes aqueuses (AFFF) dans des zones d'entraînement désignées pour la lutte contre les incendies au cours de plusieurs décennies sont étroitement liés à la grave contamination des sols et des eaux souterraines par des substances perfluorées et polyfluoroalkyles (PFAS). Des réglementations de plus en plus strictes exigent des mesures d'atténuation et des technologies de traitement permettant de contrôler efficacement la propagation des PFAS ou de nettoyer les sites touchés. Un large éventail de technologies de traitement a été mis au point pour traiter l'eau contaminée, alors que peu se sont concentrées sur les technologies de traitement des sols efficaces disponibles pour les praticiens de sites contaminés. La littérature a documenté que les argiles modifiées, produites en insérant des cations organiques (par exemple un surfactant ammonium quaternaire) dans les sites d'échange d'argiles extensibles (par exemple des argiles à smectite), peuvent immobiliser une gamme de contaminants organiques dans l'assainissement des sols. L'objectif de cette étude est d'évaluer la performance des argiles modifiées (matériaux à base de bentonite) en tant qu'amendement du sol permettant de réduire la mobilité ou la lixiviabilité des dizaines de PFAS détectés dans les sols impactés, ce qui pourrait contribuer à la mise au point de solutions peu coûteuses et faciles à utiliser. mettre en œuvre une technologie de dépollution des sols.

Les profils PFAS de quatre sols contaminés par AFFF et des lixiviats de sol ont d'abord été caractérisés à l'aide d'une CLHP-U couplée à une spectrométrie de masse à haute résolution afin de démontrer la présence significative de PFAS anioniques, cationiques et neutres / zwitterioniques. Au cours de la première étape, un test de lixiviabilité dans le sol modifié selon la méthode 1313 de la lixiviation de la toxicité caractéristique (TCLP) de la méthode USEPA a été utilisé pour déterminer l'effet de la dose de sorbant, du temps d'équilibrage et des activités microbiennes potentielles sur un sol fortement contaminé. La réduction significative (95 ~ 99%) des PFAS anioniques du lixiviat du sol, y compris le perfluorooctane sulfonate (SPFO), le perfluorohexane sulfonate (PFHxS) et le perfluorooctane carboxylate (PFOA), peut être atteinte en 1 à 4 jours avec une dose aussi faible. comme 0,5% (p / p). Il y avait également une réduction significative de plusieurs PFAS cationiques et neutres / zwitterioniques (70 ~ 99%), mais une posologie plus élevée était nécessaire. Pendant ce temps, le pH de la solution et la force ionique des fluides d’extraction ne montrent aucun impact détectable sur la lixiviabilité du PFAS. Dans la seconde étape, une évaluation comparative a été réalisée entre l'argile modifiée, le charbon actif en grains (GAC) et le biochar. Les argiles modifiées et le GAC ont permis une réduction significative du PFAS, tandis qu'une réduction négligeable du PFAS a été constatée dans les sols modifiés au biochar. Les argiles modifiées ont mieux performé que le CAG en ce qui concerne l'absorption de PFAS anioniques et neutres / zwitterioniques, tandis que le CAG était le sorbant le plus efficace pour prévenir la lixiviation des PFAS cationiques. Dans la troisième étape, la performance des argiles modifiées sur le PFAS cationique a été améliorée en ajoutant une argile bentonite naturelle, ce qui peut être attribué à la forte affinité des minéraux naturels de montmorillonite pour les PFAS cationiques. Les résultats suggèrent que les utilisations combinées des minéraux d'argile naturels et modifiés de l'argile bentonite constituent une solution viable, peu coûteuse et efficace, pour l'assainissement des PFAS par le sol.

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Nomenclature

Acronym	**Name**
PFAS	Perfluoroalkyl and polyfluoroalkyl substances
AFFFs	Aqueous film-forming foams
PFAA	Perfluoroalkyl acid
PrePFAA	Precursors to perfluoroalkyl acid
PFCA	Perfluoroalkyl carboxylic acid
PFPrA	Perfluoropropanoic acid
PFBA	Perfluorobutanoic acid
PFPeA	Perfluoropentanoic acid
PFHxA	Perfluorohexanoic acid
PFHpA	Perfluoroheptanoic acid
PFOA	Perfluorooctanoic acid
PFNA	Perfluorononanoic acid
PFDA	Perfluorodecanoic acid
PFUdA	Perfluoroundecanoic acid
PFDoA	Perfluorododecanoic acid
PFTrDA	Perfluorotridecanoic acid
PFTeDA	Perfluorotetradecanoic acid
PFSA	Perfluoroalkyl sulfonic acid
PFPrS	Perfluoropropane sulfonate
PFBS	Perfluorobutane sulfonate
PFPeS	Perfluoropentane sulfonate
PFHxS	Perfluorohexane sulfonate
PFHpS	Perfluoroheptane sulfonate
PFOS	Perfluorooctane sulfonate
PFNS	Perfluorononane sulfonate

Acronym	**Name**
PFDS	Perfluorodecane sulfonate
PFECHS	Perfluoro-4-ethylcyclohexanesulfonate
FBSA	Perfluorobutane sulfonamide
FHxSA	Perfluorohexane sulfonamide
FOSA	Perfluorooctane sulfonamide
MeFOSA	Methyl-perfluorooctane sulfonamide
EtFOSA	Ethyl-perfluorooctane sulfonamide
FOSAA	Perfluorooctane sulfonamideacetic acid
MeFOSAA	Methyl-perfluorooctane sulfonamidoacetic acid
EtFOSAA	Ethyl-perfluorooctane sulfonamidoacetic acid
4:2 FTSA	4:2 Fluorotelomer sulfonic acid
6:2 FTSA	6:2 Fluorotelomer sulfonic acid
8:2 FTSA	8:2 Fluorotelomer sulfonic acid
10:2 FTSA	10:2 Fluorotelomer sulfonic acid
3:3 FTCA	3: 3 Fluorotelomer carboxylic acid
4:3 FTCA	4: 3 Fluorotelomer carboxylic acid
5:3 FTCA	5: 3 Fluorotelomer carboxylic acid
7:3 FTCA	7: 3 Fluorotelomer carboxylic acid
6:2 FTUA	6: 2 Fluorotelomer unsaturated acid
8:2 FTUA	8: 2 Fluorotelomer unsaturated acid
10:2 FTUA	10: 2 Fluorotelomer unsaturated acid
PFHxSAm	Perfluorohexanoic sulfonamidoalkyl amine
PFOSAm	Perfluorooctane sulfonamidoalkyl amine
PFHxSAmS	Perfluorooctane sulfonamidoalkyl ammonium salt
PFOSAmS	Perfluorooctane sulfonamidoalkyl ammonium salt
PFOAAmS	Perfluorooctaneamide ammonium salt
6:2 FTAB	6:2 Fluorotelomer sulfonamide betaine

Acronym	**Name**
PFOAB	Perfluorooctaneamidoalkyl betaine
PFOSB	Perfluorooctanesulfonamidoalkyl betaine
PFOANO	Perfluorooctane alkylamido amine oxide
PFOSNO	Perfluorooctane sulfonamidoalkyl amine oxide
MPFBA	Perfluoro-n-[$^{13}C_4$]butanoic acid
M5PFPeA	Perfluoro-n-[$^{13}C_5$]pentanoic acid
M5PFHxA	Perfluoro-n-[1,2,3,4,6-$^{13}C_5$]hexanoic acid
M4PFHpA	Perfluoro-n-[1,2,3,4-$^{13}C_4$]heptanoic acid
M8PFOA	Perfluoro-n-[$^{13}C_8$]octanoic acid
M9PFNA	Perfluoro-n-[$^{13}C_9$]nonanoic acid
M6PFDA	Perfluoro-n-[1,2,3,4,5,6-$^{13}C_6$]decanoic acid
M7PFUdA	Perfluoro-n-[1,2,3,4,5,6,7-$^{13}C_7$]undecanoic acid
MPFDoA	Perfluoro-n-[1,2-$^{13}C_2$]dodecanoic acid
M2PFTeDA	Perfluoro-n-[1,2-$^{13}C_2$]tetradecanoic acid
M3PFBS	Sodium perfluoro-1-[2,3,4-$^{13}C_3$]butanesulfonate
M3PFHxS	Sodium perfluoro-1-[1,2,3-$^{13}C_3$]hexanesulfonate
M8PFOS	Sodium perfluoro-1-[$^{13}C_8$]octanesulfonate
M6:2 FTUA	2H-Perfluoro-[1,2-$^{13}C_2$]-2-octenoic acid
M8:2 FTUA	2H-Perfluoro-[1,2-$^{13}C_2$]-2-decenoic acid
M6:2 FTSA	Sodium 1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]-octane sulfonate ***(6:2)***
M8:2 FTSA	Sodium 1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]-decane sulfonate ***(8:2)***
d5-EtFOSA	N-ethyl-d_5-perfluoro-1-octane sulfonamide
d-MeFOSA	N- methyl-d_3-perfluoro-1-octane sulfonamide
d-MeFOSAA	N-methyl-d_3-perfluoro-1-octanesulfonamidoacetic acid
M8FOSA	Perfluoro-1-[$^{13}C_8$]octanesulfonamide
ACN	Acetonitrile
MeOH	Methanol

Acronym	**Name**
FS100	FLUORO-SORB100®
GAC	Granular activated carbon

Contribution of Authors

This thesis is presented in a traditional format. Chapters 1 and 2 are introduction and literature review, followed by materials and methods (chapter 3). Chapter 4 presents the results and discussions, and Chapters 5 and 6 are conclusions and suggested future directions. Supplementary information is provided at the end of the thesis.

The candidate Chenyang Wang, under the supervision of Dr. Jinxia Liu, conducted the design and performance of the experiment work, data analysis, and manuscript writing. Dr. Bei Yan (McGill University) contributed to revising the experimental design and performing part of the instrumental analysis. Dr. Gabriel Munoz (Université de Montréal) contributed to performing part of the instrumental analysis.

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1. Introduction

1.1 Background

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) have been widely detected in environmental compartments [1-5], wildlife [6], and humans [7-9]. It is a critical issue since PFASs are chemically persistent [10], bioaccumulative [6, 9], and can pose risks to humans and wildlife [11-16]. The ubiquitous distribution of PFASs is due to their wide applications in various industries and products [17], with an important application of PFASs as the major components in aqueous film-forming foams (AFFFs) that are used for fighting Class B fires. PFASs effectively lower the surface tension and accelerate the formation of the aqueous film, which cuts off oxygen contact to extinguish hydrocarbon-based fuel fires [18].

Repeated discharges of AFFFs in designated firefighting training areas over decades are closely linked to severe soil and groundwater contamination by PFASs [1, 5, 10, 19, 20]. In many instances, PFASs are directly discharged into soils with little or no treatment, and thereby leaching from soils contributes to PFASs load in groundwater and surface water. The elevated levels of PFASs are widely observed in the soil and water samples geographically relevant to the impacted sites, even when the regular discharges of AFFFs discontinued for years [1, 10].

Increasingly stringent regulations require treatment technologies that can effectively control the spread of PFASs or clean up impacted sites. Despite many efforts to develop innovative treatment technologies, the treatment technologies that can be applied to a large scale, being cost-effective, easy to implement and without giving rise to secondary issues, are very limited. Activated carbon and ion exchange resin are probably the only ones that have been used in actual projects of AFFF-impacted water treatment [21, 22]. In addition, most of the development efforts have been targeting anionic PFASs, such as perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA). Only a few studies examined the treatment efficacy towards non-anionic PFASs. Furthermore, few available treatment technologies focus on soil, which can retain a significant amount of PFASs; many PFASs are highly hydrophobic or have functional groups that engage strong soil interactions.

PFOS and PFOA are the most detected and studied PFASs at AFFF-impacted sites. In recent years, an increasing number of classes of novel PFASs have been identified in AFFF concentrates [20, 23-27]. The novel PFASs span various types of anionic, zwitterionic, neutral, and cationic fluorosurfactants, containing the functional groups such as ammonium, amine oxide, sulfonamide, betaine and many others. Most of the novel PFASs are considered as the "precursors" to perfluoroalkyl acids (PFAAs) since they have the potential to biologically or chemically degrade to perfluoroalkyl carboxylic acids (PFCAs) or perfluoroalkyl sulfonic acids (PFSAs) [4, 10, 26]. The previous investigations focus mainly on legacy anionic groups (i.e. PFCA and PFSA groups) with little consideration of the significant presence of the non-anionic groups.

Increasingly stringent regulations require treatment technologies that can effectively control the spread of PFASs or clean up impacted sites. So far, a broad array of treatment technologies has been studied for treating PFAS contaminated water, while few treatment technologies for contaminated soils have been developed or even attempted. Technologies targeting water treatment in full-scale installations include granular activated carbon (GAC) adsorption, ion exchange, and membrane filtrations (e.g. reverse osmosis) [21, 22]. Sorption-based physical separation is the prevailing approach to control PFAS in real-world water treatment scenarios. A similar technique that can be used towards soil remediation is using sorbent amendment, which relies on sorbents that have a high affinity for target contaminants to stabilize or immobilize contaminants. The method has been proven effective in reducing the leachability and bioavailability of some organic pollutants such as PCB, PAHs, and DDT in soils [28-31], while limited studies have been conducted targeting PFASs, even less non-anionic PFASs.

In a few reported studies focusing on PFAS, a range of soil amendments have been evaluated, including activated carbon, compost soil, montmorillonite, modified palygorskite (MatCARE^{TM}), biochar, pulverized zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride [32-35]. The studies concluded that activated carbon and modified palygorskite performed significantly better than other materials in preventing PFAS leaching. Compared to the single study on using modified palygorskite to retain PFASs, there is a rich literature on the use of modified clays for water treatment and soil remediation targeting a range of inorganic and organic pollutants [36-41]. Modified clays are made by inserting cation surfactants (e.g., quaternary ammonium surfactant) in the exchange sites of expandable clays such as smectite. The promising preliminary data suggest the modified clay can be a low-cost alternative to activated carbon for immobilizing PFAS, particularly for soil remediation or mitigation purposes [40]. Therefore, there is a strong need to develop further understanding of the efficacy and efficiency of modified clays in PFAS treatment and elucidate sorption mechanisms.

The overarching goal of the research is to evaluate the performance of modified clays as a soil amendment in reducing the mobility or leachability of anionic, cationic, neutral and zwitterionic PFAS groups detected in AFFF-impacted soils. The laboratory-scale batch tests modified based on a USEPA method were conducted to evaluate the leachability of PFAS in four contaminated soils with and without sorbent amendment. Leaching kinetics, required dosage of modified clays, and the effect of solution pH were assessed using the most heavily contaminated soil. A comparative assessment of modified clays, granular activated carbon (GAC) and biochar was provided as well. Finally, a potential approach to improve the performance of modified clays on cationic PFAS reduction in practical applications was proposed and tested. The findings provide robust data to allow future applications of the low-cost and easy-to-implement soil remediation technology for PFAS-contaminated soils.

1.2 Research Objectives and Approaches

Four specific objectives were proposed.

- Objective 1 was to demonstrate the significant presence of anionic, cationic, and neutral/ zwitterionic PFAS in AFFF-impacted soil and soil leachates;
- Objective 2 was to evaluate the efficacy and efficiency of new commercially available modified clays in reducing PFAS leaching;
- Objective 3 was to provide a comparative assessment of the modified clays, granular activated carbon and biochar;
- Objective 4 was to develop a method to improve the performance of modified clays in practical applications.

The study utilized batch sorption and desorption tests to evaluate the performance of a newly developed modified clay that is manufactured by CETCO (USA), a division of Mineral Technologies, Inc. Since there is no acceptable leachability standard method that has been specifically developed for PFASs, the existing US EPA method for evaluating leachability of solid hazardous materials was modified and applied to four field contaminated soils. The study included 49 quantitatively evaluated PFAS, for which authentic standards are available, and cover anionic, cationic, zwitterionic and neutral groups. Chemical analysis was performed using UHPLC coupled with high-resolution mass spectrometry.

2. Literature Review

2.1 PFAS Properties and Applications

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are highly fluorinated aliphatic substances that feature the hydrophobic moiety of $–C_nF_{2n+1}–$. PFAS-based surfactants contain one perfluoroalkyl tail and one hydrophilic head group [42]. The number and positions of fluorine atoms are closely related to the properties of PFAS [17]. Due to the strong C-F bond, many PFAS compounds are remarkably stable under the heat, acid/base, and reduction/oxidation condition [17, 43]. Meanwhile, the perfluoroalkyl tail is not only hydrophobic but also lipophobic, which imparts unique properties for some PFAS to repel both water and oil or fat [17, 44]. As surfactants, PFASs are more surface-active than the corresponding hydrocarbon surfactants; a similar level of surface tension reduction can be achieved with a smaller quantity of fluorinated surfactants [17].

Due to the above properties, PFASs have been included in various industries and products over the past six decades, such as protective coating, textiles, electronics, antifogging agents, adhesives, cosmetics, antistatic agents, crystal growth regulators, polishing chemicals, dispersants, electroless metallization, flotation of minerals, etching and active agents in AFFFs etc. [17, 45]. Aqueous film-forming foams (AFFFs), used as fire suppressants for hydrocarbon fuel fires, are one of the most critical products containing PFASs. AFFFs can effectively lower the surface tension and accelerate the formation of the aqueous film between oxygen and fuel to prevent further combustion. There are two principal processes of manufacturing PFASs or fluorinated surfactants, electrochemical fluorination and telomerization [44].

2.2 Risks and Regulations

Several decades after PFASs were invented, PFOS was detected in the environment in 2001. Giesy et al. (2001) reported that PFOS was detected in the tissues of wildlife occupying different trophic levels, which also provided evidence of PFAS bioaccumulation [6]. Since then, increasing types of PFASs have been detected in various environmental compartments such as rivers and oceans, drinking water, groundwater, surface soil, sediment, sewage sludge, wastewater treatment plants, house dust, indoor air, landfills, etc. [1-5, 70, 71]. PFASs are also detectable in humans. In 2006, So M.K. et al. reported the detection of PFASs in human breast milk in China [7]. According to the 2017 data from the National Health and Nutrition Examination Survey (NHANES), PFASs have been found in more than 98% of the serum samples in the U.S. [8]. Due to the stability of C-F bonds, PFASs are highly persistent in the environment and human beings. The half-life of PFOS and PFOA in humans are in the range of 2.3 to 5.4 years [72].

The ubiquitous existence in humans and the difficulties of elimination have caused great concerns on the health risks of PFASs. Numerous investigations have been conducted. Apelberg et al. (2007) identified the negative associations between PFOS/PFOA cord serum concentrations and baby weight/size at birth [16]. Granum et al. (2013) indicated that prenatal exposure to PFAS may be related to lower vaccine antibody levels and immunosuppression health outcomes in early childhood [12]. Barry et al. (2013) reported that PFOA exposure was relevant to kidney and testicular cancer [11]. Fisher et al. (2013) have observed a noteworthy association with PFHxS and high cholesterol [14]. In addition, hepatotoxicity, developmental toxicity, neurotoxicity, and endocrine disruption were observed to be linked with PFASs [72].

Considering the persistence, bioaccumulation, and potential health threats of PFASs, regulations and strategies have been developed to limit the use of long-chain PFASs. 3M Company, which was the largest PFOS producer, ceased the production of PFOS and its derivatives in 2002. DuPont, one of the major producers of fluorotelomer-based PFASs, phased out PFOA production by the end of 2015 [72]. Over the years, the raw materials of the industrial applications and commercial products have been shifted to short-chain PFASs (e.g. PFBA, PFBS) that are considered to be less bioaccumulative and probably less toxic. PFOS is listed as one of the new POPs under the *Stockholm Convention*, whereas PFOA is included in the chemicals recommended for listing and PFHxS has been under review by POPs Review Committee [73]. In 2016, The United States Environmental Protection Agency (US EPA) issued health advisory levels of PFOS and PFOA (combined) in drinking water at 70 ng/L [74]. Recently, US EPA implemented the PFAS Action Plan as a guide regarding PFAS issues as a result of the discovery of a high number of contaminated sites in the USA [75].

2.3 Contamination in AFFF-Impacted Sites

AFFFs were developed in the 1960s as a way to quickly respond to severe hydrocarbon fuel fires [46]. An early inventory report concluded that there were 4.6 million gallons of PFOS-based AFFFs in the US in 2004 [47]. With the introduction of stringent PFAS regulations, developing fluorine-free fire-fighting foams to replace PFAS-base AFFF has been seen as a critical step to move away from PFAS. However, AFFFs containing fluorinated surfactants are still dominant in the current global market. Repeated discharges of AFFFs in designated firefighting training areas over decades are found to be closely linked to severe soil and groundwater contamination by PFASs [1, 5, 10, 19, 20]. Considering the use of AFFFs, most of PFASs in AFFFs are directly discharged into soils; nonetheless, leaching from soils contributes to PFASs load in groundwater and surface water. The significantly elevated levels of PFASs have been observed in the soil, water, sediment and wildlife samples geographically relevant to the impacted sites [1, 10, 48], even when the regular discharges of AFFFs discontinued years ago [10]. According to Milley et al. (2018), 152 airports are likely to have PFAS contamination as a result of AFFF usage, and the surface waters are estimated to be impacted near 152 to 420 airport sites in Canada [49]. Not only airports, military bases, fire-fighting training areas, areas with emergency responses to the accidents [50], and AFFF storage sites are possible sites of contamination.

Fluorosurfactants in AFFFs can be in the form of anion, cation, zwitterion or non-ion [44]. PFOS and PFOA are the most widely detected and studied anionic PFASs, but significant presence of other novels PFASs that can contain functional groups such as quaternary ammonium, amine oxide, sulfonamide, and betaine needs to be addressed. Multiple studies have focused on the identification of PFAS composition in AFFFs. In 2012, Place and Field identified 10 classes of PFAS including the anionic, cationic and zwitterionic groups with perfluoroalkyl chain lengths from 4 to 12 by the combination of fast atom bombardment mass spectrometry (FAB-MS) and high-resolution quadrupole-time-of-flight mass spectrometry (QTOF-MS) [24]. Agostino and Mabury (2014) compared the results of total organofluorine content using combustion ion chromatography (CIC) with the results of specific PFAS analysis by LC-MS/MS, demonstrating the presence of unknown PFAS in environmental and biological samples [27]. In addition, they determined 103 compounds as 12 novel and 10 infrequently reported PFAS classes [27]. Backe et al. (2013) derived a new analytical method to quantitively evaluate 26 novel and 21 legacy PFAS in AFFF-contaminated groundwater samples by nonaqueous large-volume injection via LC-MS/MS [23]. Recently, Barzen-Hanson et al. (2017) discovered 40 novel PFAS compounds and another 17 new classes using liquid chromatography quadrupole time-of-flight mass spectrometry [20]. Gabriel et al. (2016) presented the existence of the unknown PFAS and developed a new analytical method to quantify the PFAS in sediments using the ultra-high performance liquid chromatography coupled to Orbitrap mass spectrometry through polarity-switching electrospray ionization [51].

2.4 Treatment of PFAS contaminated water

The dominant PFASs detected in the impacted aqueous phase are legacy anionic PFAS such as PFOS, PFOA, PFBS, PFBA, PFHxS, PFHxA, and 6:2 FTSA [3, 5, 52, 53]. Previous studies showed that conventional water treatment processes (i.e., coagulation, flocculation, sedimentation, and filtration) have little effect on PFAS removal (<20%) from water by comparing PFAS concentrations in influent and effluent of full-scale plants [21, 22, 54-56]. Besides, chlorination, ozonation, advanced oxidation, UV irradiation, and low-pressure membranes processes seem to be ineffective on PFAS reduction [21]. Effective technologies include adsorption by granular or powdered activated carbon (GAC, PAC), ion exchange, reverse osmosis, and high-pressure membranes [21, 22].

GAC adsorption can effectively remove the long-chain PFAS in drinking water treatment plants at full-scale [57]; however, a frequent reactivation or filter replacement is necessary to ensure the performance of GAC [21, 58]. Two to three times of replacement of GAC filter per year could maintain satisfactory performance on PFAS removal [58], though, the performance and the cost need to be balanced in the real-world scenario. However, GAC was not able to remove much short-chain PFAS especially PFBA and PFBS [57]. PAC was also studied to compare with GAC, but only at bench scale. Faster kinetics and higher removal efficiency were observed on PAC than GAC [59, 60].

Resin treatment is a promising technology to remove the PFAAs from water by anion exchange processes. The mechanisms are hypothesized to be the combination of electrostatic interactions and adsorption via hydrophobic interactions [21, 61]. Deng et al. (2010) examined six anion exchange resins with different polymer matrix, porosity, and functional groups on the performance of PFOS reduction from model wastewater. The results presented quick kinetic and significant PFOS uptake capacity (up to 4-5 mmol/g on IRA67 and IRA58) [61]. Woodard et al. (2017) have compared the ion exchange resin and GAC on PFOS and PFOA removal in the pilot scale. Both resin and GAC could achieve US EPA health advisories level that is 70 ppt of PFOS and PFOA combined, while resin has great advantages over GAC on the volume treated (over eight times) before exceeding the standards [62]. Furthermore, a significant improvement in shorter chain PFAS removal has been reported for ion exchange resins compared to GAC [21].

The effectiveness of reverse osmosis (RO) on PFAS reduction has been reported [21, 22, 63, 64]. RO can achieve high rejection of most PFASs including shorter chain PFAS such as PFBA [22]. However, the large volume of rejected solution (up to 20% of the feed) containing PFAS needs to be further treated. US EPA suggests that RO might be suitable for treating water at the point-of-use because of the low volume of water requiring treatment.

2.5 Treatment of PFAS-contaminated soil

Although a broad array of water treatment technologies has been studied, few practical soil remediation technologies are available. The major PFAS classes in AFFF-impacted groundwater and surface water systems are anionic groups, while non-anionic PFAS are negligible. Different from the PFAS composition in water, the cationic, neutral and zwitterionic PFAS can also make up a significant fraction of total PFAS load in impacted soils [51]. Since the environmental fate and behaviors of non-anionic PFAS are not fully studied and understood, this could be one of the significant challenges regarding soil remediation.

The technologies and mechanisms applied in water treatment could provide some clues in developing the soil remediation methods. Similar to adsorption in water treatment, sorbents can also be used in soil remediation as amended materials to stabilize the contaminants in the solid phase, reducing the bioavailability and mobility of some organic pollutants. Soil amendment has been proven to be effective for several organic contaminants. Tomaszewski et al. (2007) evaluated the performance of activated carbon amended sediment collected from Lauritzen Channel on DDT reduction after one month of treatment by measuring the aqueous equilibrium concentrations and uptake in semipermeable membrane devices (SPMDs) [65]. The results showed that the DDT equilibrium concentration in the aqueous phase was decreased by up to 83% and SPMD uptake declined up to 91% [65]. Reduction in PCB and PAH leachability and bioavailability has been reported in both laboratory scale and field studies [66-68]. However, limited soil amendment studies have been done targeting PFAS, especially non-anionic PFAS. Das et al. (2013) conducted soil treatability studies by mixing 10 wt% MatCARE^{TM} (modified palygorskite) with PFOS contaminated soils, maintaining 60% of the maximum water holding capacity at 25 °C and 37°C for a year. They reported effective immobilization with only 0.5 – 0.6% of leaching in the aqueous phase [32]. Hale et al. (2017) evaluated the performance of GAC, compost soil, and montmorillonite as amended materials in AFFF-impacted soils collecting from a Norwegian airport [33]. The single-step batch tests showed the effectiveness order on leaching PFAS reduction was AC (94% – 99%) > compost soil (29% – 34%) ≈ montmorillonite (28% – 40%) [33]. Sorengard et al. (2019) compared the performance of PAC, Rembind®, pulverized zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride as additives with dosage of 2% with soils spiked with 14 PFAS, demonstrating that the stabilization and solidification (S/S) efficiency was related to PFAS chain length, functional group, and sorbent type [34]. PAC or Rembind® was proven to be the best additive among all the amended materials [34]. Kupryianchyk et al. (2016) suggested activated carbon, but not biochar could be a useful additive to reduce the mobility of PFOS, PFOA, and PFHxS [35].

The literature has documented that modified clays, produced by inserting cation surfactants (e.g., quaternary ammonium surfactant) in the exchange sites of expandable clays, usually smectite groups, can enhance the immobilization of PFAS in soil remediation [40]. The hypothesized mechanisms included hydrophobic interaction and electrostatic interaction [40, 69]. The study of activated carbon is massive and mature, while there is an inadequate understanding of modified clays both in terms of applications and fundamental mechanisms. Smectite clays are of low cost and quite resistant to fouling by natural organic matter, which makes clays are attractive alternatives to GAC that is more expensive and prone to fouling. Hence, further studies are required in this area to bridge the knowledge gap.

3. Materials and Methods

3.1 Chemicals and Standards

Forty-nine of PFASs (see structures in Figure 1) were quantitatively analyzed, including anionic, cationic, zwitterionic and neutral compounds that have been detected in AFFF formulations or AFFF-impacted sites. Most of the analytical standards were obtained from Wellington Laboratories (Whitby, ON, Canada), including twelve perfluoroalkyl carboxylic acids (PFCAs, number of perfluoroalkyl carbons (n) = 3 - 14), eight perfluoroalkyl sulfonic acids (PFSAs, n = 3 - 10), four fluorotelomer sulfonic acids (FTSAs, n = 4, 6, 8, and 10), three fluorotelomer unsaturated acids (FTUAs ,n= 6, 8, and 10), perfluorobutane sulfonamide (FBSA), three perfluoroalkyl sulfonamides (FOSAs), including perfluorooctane sulfonamide (FOSA), methyl-perfluorooctane sulfonamide (MeFOSA), ethyl-perfluorooctane sulfonamide (EtFOSA), three perfluorooctane sulfonamideacetic acids (FOSAAs), including perfluorooctane sulfonamideacetic acid (FOSAA), methyl-perfluorooctane sulfonamidoacetic acid (MeFOSAA), ethyl-perfluorooctane sulfonamidoacetic acid (EtFOSAA), perfluoro-4-ethylcyclohexanesulfonate (PFECHS), 6:2 fluorotelomer sulfonamide betaine (6:2 FTAB), perfluorohexanoic sulfonamidoalkyl amine (PFHxSAm) and perfluorohexanoic sulfonamidoalkyl ammonium salt (PFHxSAmS). Standards of x:3 fluorotelomer carboxylic acids (4:3 FTCA, 5:3 FTCA, 7:3 FTCA) were donated by DuPont USA (Wilmington, DE, U.S.A.). Perfluorohexane sulfonamide (FHxSA) and 3:3 FTCA were purchased from Synquest Laboratories (Alachua, FL, U.S.A.). Perfluorooctane sulfonamidoalkyl amine (PFOSAm), perfluorooctane sulfonamidoalkyl ammonium salt (PFOSAmS), perfluorooctaneamidoalkyl betaine (PFOAB), perfluorooctanesulfonamidoalkyl betaine (PFOSB), perfluorooctane alkylamido amine oxide (PFOANO), perfluorooctane sulfonamidoalkyl amine oxide (PFOSNO), perfluorooctaneamide ammonium salt (PFOAAmS) were custom-synthesized at the Beijing Surfactant Institute (Beijing, China). Isotope-labeled internal standards (IS), including MPFBA, M5PFPeA, M5PFHxA, M4PFHpA, M8PFOA, M9PFNA, M6PFDA, M7PFUdA, MPFDoA, M2PFTeDA, M3PFBS, M3PFHxS, M8PFOS, M6:2 FTUA, M8:2 FTUA, M6:2 FTSA, M8:2 FTSA, d5-EtFOSA, d-MeFOSA, d-MeFOSAA, and M8FOSA, were also purchased from Wellington

Laboratories (Whitby, ON, Canada). HPLC-Grade water, formic acid (Optima LC/MS grade), methanol and acetonitrile of LC/MS grade, ammonium acetate, 0.1N sulfuric acid, sodium chloride, 1N and 5N sodium hydroxide solutions, and 6N hydrochloric acid solution were obtained from Fisher Scientific (Ottawa, ON, Canada). Sodium azide, potassium persulphate and ENVI-Carb graphite cartridges (250mg/6mL) were from Sigma Aldrich (St. Louis, MO). Nitric acid was purchased from ACP Chemicals Inc. (Montreal, QC, Canada), and N_2 gas was obtained from Praxair (Mississauga, ON, Canada). The deionized water used in all the experiments was acquired from LabChem (Zelienople, PA).

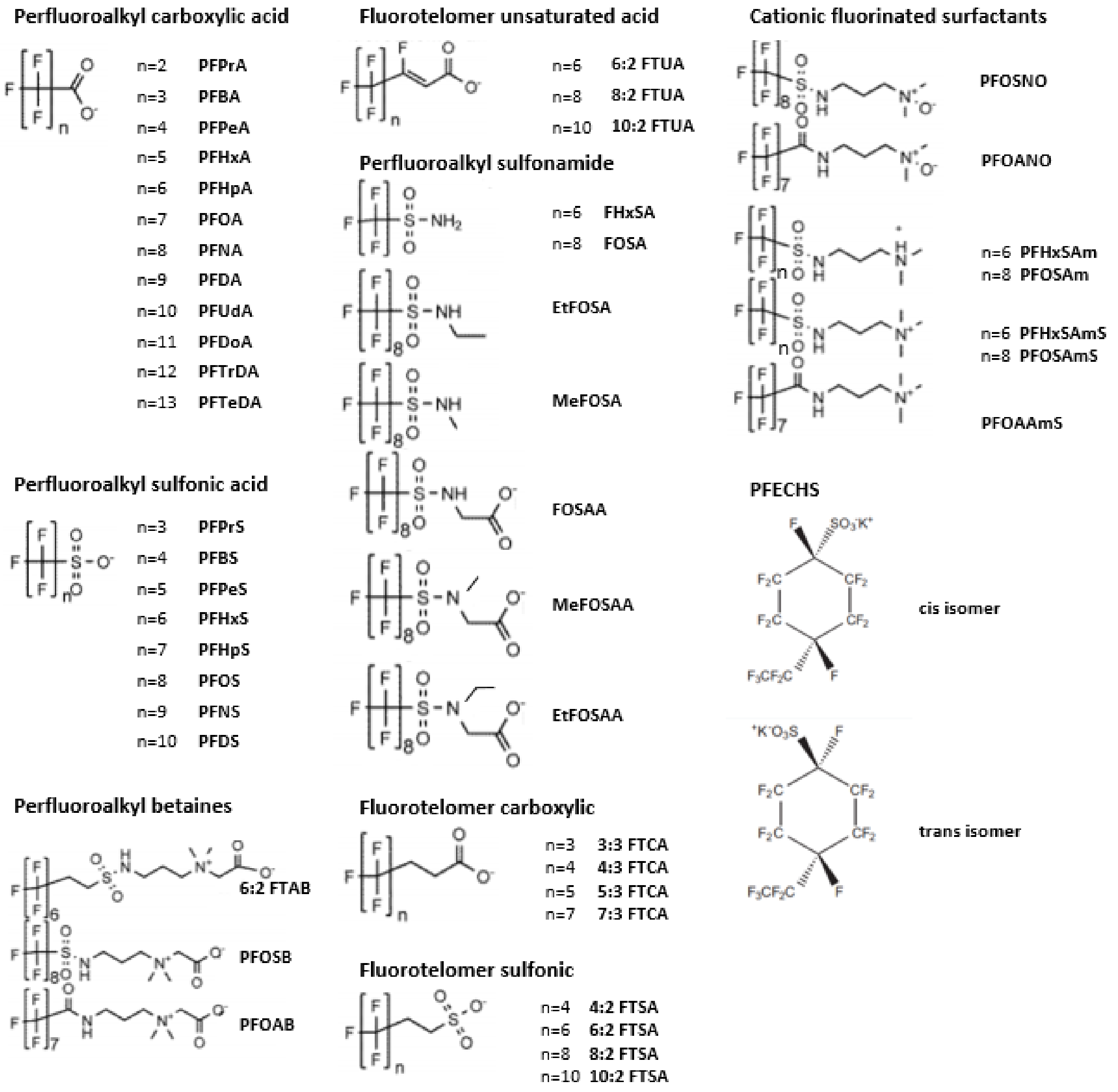


Figure 1 The structures of 49 quantitively evaluated PFASs

3.2 Soil Characterization

Four AFFF-contaminated soils with different sources and properties were used in the study. Two soils (CAN1, CAN2) were donated by Environment Canada and likely collected from two Canadian AFFF sites, and CETCO Minerals Technologies Inc. collected the other two soils (US1, US2) from one Airforce Base in the USA. Soil characterization (**Table 1.** Properties of the four soils collected from the US and Canada that were used in **Table 1**) was performed by A&L Canada Laboratories Inc. using standard methods. Details on the soil physical-chemical properties are provided in SI. Collected soils were air-dried for 48h and the fraction that passed through 2-mm sieve was retained for the experiments. The moisture content of air-dried soils was measured by weighing the soil samples in triplicate before and after 24-h oven drying at 105 °C. The results show that the air-dried soils have negligible water content (**Table S2**). The results throughout the thesis are expressed as per gram of oven-dry soil weight where it is applicable.

Table 1. Properties of the four soils collected from the US and Canada that were used in the study

ID	Textural Class	Sand %	Silt %	Clay %	[a] Organic Matter %	[b] CEC (meq/100g)	[c] pH
CAN1	Loamy Sand	89.2	0.8	10.0	0.4	32.6	8.0
CAN2	Loamy Sand	83.2	4.8	12.0	0.6	6.8	6.0
US1	Sand	91.2	0.8	8.0	0.3	7.3	7.9
US2	Sand	93.2	0.8	6.0	0.4	7.4	7.9

[a] Organic matter content was analyzed by weight loss on ignition at 360 °C using an analytical balance.

[b] CEC was calculated as the sum of Ca^{2+}, Mg^{2+}, and Na^{+} concentrations from a Mehlich 3 extract

[c] pH was determined by measuring 1:1 deionized water soil extract using pH electrode and mV meter

3.3 Sorbent Characterization

The modified clay FLUORO-SORB 100® (FS100), which is a bentonite-based material modified with quaternary ammonium surfactants, was obtained from CETCO Minerals Technologies Inc. The particle size distribution of FS100 was determined using laser diffraction particle size analyzer LA-950 (HPRIBA Ltd., Japan). X-ray diffraction (XRD) was conducted to demonstrate the structural difference between the modified clay and non-modified bentonite clay. The detailed information about sorbent characterization is provided in SI. The capability of reducing PFAS leachability was also compared with a commercial granular activated carbon Filtrasorb 400 (CalgonCarbon, USA) and hardwood-based biochar purchased from Charcoal House (USA) with a catalogue number of SKU-A 279. Both the GAC and biochar were previously characterized, and the details can be found in two recent publications by Zhi and Liu [76, 77].

3.4 Soil leaching test

The soils were first subjected to solvent extraction using methanol - ammonium acetate extraction fluid to reveal the full extent of PFAS contamination [78]. The extraction method as detailed by Munoz et al. (2018) [78] was recently developed to enhance the recovery of cationic and zwitterionic PFAS from soils, whereas the ASTM standard method (Method D7968-17a) using methanol-ammonia hydroxide was found to be ineffective for the recovery of non-anionic PFAS retained by soils [79, 80].

The soils were subjected to a leachability test where the soil-retained PFASs are mobilized under a simulated natural weathering condition. As no standardized method has been developed to assess the leachability of PFAS, the US EPA method 1311 Toxicity Characteristic Leaching Procedure (TCLP) was modified for the present study. The main modifications were focused on minimizing the sorption to vessel walls. Glassware was replaced with polypropylene or high-density polyethylene vessels, and all filtration steps were eliminated and substituted by high-speed centrifugations. Briefly, five grams of air-dried sieved soil was weighed into each 60 mL HDPE bottle, and 50 mL of deionized water was added to reach the water-to-solid ratio of 10. The vessels were shaken on a horizontal shaker for 8 days, at 150 rpm and 20 °C in the dark. At each sampling time, a subsample was pipetted out and centrifuged at 20,000 x *g* for 10 minutes, and the supernatant was taken out for further dilution with methanol and water (Methanol:Water 80:20). Diluted samples were stored at -20 °C and spiked with the internal standards before the chemical analysis using the high-resolution mass spectrometry as described below.

Sorption losses of PFAS caused by containers needed to be considered. Recoveries of eight representative PFASs, including anionic, zwitterionic and cationic groups from aqueous solutions, were examined in 60-mL HDPE bottles in triplicates using three groups (i.e. anion, zwitterion, and cation) of PFAS spiked solutions respectively. Anionic PFAS included PFOS, PFOA, PFDA, 6:2 FTSA, 8:2 FTSA, zwitterionic PFAS included PFOAB and PFOSB, and PFOAAmS was the representative cationic PFAS.

HDPE bottles showed little adsorption of legacy anionic PFAS (less than 5%) but some adsorption of newly-identified zwitterionic and cationic PFAS (20%-45%). Given that the performance evaluation of amended materials was determined based on the PFAS concentration decrease in soil leachate before and after soil amendment divided by PFAS leaching concentration before the addition of soil amendment, the adsorption on the containers does not affect the assessment of the sorbent performance. However, some zwitterionic and cationic PFAS concentrations in the soil leachate could be underestimated because of the adsorption to container walls. The detailed results of the recovery test are provided in **Figure S3** in SI.

The pH effect of the leaching solution on PFAS leachability was also evaluated based on US EPA method 1312 (Synthetic Precipitation Leaching Procedure) and method 1311 TCLP (Toxicity Characterization Leaching Procedure). A leaching test, without soil amendment but with different extraction fluids, was conducted on Soil CAN1. Deionized water was set to be the control group of four extraction fluids with pH ranging from 4.0 to 8.0. Diluted 60/40 weight percent mixture of sulfuric acid/nitric acid and 0.01 N sodium hydroxide were used to adjust the pH of extraction fluid according to **Table S3**. Two grams of sieved CAN1 soil was added in each bottle, then the 40 mL of Extraction Fluid was added, and Parafilm was used outside the screw caps for a tight seal. The bottles were secured onto a rotary agitation device and rotated at 30 ± 2 rpm at 25 °C for 3 days. The pH of the leaching solution was not found to have any statistically significant impact of PFAS leachability as discussed in Section 4.3, and therefore later tests were performed using deionized water without pH adjustment.

3.5 Comparison of sorbents

A heavily contaminated soil (CAN1) was first tested to evaluate the effect of FS100 dosage, equilibration time, leaching kinetics, and potential microbial activities on PFAS leaching. A range of FS100 dosages from 0.1% to 5% (weight percentage of soil mass) was assessed using the modified soil leachability test, where the addition of soil was followed by the addition of FS100, and then the leaching solution (deionized water) to reach the water-to-solid ratio of 10. Sodium azide was added to prevent the potential microbial activities, meanwhile, a group of control without sodium azide was set for comparison. A similar sampling procedure as what was developed above was performed. Then a comparative assessment was conducted at the dosage of 0.5 wt% for all the sorbents (modified clay, GAC, and biochar) and for all four soils.

We also tested an approach to improve the performance of modified clays on cationic PFAS immobilization by comparing the single FS100 soil amendment system with the FS100/natural bentonite combined system. The same methodology was used as previous tests.

3.6 Instrumental Analysis

Quantitative analysis was performed by ultra-high-performance liquid chromatography coupled to a high-resolution accurate-mass Orbitrap mass spectrometry (UHPLC-HRMS). The separation was achieved by a Thermo Hypersil Gold aQ column (100 mm × 2.1 mm, 1.9 µm particle size). The Q-Exactive Orbitrap mass spectrometer (Thermo Fisher Scientific, Waltham, MA, USA) was operated in Full Scan MS mode (mass scan range: 150-1000 *m/z*) with a resolution setting of 70,000 FWHM at 200 *m/z*. The instrumental analysis method applied in this case was developed from the previous studies [51, 78, 81, 82]. More details on the instrumental analysis method are provided in **Table S4** in the SI.

3.7 Data Analysis

The capability of amended material to reduce the PFAS leachability (i.e. to stabilize the PFAS in soils) was directly interpreted by the PFAS aqueous concentration decrease in the leachate which can be calculated as follows:

$$Concentration\ Decrease_{PFAS}\ (\%) = 100\%\ (C_{NAm,PFAS} - C_{Am,PFAS})/C_{NAm,PFAS}$$

$$Concentration\ Decrease_{TOTAL\ PFAS}\ (\%) = 100\%\ (\sum C_{NAm,PFAS} - \sum C_{Am,PFAS})/\sum C_{NAm,PFAS}$$

where $C_{NAm,PFAS}$ is the equilibrium concentration of a leached PFAS in the soil leachate when there was no amended material added, and $C_{Am,PFAS}$ is the equilibrium concentration of the same PFAS in the soil leachate in the presence of amended sorbent material.

Additionally, the leaching PFAS fraction of the total PFAS retained by soil was determined by the following formula:

$$F_{Leaching\ PFAS} = C_{Leaching\ PFAS}\ (\mathrm{ng/g})\ /\ C_{PFAS\ retained\ by\ soil}(ng/g)$$

where $C_{PFAS\ retained\ by\ soil}$ was determined by performing soil solvent extraction by ammonium acetate.

4. Results and Discussion

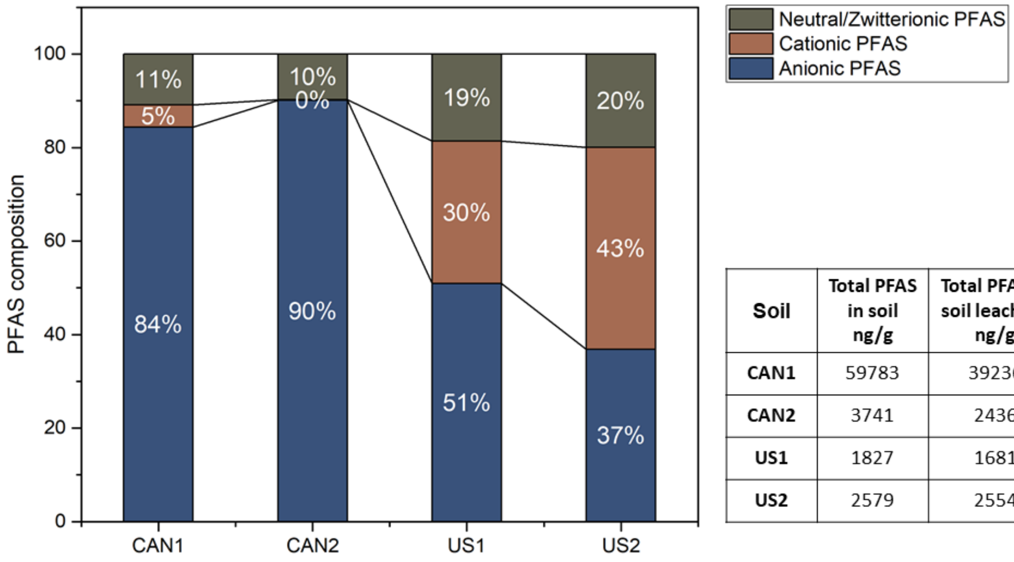
4.1 PFASs retained by soils

PFASs retained by soils were determined by conducting multi-cycles of methanolic ammonium acetate soil extraction [78]. Among forty-nine quantitatively monitored PFAS, forty PFASs were detected in soil CAN1, thirty-seven were observed in CAN2, thirty-two PFASs were identified in US1, and twenty-five PFASs were found in US2. The concentration of total PFASs retained in four soils ranged from 1,827 to 59,783 ng/g. PFOS, an anionic PFSA, was the most dominant PFAS detected in all the four soils with the concentration ranging from 868 to 44,441 ng/g and accounting for 33.7% ~ 74.3% of total PFASs. Other abundant anionic PFASs observed in all four AFFF-contaminated soils included 8:2 FTS (18 –1,109 ng/g), 6:2 FTS (4.4 - 731 ng/g), PFHxS (12 - 483 ng/g), PFHxA (4.9 - 148 ng/g), and PFOA (14 - 108 ng/g). Some neutral, zwitterionic, and cationic PFASs were also detected at relatively high concentration, such as PFHxSAm (17 - 4,111 ng/g), PFHxSAmS (109 - 3,098 ng/g), FHxSA (91 - 545 ng/g), FOSA (81 - 366 ng/g), PFOSAmS (17 - 206 ng/g), and PFOSAm (1.2 - 145 ng/g). The detection of other PFASs aside from anionic PFAS was expected, given that several studies have reported that non-anionic PFASs make up a large fraction of fluorosurfactants in AFFF formulations [51]. The data once again demonstrated that analyzing the soil leaching behaviors of both anionic and non-anionic PFAS groups is critical; otherwise, a significant fraction of PFASs would be missed out, jeopardizing comprehensive risk assessment or developing effective remediation strategies and technologies. Details of PFAS profiles for four AFFF-contaminated soils are provided in **Table S5** in the SI.

4.2 PFAS leaching profiles

Overall, the types of PFAS detected in soil leachate were fewer than those in soil solvent extracts. Thirty-one PFASs were observed in CAN1 leachate, twenty-two PFASs were determined in CAN2 leachate, twenty-three PFASs were identified in the US1 leachate, and twenty-two PFASs were found in US2 leachate. The PFAS compositions of four contaminated soil leachates were soil dependent as shown in **Figure 2**, which demonstrated that although the anionic group is dominant, the presence of cationic and neutral PFASs in soil leachate is significant. Considering approximate 20-45% of adsorption on containers, the concentrations of zwitterionic/neutral and cationic groups were, in fact, underestimated in the leachate. The PFAS leaching profiles and contribution of each individual PFAS to the total PFASs of four AFFF-contaminated soils were illustrated in **Figure 3**. The abundant PFASs detected in the soil leachates included anionic group (e.g. PFOS, PFHxS, and 6:2 FTS), cationic group (e.g. PFHxSAmS, PFOSAmS, and PFOANO), and neutral/zwitterionic group (e.g. FHxSA, FOSA, PFHxSAm and PFOSAm). Their general abundance roughly reflected their dominance in soils, as demonstrated by the concentrations in the methanolic ammonium acetate extracts (Section 4.1). The concentration of each individual PFAS found in each soil leachate is tabulated in **Table S6** of SI. The fraction of total leachable PFAS relative to total solvent extractable for the two soil samples collected in Canadian sites was around 65%, and that fraction for the two US soils was more than 90%. The difference in leachability may suggest that soil textural class may play a role, and the sandy soils have the tendency to leach out most of the PFASs (65-99%) they retain. Whether such a trend can be generalized to other soils needs caution as the sorption and desorption of non-anionic PFAS by soils have not been adequately evaluated.

The pH effect of extraction fluid on PFAS leaching was evaluated using Soil CAN1. **Table S19** demonstrated the individual and total PFAS concentration in the CAN1 leachate at pH ranging from 4.0 to 8.0. No significant pH effect was observed on either the total PFAS or individual PFAS. The addition of the acid mixture or base to the extraction fluid created a range of ionic strength. Though not directly evaluated, ionic strength at the low to moderate level also did not appear to have an impact on PFAS leaching. Previously, the sorption of several anionic PFASs (e.g., PFOS, PFOA) onto sediments exhibited a pH-dependent trend with weaker sorption observed at higher solution pH, attributed to the increasing electrostatic repulsion between the negative surface charge of sediment and the PFAS anions. However, since only one soil was evaluated in this study, further assessment of the effect of pH on PFAS desorption or mobilization is needed in the future as part of the efforts to develop standardized leachability test methods.



(a)

Soil	Total PFAS in soil ng/g	Total PFAS in soil leachate ng/g	$F_{LEACHING\ PFAS}$ %
CAN1	59783	39236	65.6%
CAN2	3741	2436	65.2%
US1	1827	1681	92.0%
US2	2579	2554	98.9%

(b)

Figure 2. (a) The cationic, anionic, neutral, and zwitterionic PFAS composition relative to the total PFASs detected in the soil leachate of four contaminated Soils; **(b)** Total PFAS concentrations in soil (solvent extractable), total leachable PFAS concentration, and fractions of leachable PFAS relative to total solvent extractable PFAS.

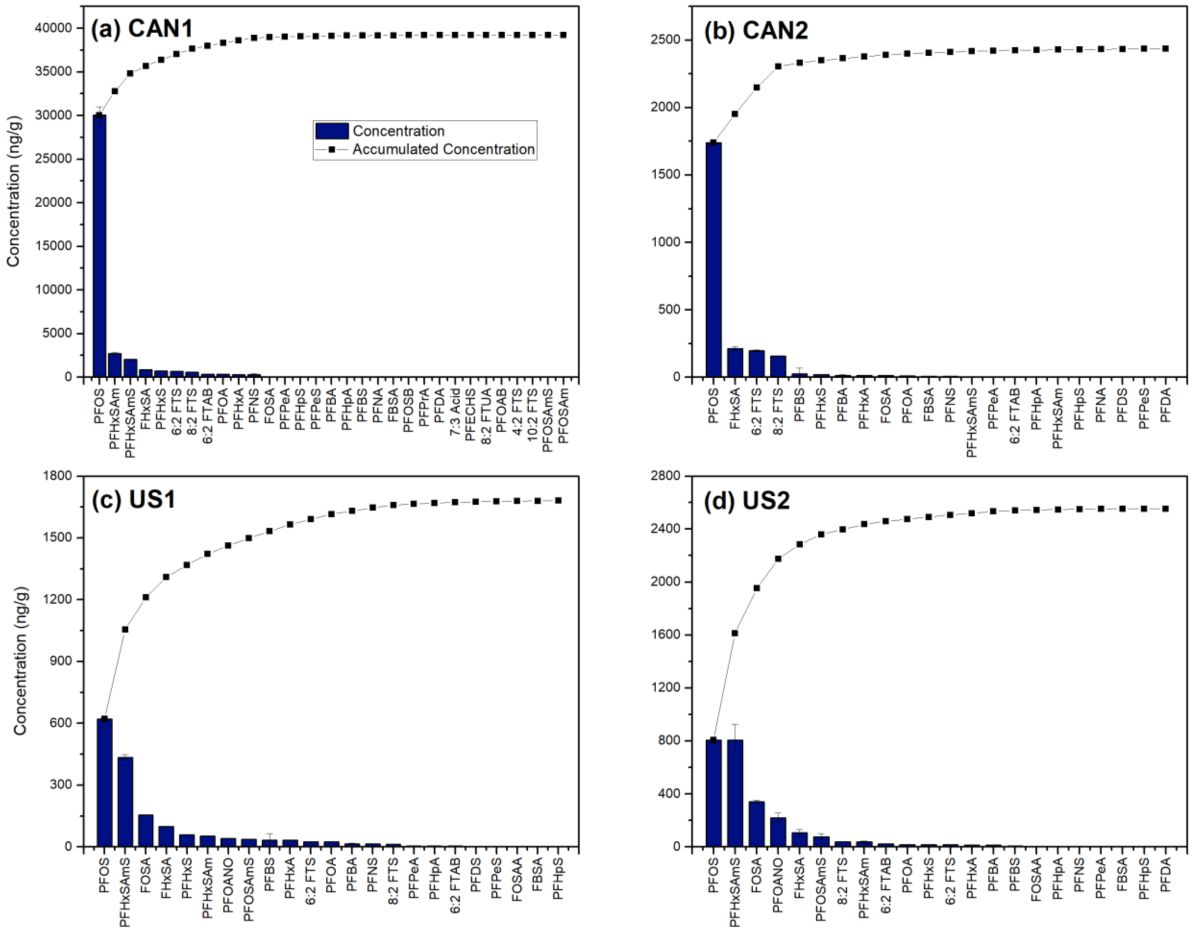
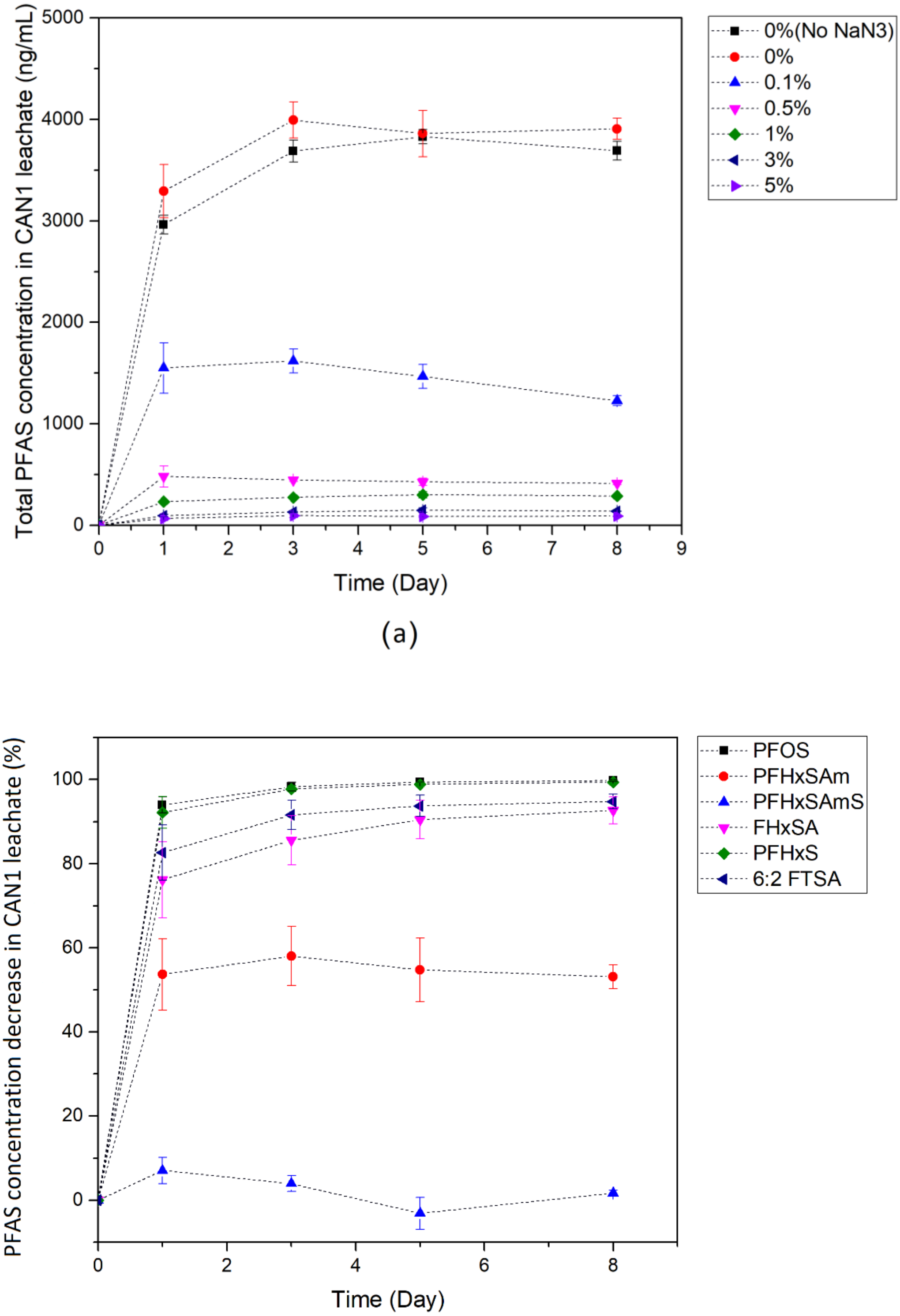


Figure 3. The PFAS profile in the leachate of each soil and contribution of each PFAS to the total PFAS concentrations. A total of 49 PFAS analytes were monitored, and the ones below detection limits are not shown.

4.3 Effect of FS100 dosage on reducing PFAS leachability

The decrease in leachability of total and individual PFAS at different FS100 dosages was examined on the most heavily contaminated soil CAN1. The repetitive sampling of CAN1 leachate during the eight days showed relatively rapid equilibration in the solid/water system. For the soil without sorbent amendment, the equilibrium can be reached at around day 3 or day 4, with a significant increase of PFAS concentration in aqueous phase observed in the first 24 hours (approximate 80% of the equilibrium concentration). As for the soil with the FS100 amendment, equilibriums can be reached within 24 hours, although some variability among different PFAS compounds was observed. The leaching kinetics of total PFAS in CAN1 was illustrated in **Figure 4 (a)**. Control without the addition of sodium azide was set to evaluate the effect of microbial activity during the batch test. No significant difference in PFAS analytes concentration was observed between the samples (no amendment, with sodium azide) and the control groups (no amendment, no sodium azide), which suggests the effect of degradation or microbial activity was negligible in this case. Previously, it was found the structural analogue of PFHxSAmS with eight perfluorinated carbons (e.g., PFOSAmS) can undergo aerobic transformation to form PFOS, but with a half-time of more than 500 days [26]. Similarly, FOSA can also undergo biotransformation but also at a very slow rate [83]. Given the relatively short duration of the leachability test, it is not unexpected that the effect of microbial activity can be ignored. We suspect that the soils used in the study are quite weathered, and labile functional groups probably had already been transformed.

At equilibrium, about 70% of the decrease in total PFAS aqueous concentration in CAN1 leachate can be achieved with 0.1 wt% of FS100, while 0.5 wt% of FS100 can result in 90% of total PFAS removal in the aqueous phase . Dosages of 1 wt%, 3 wt%, and 5 wt% of FS100 can achieve 92%, 96%, and 98%, respectively, of total PFAS removal in the soil leachates. **Figure 4 (b)** demonstrated the time course of the concentration decrease (%) in CAN1 leachate of the six dominant PFAS (PFOS, PFHxS, 6:2 FTSA, FHxSA, PFHxSAm, and PFHxSAmS) in the presence of 0.5% of FS100. For the six compounds, the equilibrium can be reached within 1-4 days. A dosage of 0.5 wt% of FS100 can achieve more than 90% of removal for anionic PFOS, PFHxS, and 6:2 FTSA. As for neutral/zwitterionic compounds such as FHxSA and PFHxSAm, 0.5 wt% of FS100 can around between 55% to 90% concentration decrease in the soil leachate. For cationic PFAS (PFHxSAmS), the little concentration decrease in the aqueous phase was observed.

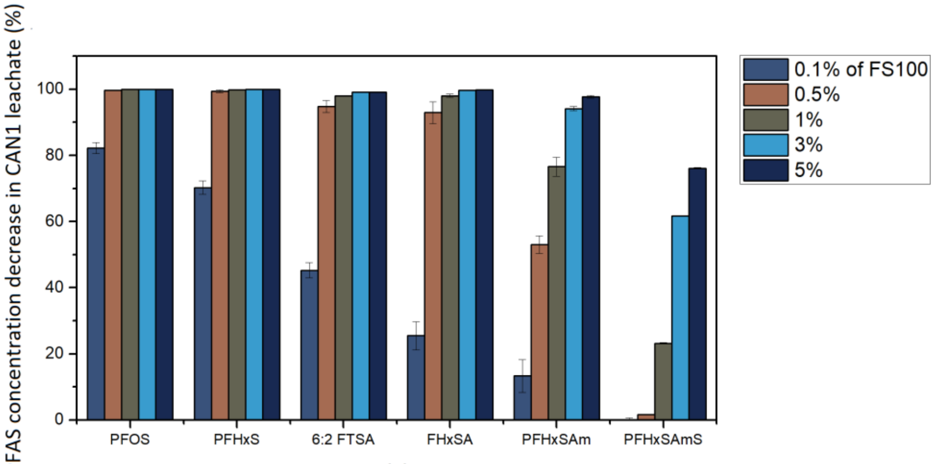


(a)

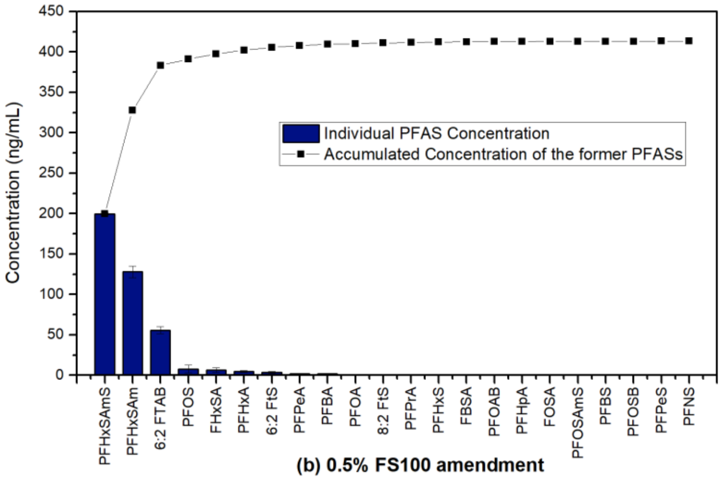
(b)

Figure 4. (a) The leaching kinetics of total PFAS from Soil CAN1 with the addition of 0.1 – 5 wt% of FS100; **(b)** the leaching kinetics of the six dominant PFAS (PFOS, PFHxS, 6:2 FTSA, FHxSA, PFHxSAm, and PFHxSAmS) from Soil CAN1 with the addition of 0.5% of FS100.

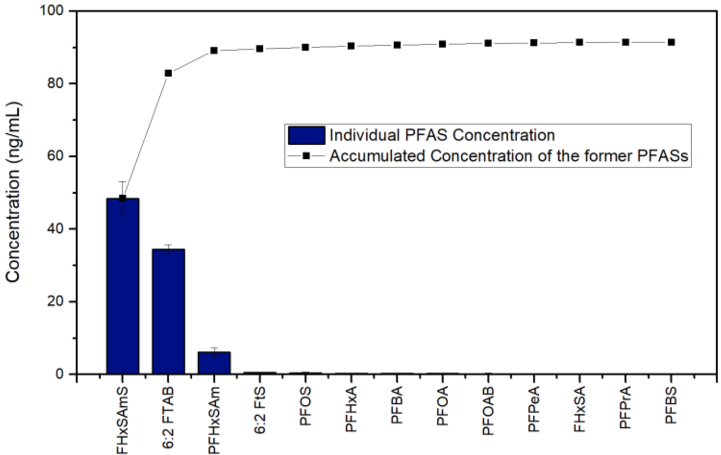
The effect of sorbent dosage is illustrated in **Figure 5**. **Figure 5 (a)** shows the removal efficiency of 6 dominant PFAS in CAN1 leachate at equilibrium with 0.1 wt% - 5 wt% of the FS100 amendment. For anionic PFAS and the neutral compound FHxSA, 0.5 wt% is a critical point. For PFOS and PFHxS, when the dose was 0.5% or higher, the concentration decrease in soil leachate can reach almost 99.9%. For 6:2 FTSA and FHxSA, 0.5 wt% can achieve about 90% of them stabilized in soil. However, for neutral PFHxSAm and cationic PFHxSAmS, the increasing trend in immobilization as the amount of the sorbent increases appeared to be moderate. Therefore, the order by which FS100 exhibits removal efficiency can be estimated as: anionic PFAS> neutral/zwitterionic PFAS> cationic PFAS. **Figure 5 (b)** and **(c)** show the PFAS leaching profile from Soil CAN1 in the presence of 0.5 wt% and 5 wt% FS100, respectively, and contribution of each PFAS to the total PFAS concentrations. Comparing to the PFAS profile without the addition of the amendment (**Figure 3a**), it is clear that both the type of PFAS detected and the total PFAS concentration decreased substantially. Before the addition of FS100, thirty-one types of PFAS were detected, and the total PFAS concentration in soil leachate was as high as 3923.6 ng/mL. After amended with 0.5 wt% of FS100, twenty-two of individual PFASs were identified in the leachate, and the total PFAS concentration declined to 413 ng/mL. If the dose was determined to be 5 wt%, only thirteen types of PFAS were found in the amended CAN1 leachate, and the total PFAS concentration was significantly dropped to 91.4 ng/mL. Detailed data of leaching kinetics, the effect of sorbent dosage, and potential microbial activity effect on each major individual PFAS in CAN1 soil leachate were listed in SI.



(a)



(b) 0.5% FS100 amendment



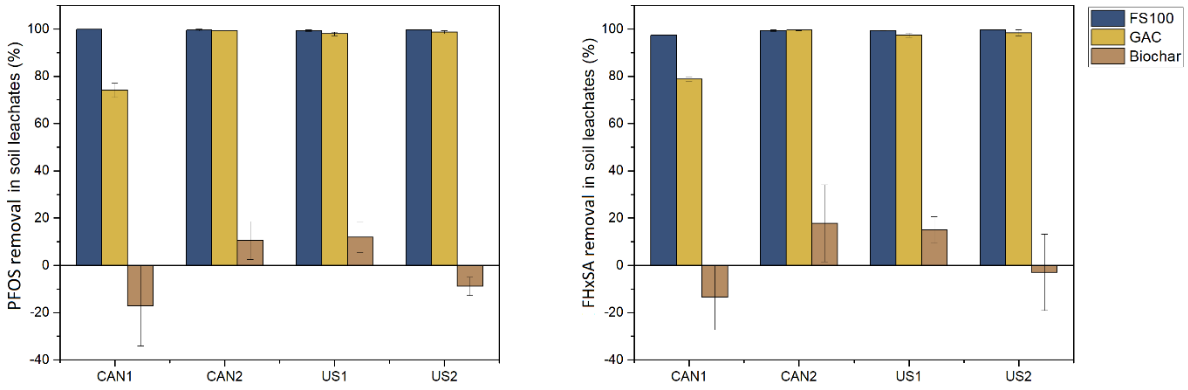
(c) 5% FS100 amendment

Figure 5. (a) The decrease in leachability of six dominant PFAS in from Soil CAN1 with the addition of 0.1 wt% - 5 wt% of FS100 when measured after 8-day equilibration; the PFAS that leached out from Soil CAN1 with the addition of **(b)** 0.5 wt% and **(c)** 5.0% wt% of FS100 amendment.

4.5 Comparative assessment among FS100, GAC and Biochar

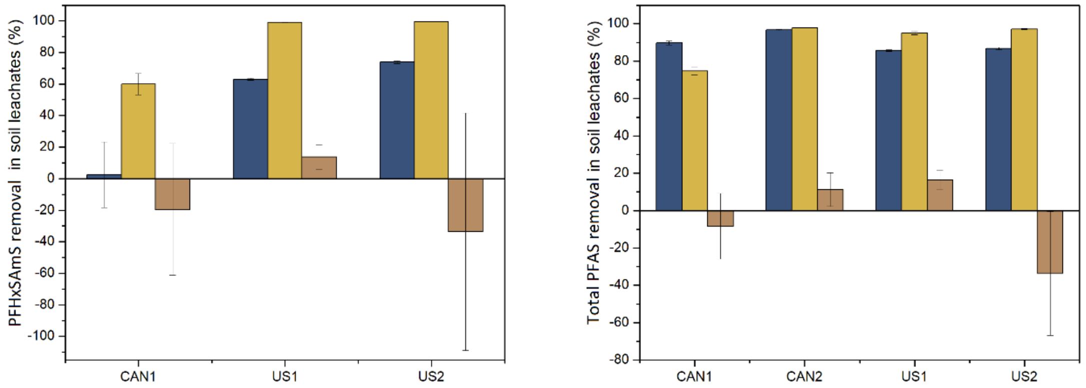
A comparative assessment of the performance of FS100, GAC and biochar at 0.5 wt% on reducing the mobilization of PFAS was provided, using the four contaminated soils. The performance of amended sorbents was closely related to the PFAS type and PFAS concentration. As shown in **Figure 6**, the extent of PFOS immobilization in four soils followed the order of FS100 > GAC >> Biochar. FS100 can achieve more than 99% of PFOS removal in all four soil leachates whose initial PFOS concentrations ranged from 62.1 to 3005 ng/mL. Meanwhile, GAC can also achieve approximate 99% of PFOS decrease in the aqueous phase when the starting PFOS concentration was 62.1 to 174 ng/mL in Soil CAN 2 and the two US soils. For Soil CAN1 leachate, the initial PFOS concentration was as high as 3005 ng/mL and GAC can only remove about 74.1% PFOS. In addition, biochar showed almost no removal of PFOS in the soil slurry; the PFOS decreased in the leachate ranged from -37.7% to 11.9%. The negative value may be a result of PFOS generation from precursor substances, compounded by measurement variability of LC-MS/MS analysis, which routinely experiences ±20% variation. Previously, Kupryianchyk et al. (2016) found that there was small to no PFOS decrease in pore water concentration when two biochars from different feedstocks (i.e. mixed wood and paper mill waste) were amended to PFAS-contaminated soil [35], which was consistent with our findings. Hale et al. (2017) also observed the increase of certain PFAS (e.g. PFBS, PFHxS and PFNA) after the soil amendment addition [33]. Further studies are needed to understand the mechanisms of such increases, which we hypothesize due to the transformation of polyfluoroalkyl substances (or precursors). It is probably a case-by-case scenario regarding the possible generation of PFAAs from the precursors, depending on the types of AFFFs applied to the soil and the length and condition of natural attenuation processes.

For neutral/zwitterionic PFAS, the observed removal efficiency was in the same order as that of anionic PFAS: FS100 > GAC >> Biochar. **Figure 6 (b)** demonstrated the decrease in leachability of FHxSA (a representative of neutral/zwitterionic PFAS) in the soil leachates with the addition of 0.5% of FS100, GAC, or biochar. FS100 can achieve around 99% of FHxSA removal in all soil leachates whose FHxSA concentrations ranged from 9.9 to 86.0 ng/mL. Meanwhile, GAC can achieve an approximate 99% of the decrease in the leachates when the FHxSA concentration ranged from 9.9 to 21.3 ng/mL. For CAN1 leachate with the initial FHxSA concentration of 86.0 ng/mL, GAC achieved 78.7% of removal. Similar to PFOS, biochar showed little decrease of FHxSA in leachates; for four contaminated soils, the FHxSA aqueous concentration decrease in the leachates ranged from -39.6% to 18.5%. In terms of cationic PFAS, the effectiveness order of three sorbents is: GAC > FS100 >> Biochar. **Figure 6 (c)** illustrated the decrease in leachability of PFHxSAmS (a representative of cationic PFAS) in 3 soil leachates (CAN1, US1, and US2) with the addition of 0.5% of FS100, GAC, or biochar. In this case, only three soil leachates were evaluated, as **Figure 2 (a)** showed that the cationic compound was negligible in Soil CAN2. When the initial PFHxSAmS concentration in soil leachate was in the range of 43.5 to 80.7 ng/mL, GAC can achieve 99% of PFHxSAmS removal while FS100 can achieve 63.0% to 73.8% removal in the leachates. When the PFHxSAmS concentration was relatively high (i.e., 203 ng/mL in CAN1 leachate), GAC can achieve 60.3% of removal while FS100 can only achieve 4.6% of removal. Clearly, when the PFHxSAmS concentration was at 203 ng/mL or higher, a dosage of higher than 0.5 wt% would be required for GAC or FS100 to achieve a significant decrease in leachability of the cationic PFAS. Biochar overall showed almost no removal of PFHxSAmS (from -44.4% to 13.8%). The experimental data for other individual PFAS in the presence of 0.5 wt% of FS100, GAC or biochar are provided in the SI.



(a) PFOS

(b) FHxSA



(c) PFHxSAmS

(d) Total PFASs

Figure 6. Comparison of three sorbents (Fluorosorb 100, granular activated carbon and wood biochar) in decreasing the leachability of (a) PFOS, (b) FHxSA, (c) PFHxSAmS and (d) total PFAS from four contaminated soils at the sorbent dosage of 0.5 wt% (n = 3).

A comprehensive evaluation of three sorbents in decreasing the leachability of total PFASs was conducted and demonstrated in **Figure 6 (d)**. Similar to what has been discussed above for several individual PFAS, the biochar overall had minimal capacity to immobilize any PFAS, and therefore, total PFAS removal efficiency of biochar in the soil leachates was very low, -28.91% ~ 16.41%. Therefore, the following discussion will only focus on FS100 and GAC. Soil CAN1 is the most heavily contaminated soil with 3924 ng/mL as the initial total PFAS concentration in soil leachate. **Figure 2 (a)** further shows that anionic, neutral and zwitterionic PFAS families accounted for 95% of total PFAS while cationic PFAS only 5%. The more effective removal of PFOS by FS100 than GAC is consistent with the observation, and the total PFAS removal from the soil leachate by FS100 (89.7%) is also higher than that of GAC (74.75%). For

Soil CAN2, the cationic PFAS composition is negligible and the initial concentration of total leachable PFAS (244 ng/mL) was much lower than that of CAN1. Both FS100 and GAC achieved similarly high removal (more than 96%) of total PFAS and also were higher than those of Soil CAN1. For the Soil US1 and US2, the initial leaching concentrations were at a similar level (255 ng/mL for US2) or lower (168 ng/mL for US1) than that of CAN2. Nevertheless, **Figure 2(a)** shows that cationic PFASs make up a significant fraction of total leachable PFAS, 30% and 43%, respectively from US1 and US2. Hence, GAC (95.0% of immobilization for US1 and 97.2% for US2) performed better than FS100 (85.7% of immobilization for US1 and 86.7% for US2). It appears that the type of PFAS leaching out soils (without the presence of any soil amendment), as well as initial leachate concentration, are two critical factors that affect the sorbent performance in stabilizing the AFFF contaminated soils.

4.6 Suggested improvement for removing cationic PFASs

Since the performance of FS100 for cationic PFAS immobilization was less ideal than that for anionic and neutral PFASs, for soils that contain high levels of cationic PFASs, other measures are necessary for reducing the total PFAS levels. According to a preliminary study (see **Table S24** and **Table S25** in SI), Na^+-bentonite clays can take up a significant amount of cationic PFOAAmS owning to cation exchange processes. With the addition of natural Na^+ saturated bentonite at 1/5000 g/mL of solid to liquid ratio, the aqueous concentration of PFOAAmS was significantly decreased from initial 2080 ppb to 33.5 ppb at equilibrium. It is hypothesized that the bentonite clay minerals with negative charges have a great affinity to the cationic PFOAAmS due to electrostatic force, and the expandable property of bentonite would create spaces for the retained PFOAAmS. Hence, a combined FS100/bentonite system (0.5 wt% FS100 + 0.5wt% bentonite) was tested for the performance of PFAS immobilization. The comparison between the FS100/Bentonite combined system and the single FS100 soil amendment system is demonstrated in **Figure 7** for every cationic PFAS found in the three soils which leached out cationic PFASs. The removal of PFHxSAmS went from 2.6% to 32.3% in CAN1 leachate, 63.0% to 79.3% in US1 leachate, and 73.8% to 84.6% in US2 leachate. Similar trends were observed for PFOSAmS and PFOANO, the cationic PFAS with relatively high concentrations in US1 and US2 (around 36ng/g ~ 220ng/g). The results show that the cation exchange process is involved in removing the PFASs from the soil-water slurry and the hydrophobic effect plays a less important role than cation exchange. However, the direct application of sodium bentonite clay into soils might create some complications. Being a less stable form of bentonite clay, the Na^+-bentonite may quickly lose its cation exchange sites when calcium ions in solution may quickly exchange with clay-bound Na^+. So, Na^+-bentonite may be only effective for a short period. The swelling property of the bentonite clay may pose some challenges in field applications if applied at a high dosage.

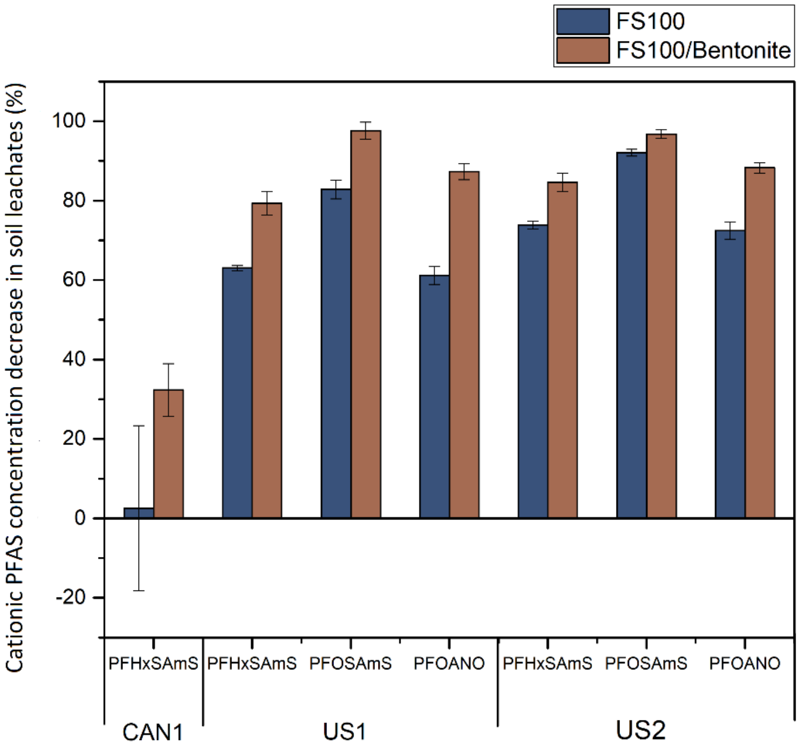


Figure 7. Comparison of FS100 alone (0.5 w%) and FS100/bentonite (0.5 wt% for each clay) in reducing the leachability of three cationic PFAS from three contaminated soils; Soil CAN2 was not selected for the test as the cationic PFAS composition was negligible (less than 1%) (n = 3).

5. Conclusions

The analysis of PFAS profiles of four AFFF-contaminated soils and their leachates demonstrated the significant presence of the anionic and non-anionic PFAS, while the latter has not been sufficiently discussed in the literature. A new clay-based material FLUORO-SORB 100® (FS100) was tested for its capability of reducing the leachability of anionic, neutral/zwitterionic and cationic PFAS retained by the impacted soils in a soil-water slurry system constructed based on a modified US EPA leachability test. A significant decrease in aqueous phase concentration of anionic PFAS (95~99%) can be achieved in 1-4 days with FS100 dosage as low as 0.5 wt%. Considerable removal of several non-anionic PFAS (70~99%) can be achieved with a higher dosage than 0.5 wt%. Comparative assessment between the clay-based sorbents and two commercial carbon-based sorbents showed that the order of effectiveness for anionic and neutral PFAS reduction is FS100> GAC>> Biochar, while for cationic PFAS immobilization the order is GAC> FS100>> Biochar. Besides the sorbent type, the performance of the amended materials is also soil dependent, related to soil properties and soil PFAS profiles (i.e., PFAS composition and concentration), suggesting that comprehensive characterization of soil properties and thorough identification and quantification of PFAS prior is essential to ensuring the right amendment was selected in the site remediation projects. Furthermore, the performance of modified clays on the cationic PFAS immobilization can be improved by adding natural bentonite clays, though potential limitations imposed by the property of natural clays have to be considered.

The findings contribute to developing low-cost and easy-to-implement soil remediation technology for PFAS-contaminated soils and water. For example, the amended materials can be used for in-situ PFAS immobilization by directly mixing with the contaminated soils or for groundwater remediation using permeable reactive barriers. This study is currently only limited to lab-scale testing, as well as all the other published studies on soil amendment for PFAS treatment [32-35]. The lab results warrant further testing in pilot-scale or actual field testing.

Though the study shows an effective and potentially low-cost method to immobilize the soils retained by soils, how the trapped PFASs can be completely destructed needs further studies to allow the spent clays to be safely disposed of. A few studies have shown that PFASs immobilized between clay layers can be chemically reduced by hydrated electrons that are generated by indole types of chemicals activated by UV light [84]. In addition, thermal treatment such as smoldering [85] or hydrothermal treatment [86] can be tested aside from the traditional incineration method.

6. Suggested Future Directions

The study has presented an effective and potentially cost-efficient remediation approach targeting the PFAS-impacted soils using modified clays in the laboratory scale; however, several issues need to be addressed in the future.

First of all, the mechanisms of PFAS adsorption by natural and modified clays need to be elucidated to further understand the adsorption process and provide a solid theoretical foundation of this soil remediation method by clay minerals. So far, the prevailing hypothesized mechanisms include the electrostatic force and hydrophobic effect; however, direct evidence is necessary to support this hypothesis.

Meanwhile, the feasibility and practicality of this method in real-world site remediation scenarios need to be comprehensively evaluated. Column study and full-scale study for long terms are suggested to simulate the field conditions. Long-term adsorption capacity and the factors that may influence the capacity (such as pH, co-contaminants, etc.) should be assessed. The desorption test is recommended to study the potential for the release of PFAS from the clays over time influenced by various environmental factors. Furthermore, detailed operational issues also need to be solved, such as how the amendment materials can be homogeneously mixed with soil in a bulk volume, an issue that probably requires the involvement of geotechnical engineers.

Finally, the treatment following the PFAS immobilization process, that is, how the trapped PFASs can be destructed completely needs to be considered in the future. Soil amendment serves to reduce the mobility of PFAS, yet the contaminants remain in soils. As we have suggested in conclusion, besides traditional incineration and biodegradation, more innovative methods are under development including chemical reduction by hydrated electrons, thermal methods such as smoldering and hydrothermal treatment. How PFASs trapped in clay particles can be destroyed thermally or chemically has not been evaluated.

Supplemental Information

S.1. Materials and methods

S.1.1. Details on soil characterization

S.1.2. Particle size distribution of FS100

S.1.3. X-ray diffraction (XRD) of FS100

S.1.4. Additional experimental details for evaluating the effect of pH

S.1.5. Control test

S.1.6. Details on the instrumental analysis method

S.2. Results and Discussions

S.2.1. Profiles of retained PFASs for four AFFF-contaminated soils

S.2.2. Profiles of leaching PFASs in four AFFF-contaminated soils

S.2.3. Leaching PFASs at different FS100 dosages

S.2.4. Leaching PFASs at different pH

S.2.5. Comparison of FS100, GAC, Biochar, and FS100/Bentonite on PFAS reduction

S.1. Materials and methods

S.1.1 Details on soil characterization

Table S1. Additional details on the physical and chemical properties of four contaminated soils; CAN 1 and 2 were collected from unnamed contaminated sites in Canada and US1 and US2 were collected from an airforce base from the US.

ID	Element Composition, ppm						Percent Base Saturation, %					K/Mg Ratio	Saturation P, %	Saturation Al, %
	P	K	Mg	Ca	Na	Al	K	Mg	Ca	H	Na			
CAN1	8	20	110	6250	92	33	0.2	2.8	95.8	-	1.2	0.07	1	0
CAN2	29	78	44	980	30	865	2.9	5.4	72.1	17.6	1.9	0.54	4	1.1
US1	24	20	103	1270	14	428	0.7	11.7	86.9	-	0.8	0.06	2	0
US2	20	13	80	1330	12	429	0.5	9.0	89.9	-	0.7	0.06	1	0

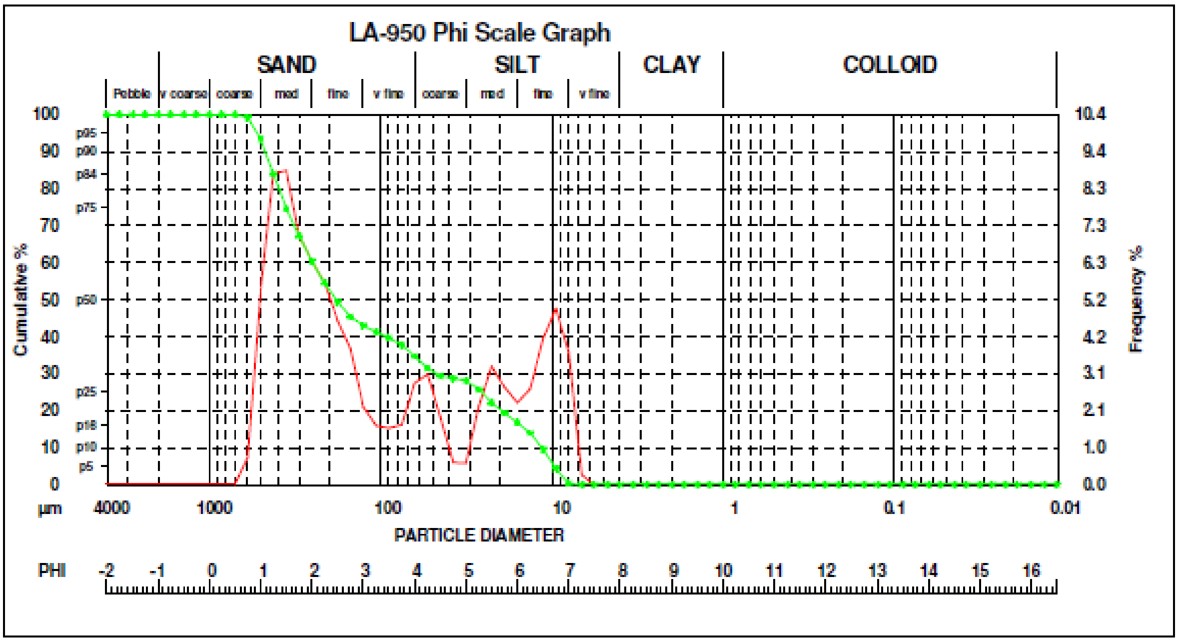
S.1.2 Moisture content of four contaminated soils

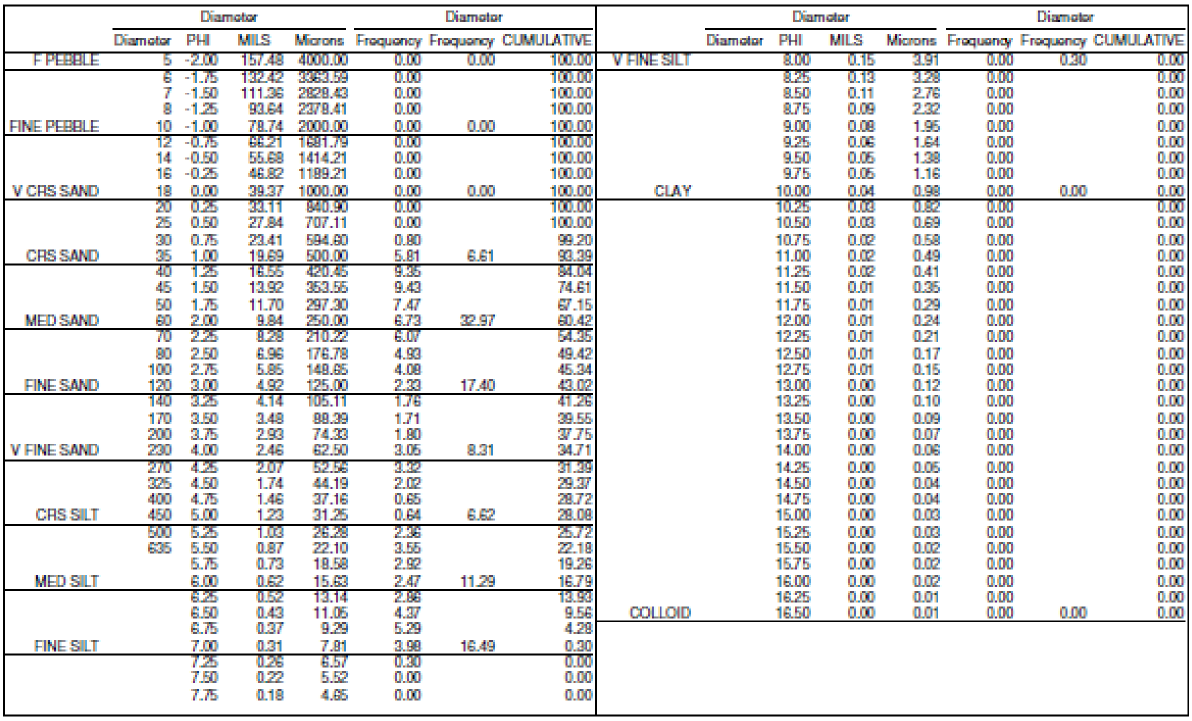
The moisture content of soils can be determined by the difference between the soil mass before the oven drying and the soil mass after the oven drying.

Table S2 Four air-dried soil samples mass before and after 24 hours of oven drying at 105 °C

Soil	**Mass before oven dry (g)**	**Mass after oven dry (g)**
CAN1	3.01	3.02
	3.01	3.02
	2.84	2.84
CAN2	2.91	2.91
	2.85	2.86
	2.97	2.97
US1	2.99	2.99
	3.08	3.08
	3.10	3.10
US2	3.00	3.00
	3.04	3.04
	3.11	3.09

S.1.3 Particle size distribution of FS100





	Diameter				Diameter		
	Diameter	PHI	MILS	Microns	Frequency	Frequency	CUMULATIVE
F PEBBLE	5	-2.00	157.48	4000.00	0.00	0.00	100.00
	6	-1.75	132.42	3363.59	0.00		100.00
	7	-1.50	111.36	2828.43	0.00		100.00
	8	-1.25	93.64	2378.41	0.00		100.00
FINE PEBBLE	10	-1.00	78.74	2000.00	0.00	0.00	100.00
	12	-0.75	66.21	1681.79	0.00		100.00
	14	-0.50	55.68	1414.21	0.00		100.00
	16	-0.25	46.82	1189.21	0.00		100.00
V CRS SAND	18	0.00	39.37	1000.00	0.00	0.00	100.00
	20	0.25	33.11	840.90	0.00		100.00
	25	0.50	27.84	707.11	0.00		100.00
	30	0.75	23.41	594.60	0.80		99.20
CRS SAND	35	1.00	19.69	500.00	5.81	6.61	93.39
	40	1.25	16.55	420.45	9.35		84.04
	45	1.50	13.92	353.55	9.43		74.61
	50	1.75	11.70	297.30	7.47		67.15
MED SAND	60	2.00	9.84	250.00	6.73	32.97	60.42
	70	2.25	8.28	210.22	6.07		54.35
	80	2.50	6.96	176.78	4.93		49.42
	100	2.75	5.85	148.65	4.08		45.34
FINE SAND	120	3.00	4.92	125.00	2.33	17.40	43.02
	140	3.25	4.14	105.11	1.76		41.26
	170	3.50	3.48	88.39	1.71		39.55
	200	3.75	2.93	74.33	1.80		37.75
V FINE SAND	230	4.00	2.46	62.50	3.05	8.31	34.71
	270	4.25	2.07	52.56	3.32		31.39
	325	4.50	1.74	44.19	2.02		29.37
	400	4.75	1.46	37.16	0.65		28.72
CRS SILT	450	5.00	1.23	31.25	0.64	6.62	28.08
	500	5.25	1.03	26.28	2.36		25.72
	635	5.50	0.87	22.10	3.55		22.18
		5.75	0.73	18.58	2.92		19.26
MED SILT		6.00	0.62	15.63	2.47	11.29	16.79
		6.25	0.52	13.14	2.86		13.93
		6.50	0.43	11.05	4.37		9.56
		6.75	0.37	9.29	5.29		4.28
FINE SILT		7.00	0.31	7.81	3.98	16.49	0.30
		7.25	0.26	6.57	0.30		0.00
		7.50	0.22	5.52	0.00		0.00
		7.75	0.18	4.65	0.00		0.00
V FINE SILT		8.00	0.15	3.91	0.00	0.30	0.00
		8.25	0.13	3.28	0.00		0.00
		8.50	0.11	2.76	0.00		0.00
		8.75	0.09	2.32	0.00		0.00
		9.00	0.08	1.95	0.00		0.00
		9.25	0.06	1.64	0.00		0.00
		9.50	0.05	1.38	0.00		0.00
		9.75	0.05	1.16	0.00		0.00
CLAY		10.00	0.04	0.98	0.00	0.00	0.00
		10.25	0.03	0.82	0.00		0.00
		10.50	0.03	0.69	0.00		0.00
		10.75	0.02	0.58	0.00		0.00
		11.00	0.02	0.49	0.00		0.00
		11.25	0.02	0.41	0.00		0.00
		11.50	0.01	0.35	0.00		0.00
		11.75	0.01	0.29	0.00		0.00
		12.00	0.01	0.24	0.00		0.00
		12.25	0.01	0.21	0.00		0.00
		12.50	0.01	0.17	0.00		0.00
		12.75	0.01	0.15	0.00		0.00
		13.00	0.00	0.12	0.00		0.00
		13.25	0.00	0.10	0.00		0.00
		13.50	0.00	0.09	0.00		0.00
		13.75	0.00	0.07	0.00		0.00
		14.00	0.00	0.06	0.00		0.00
		14.25	0.00	0.05	0.00		0.00
		14.50	0.00	0.04	0.00		0.00
		14.75	0.00	0.04	0.00		0.00
		15.00	0.00	0.03	0.00		0.00
		15.25	0.00	0.03	0.00		0.00
		15.50	0.00	0.02	0.00		0.00
		15.75	0.00	0.02	0.00		0.00
		16.00	0.00	0.02	0.00		0.00
		16.25	0.00	0.01	0.00		0.00
COLLOID		16.50	0.00	0.01	0.00	0.00	0.00

Figure S1. The particle size distribution of FS100 determined by laser diffraction particle size analyzer LA-950 (HPRIBA Ltd., Japan)

S.1.4 X-Ray Diffraction (XRD) of FS100

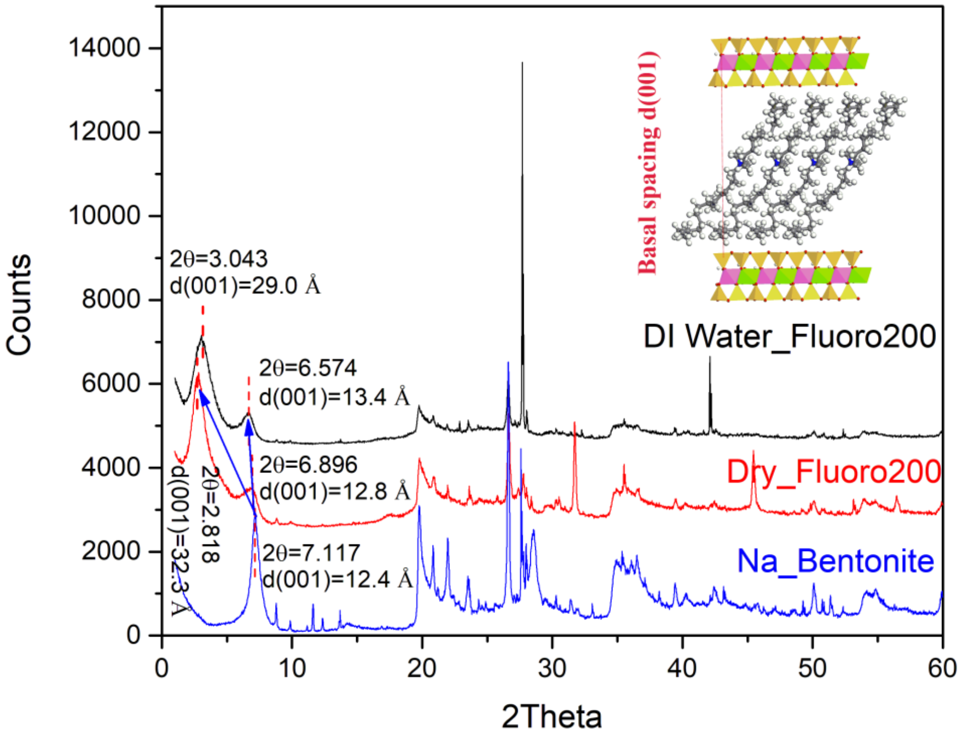


Figure S2. X-ray diffraction spectroscopy of water-saturated FS200, dry FS200, and the raw material of FS200 (Na-Bentonite). Note: FS100 and FS200 are the same products with different particle sizes.

S.1.5 Extraction fluids and samples preparation in pH effect experiment

Table S3. Preparation method and the measured pH of four extraction fluids that were used for evaluating the effect of solution pH on PFAS leachability

Extraction Fluid	Preparation Method	Actual pH
#1	Prepare 60/40 weight percent mixture of H_2SO_4/HNO_3, and dilute with DI water until the pH = 4.0-4.5 (EPA 1312 SPLP)	4.00
#2	NaCl(aq), pH = 5.6-6.0 Because the existence of CO_2	5.72
#3	DI Water + NaOH (pH = 7)	6.97
#4	Dilute NaOH(aq) with DI water until the pH = 8.0-8.5 (EPA 1313 LEAF)	8.02
Control Group	DI Water	-

Samples preparation method:

At each sampling time, 0.4 mL supernatant was pipetted into microcentrifuge tubes and subjected to high-speed centrifugation at 20,000 *x g* for 10 min. Then 100 µL supernatant was added into 4400 µL methanol/water 80/20 solvent (45 times dilution) to achieve the Phase 1 solution. The 100 µL of Phase 1 solution into was further diluted into 900 µL methanol/water 80/20 solvent (450 times dilution until now) to achieve Phase 2 solution. The samples (Phase 2 solutions) were stored at -20 °C before the analysis. The 180 µL of Phase 2 solution was transferred into HPLC vials, with the addition of 20 µL 20 ppb internal standard solution mixture (500 times dilution in total) before the analysis using the high-resolution LC-MS.

S.1.6 Recoveries of eight individual PFAS in HDPE containers

Recoveries of eight representative PFASs including anionic (PFOS, PFOA, PFDA, 6:2 FTSA, 8:2 FTSA), zwitterionic (PFOAB and PFOSB) and cationic groups (PFOAAmS) from aqueous solutions were examined in 60-mL HDPE bottles in triplicates**.** 50mL of 0.01M KCl aqueous solution was added into HDPE bottles as the background solution, and the PFAS mixture was spiked to achieve 100 ng/mL as the initial concentration. The bottles were shaken at a horizontal shaker for 8 days at 150 rpm and 20 °C. After shaking, a subsample was pipetted out and centrifuged at 20,000 g for 10 minutes, and the supernatant was pipetted out for further dilution with methanol and water. Diluted samples were stored at -20 °C and spiked with the internal standards before the analysis using high-resolution mass spectrometry.

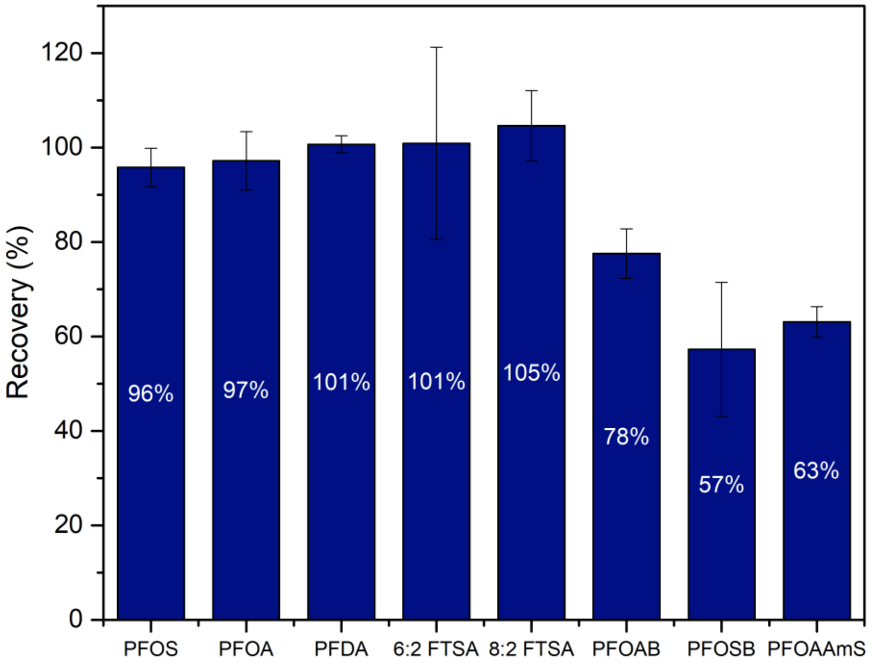


Figure S3. Recoveries of eight individual PFAS (including anionic, zwitterionic, and cationic) that were spiked into aqueous solutions in 60 mL HDPE bottles as determined by the decrease in aqueous phase concentration. The loss of PFAS was due to sorption to containers.

S.1.7 Details on the instrumental analysis method

Table S4. Details on the instrumental analysis method

Instrument	Ultra-high-performance liquid chromatography coupled to high-resolution mass spectrometry (UHPLC-HRMS)	
Software	The Dionex Ultimate 3000 LC (Chromeleon 7.2) coupled with Q-Exactive Orbitrap mass spectrometer (Xcalibur 2.3)	
Ionization	Polarity switching electrospray ionization	
Analytical column	Thermo Hypersil Gold aQ column (100 mm x 2.1 mm, 1.9 µm particle size)	
Trap column	Thermo Hypercarb (20 mm x 2.1 mm, 7 µm particle size)	
Mobile phase	A: HPLC water with 0.1% HCOOH (v/v) B: ACN with 0.1% HCOOH (v/v)	
Flow rate	0.55 mL/min	
Injection volume	10 µL	
Column temperature	40 ℃	
Gradient Profile	Time (min)	Percentage B (%)
	0	10
	7	72.5
	8.5	100
	12.5	100
	12.6	10
	14.5	10
Needle rinsing solution	1) a 1:1:1 volumetric mixture of acetonitrile, methanol, and isopropanol 2) HPLC-water containing 0.1% HCOOH	
Heated electrospray ionization source setting	Sheath gas flow rate	40 arbitrary units (a.u.)
	Auxiliary gas flow rate	15 arbitrary units (a.u.)
	Sweep gas flow rate	0 a.u.
	Capillary temperature	320 ℃
	Vaporizer temperature	350 ℃
	Spray voltage	either -4 kV or +4 kV (fast polarity-switching mode)
Orbitrap parameters	AGC target	3 x 10^6
	Maximum injection time	50 ms
	Resolution	At 70,000 FWHM at 200 m/z
	Mass scan range	150-1000 m/z (Full Scan MS mode)
Calibration	Linear regression, inverse weighting (1/x)	

S.2.Additional tables for “Results and Discussion” section

S.2.1 Profiles of retained PFAS for 4 AFFF-contaminated sandy soils

Table S5. Solvent extractable PFASs from 4 soils using the extraction method developed by Munoz et al., 2019 (n = 3)

PFAS	CAN1		CAN2		US1		US2	
	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g
PFPrA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFBA	9.3	1.7	1.9	0.2	1.7	0.1	0.0	0.0
PFPeA	22.4	4.7	5.1	0.3	3.7	0.4	1.1	0.1
PFHxA	148	32.6	13.9	0.8	25.3	1.8	6.4	0.2
PFHpA	14.3	3.4	3.2	0.3	4.3	0.4	2.8	0.0
PFOA	107.8	22.6	17.4	1.0	30.9	2.8	19.8	0.6
PFNA	12.9	2.8	1.9	0.2	0.5	0.0	0.5	0.1
PFDA	15.2	4.0	1.6	0.1	0.4	0.1	1.1	0.1
PFUnA	2.5	0.6	0.4	0.1	0.4	0.1	0.3	0.0
PFTrDA	0.2	0.3	0.0	0.0	0.0	0.0	0.0	0.0
PFTeDA	0.9	0.2	0.2	0.1	0.0	0.0	0.0	0.0
PFPrS	0.0	0.0	0.1	0.1	0.0	0.0	0.0	0.0
PFBS	5.1	1.1	1.5	0.1	0.4	0.0	0.0	0.0
PFPeS	18.3	4.2	3.3	0.2	1.5	0.2	0.3	0.0
PFHxS	483	102	45.1	2.9	52.5	4.1	15.5	0.3
PFHpS	44.3	9.5	5.3	0.3	1.4	0.2	0.7	0.0
PFOS	44441	9623	2132	137.8	867.8	86.7	1261	10.1
PFNS	0.0	0.0	0.0	0.0	18.9	1.4	0.0	0.0
PFDS	0.0	0.0	13.5	0.8	30.8	3.0	2.7	0.0
PFECHS	6.0	1.3	1.9	0.3	0.5	0.0	0.0	0.0
FBSA	1.9	0.4	10.4	1.0	0.6	0.1	0.6	0.1
FHxSA	544.9	94.6	479.6	36.3	105.7	11.4	130.3	0.2
FOSA	81.0	18.1	291.2	24.1	214.2	20.3	538.8	0.6
MeFOSA	1.3	0.3	1.0	0.1	0.9	0.2	2.0	0.1
EtFOSA	1.1	0.3	0.1	0.0	0.0	0.0	0.0	0.0
FOSAA	6.4	1.6	9.8	0.7	5.1	0.9	9.2	0.0
MeFOSAA	2.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0
EtFOSAA	8.3	2.4	3.9	0.7	0.4	0.4	0.0	0.0

3:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
5:3 Acid	0.9	0.3	0.0	0.0	0.0	0.0	0.0	0.0
7:3 Acid	6.3	1.6	1.7	0.2	2.8	0.3	0.0	0.0
4:2 FTS	0.8	0.2	0.3	0.0	0.0	0.0	0.0	0.0
6:2 FTS	731.3	158.1	195.7	14.8	19.8	2.1	5.6	0.1
8:2 FTS	1109	266.9	268.3	27.4	17.9	2.2	60.2	0.3
10:2 FTS	93.4	22.8	6.6	0.5	0.4	0.0	0.0	0.0
6:2 FTUA	0.2	0.3	0.0	0.0	0.0	0.0	0.0	0.0
8:2 FTUA	6.8	1.8	1.5	0.0	0.0	0.0	0.0	0.0
PFHxSAm	4111	926.1	33.5	1.7	34.6	0.3	25.4	0.3
PFOSAm	145.3	33.1	2.2	0.1	5.8	0.7	1.4	0.2
PFHxSAmS	3098	653.2	109.5	1.0	136.0	14.5	161.2	1.6
PFOSAmS	206.1	47.0	17.1	0.4	192.4	19.9	182.9	0.1
6:2 FTAB	4264	848.1	26.1	2.0	3.8	0.6	13.6	0.4
PFOAB	1.5	2.5	0.0	0.0	0.0	0.0	0.0	0.0
PFOSB	25.9	6.4	34.3	2.0	0.0	0.0	0.0	0.0
PFOANO	4.6	1.0	0.2	0.0	45.1	4.9	136.4	2.6
PFOSNO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
∑PFAS	59783	12901	3741	180	1827	18	2579	259

S.2.2 Profiles of leaching PFAS in 4 AFFF-contaminated sandy soil leachates

Table S6. PFASs detected in four soil leachates using the modified leaching protocol (n = 3)

PFAS	CAN1		CAN2		US1		US2	
	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g	Avg. Conc. ng/g	STD ng/g
PFPrA	8.1	1.5	0	0	0	0	0	0.0
PFBA	25.5	1.6	14.1	5.4	15.5	2.1	13.5	1.0
PFPeA	60.6	2.6	3.9	0.0	4.9	0.3	1.9	0.4
PFHxA	306.0	6.6	13.6	3.7	32.7	1.3	13.6	1.3
PFHpA	25.4	1.1	2.3	0.1	4.8	0.3	3.2	0.1
PFOA	326.3	13.8	9.1	0.4	25.0	1.1	17.7	1.5
PFNA	13.9	0.6	1.4	0.1	0.0	0.0	0.0	0.0
PFDA	7.3	0.1	0.7	0.2	0.0	0.0	0.2	0.3
PFUnA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFDoA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFTrDA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFTeDA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFPrS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFBS	16.6	0.6	25.8	43.4	33.5	30.8	7.7	2.8
PFPeS	33.3	1.1	1.0	0.0	1.6	0.1	0.0	0.0
PFHxS	724.2	29.3	18.4	0.2	59.0	0.8	15.4	0.9
PFHpS	40.8	1.9	1.8	0.0	1.1	0.1	0.4	0.1
PFOS	30050	946.8	1740	20.8	620.6	5.9	806.6	23.9
PFNS	275.5	159.1	6.5	0.4	15.1	0.2	2.9	0.3
PFDS	0.0	0.0	1.4	0.3	2.5	0.1	0.0	0.0
PFECHS	3.9	0.1	0.0	0.0	0.0	0.0	0.0	0.0
FBSA	10.8	0.5	6.6	0.4	1.3	0.1	1.1	0.4
FHxSA	860.3	28.0	212.5	15.2	98.9	1.1	108.1	22.3
FOSA	64.1	1.3	12.8	2.6	155.8	2.2	341.2	12.9
MeFOSA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
EtFOSA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
FOSAA	0.0	0.0	0.0	0.0	1.6	0.1	3.3	0.2
MeFOSAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
EtFOSAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

5:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
7:3 Acid	4.4	0.9	0.0	0.0	0.0	0.0	0.0	0.0
4:2 FTS	2.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0
6:2 FTS	661.7	33.3	197.1	5.0	25.2	1.0	15.1	0.5
8:2 FTS	590.8	9.3	156.6	2.7	13.2	0.2	38.7	0.6
10:2 FTS	1.2	0.7	0.0	0.0	0.0	0.0	0.0	0.0
6:2 FTUA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
8:2 FTUA	3.8	0.4	0.0	0.0	0.0	0.0	0.0	0.0
10:2 FTUA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFHxSAm	2729	103.0	2.2	0.5	52.6	2.6	37.7	7.7
PFOSAm	0.6	0.4	0.0	0.0	0.0	0.0	0.0	0.0
PFHxSAmS	2031	49.4	4.8	0.1	434.7	12.8	806.6	118.6
PFOSAmS	0.9	0.5	0.0	0.0	35.8	1.7	76.4	23.4
6:2 FTAB	343.7	14.4	3.2	0.1	3.6	1.1	21.8	3.0
PFOAB	3.3	0.6	0.0	0.0	0.0	0.0	0.0	0.0
PFOSB	9.9	2.4	0.0	0.0	0.0	0.0	0.0	0.0
PFOANO	0.0	0.0	0.0	0.0	41.6	1.4	220.5	35.8
PFOSNO	0	0	0	0	0	0	0	0
∑PFAS	39236	1412	2436	101.7	1681	67.4	2554	258.1

S.2.3 Leaching of PFAS with different FS100 dosages from 0.1% - 5%

Table S7. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing PFAS leachability from Soil CAN1 (n = 3)

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of Total PFAS Conc. decreased
0% (No NaN_3)	0	1.63	0.28	
	1	2963	91.3	
	3	3687	107.5	
	5	3828	70.7	
	8	3691	90.6	
0%	0	1.63	0.28	
	1	3293	261.2	
	3	3992	177.9	
	5	3860	228.0	
	8	3905	104.5	
0.1%	0	1.63	0.28	
	1	1551	248.6	52.91%
	3	1620	119.8	59.43%
	5	1466	118.1	62.02%
	8	1229	48.4	68.53%
0.5%	0	1.63	0.28	
	1	482.3	103.7	85.36%
	3	448.4	13.4	88.77%
	5	427.9	36.2	88.91%
	8	413.3	12.4	89.42%
1%	0	1.63	0.28	
	1	234.3	14.8	92.89%
	3	275.9	16.7	93.09%
	5	302.3	31.4	92.17%
	8	290.4	8.98	92.56%
3%	0	1.63	0.28	
	1	96.7	13.0	97.06%
	3	131.1	7.46	96.72%
	5	149.8	22.1	96.12%
	8	140.2	2.38	96.41%
5%	0	1.63	0.28	
	1	68.7	18.5	97.91%
	3	95.1	18.5	97.62%
	5	88.0	6.89	97.72%
	8	91.4	6.64	97.66%

Table S8. Leaching kinetics, the influence of dosages, and the impact of potential microbial activity on the decreasing PFOS leachability from Soil CAN1 (n = 3)

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of PFOS Conc. decreased
0% (No NaN_3)	0	0.13	0.13	
	1	2254	70.3	
	3	2809	104.6	
	5	2911	61.1	
	8	2816	92.4	
0%	0	0.13	0.13	
	1	2013	819.9	
	3	3094	133.9	
	5	2950	156.5	
	8	3005	94.7	
0.1%	0	0.13	0.13	
	1	963.0	236.0	52.16%
	3	945.5	93.6	69.44%
	5	742.6	94.4	74.83%
	8	533.8	49.7	82.24%
0.5%	0	0.13	0.13	
	1	121.5	58.1	93.96%
	3	52.4	31.5	98.31%
	5	17.8	3.12	99.40%
	8	7.70	4.77	99.74%
1%	0	0.13	0.13	
	1	24.4	15.1	98.79%
	3	15.4	6.70	99.50%
	5	9.67	6.74	99.67%
	8	1.77	0.53	99.94%
3%	0	0.13	0.13	
	1	8.77	5.73	99.56%
	3	5.83	2.82	99.81%
	5	12.2	16.1	99.59%
	8	0.52	0.17	99.98%
5%	0	0.13	0.13	
	1	4.51	2.72	99.78%
	3	6.25	4.15	99.80%
	5	0.41	0.08	99.99%
	8	0.41	0.20	99.99%

Table S9. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing PFHxSAm leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of PFHxSAm Conc. decreased
0% (No NaN_3)	0	0.00	0.00	
	1	176.7	4.58	
	3	250.1	5.27	
	5	270.5	19.1	
	8	254.2	18.4	
0%	0	0.00	0.00	
	1	207.8	35.1	
	3	264.6	25.4	
	5	265.4	20.1	
	8	272.9	10.3	
0.1%	0	0.00	0.00	
	1	173.9	4.88	16.32%
	3	212.2	11.5	19.78%
	5	243.1	11.9	8.41%
	8	236.6	13.4	13.31%
0.5%	0	0.00	0.00	
	1	96.3	21.6	53.67%
	3	111.0	17.9	58.06%
	5	120.1	19.3	54.74%
	8	128.0	7.23	53.09%
1%	0	0.00	0.00	
	1	31.2	5.66	84.96%
	3	44.0	7.64	83.37%
	5	56.5	11.1	78.70%
	8	63.9	7.87	76.60%
3%	0	0.00	0.00	
	1	5.88	0.39	97.17%
	3	10.3	3.46	96.11%
	5	12.6	1.08	95.27%
	8	15.9	1.52	94.18%
5%	0	0.00	0.00	
	1	3.85	1.40	98.15%
	3	7.36	3.91	97.22%
	5	4.77	1.07	98.20%
	8	6.14	1.31	97.75%

Table S10. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing PFHxSAmS leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	**Contact time Day**	**Avg. Conc. ppb**	**STD ppb**	**% of PFHxSAmS Conc. decreased**
0% (No NaN_3)	0	0.00	0.00	
	1	146.1	5.38	
	3	206.8	3.84	
	5	203.0	8.79	
	8	189.0	8.95	
0%	0	0.00	0.00	
	1	184.8	24.6	
	3	216.2	25.7	
	5	207.7	19.1	
	8	203.1	4.94	
0.1%	0	0.00	0.00	
	1	183.6	20.4	0.67%
	3	204.8	11.2	5.29%
	5	232.7	9.44	-12.04%
	8	213.8	13.4	-5.25%
0.5%	0	0.00	0.00	
	1	171.8	6.33	7.05%
	3	207.7	3.81	3.95%
	5	214.2	7.67	-3.14%
	8	199.7	1.48	1.66%
1%	0	0.00	0.00	
	1	131.5	7.44	28.83%
	3	160.7	5.07	25.68%
	5	174.0	11.9	16.25%
	8	156.0	4.36	23.21%
3%	0	0.00	0.00	
	1	52.1	3.00	71.84%
	3	75.0	2.12	65.33%
	5	81.1	3.16	60.98%
	8	77.7	0.81	61.77%
5%	0	0.00	0.00	
	1	38.4	16.3	79.23%
	3	49.0	9.53	77.35%
	5	49.7	5.34	76.07%
	8	48.5	4.58	76.14%

Table S11. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing FHxSA leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	**Contact time Day**	**Avg. Conc. ppb**	**STD ppb**	**% of FHxSA decreased**
0% (No NaN_3)	0	0.00	0.00	
	1	89.2	8.15	
	3	80.5	2.91	
	5	82.0	3.81	
	8	82.2	5.49	
0%	0	0.00	0.00	
	1	76.1	14.8	
	3	70.0	9.88	
	5	84.8	7.23	
	8	86.0	2.80	
0.1%	0	0.00	0.00	
	1	54.9	9.07	27.86%
	3	59.2	1.78	15.45%
	5	53.3	4.56	37.18%
	8	64.1	3.68	25.47%
0.5%	0	0.00	0.00	
	1	18.1	7.79	76.19%
	3	10.1	5.02	85.55%
	5	8.10	3.92	90.45%
	8	6.29	2.80	92.69%
1%	0	0.00	0.00	
	1	2.78	0.39	96.34%
	3	2.31	0.39	96.70%
	5	2.00	0.78	97.64%
	8	1.66	0.38	98.08%
3%	0	0.00	0.00	
	1	0.65	0.21	99.14%
	3	0.73	0.42	98.96%
	5	0.40	0.25	99.53%
	8	0.22	0.02	99.75%
5%	0	0.00	0.00	
	1	0.41	0.05	99.47%
	3	0.74	0.48	98.94%
	5	0.07	0.04	99.92%
	8	0.09	0.03	99.89%

Table S12. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing PFHxS leachability from Soil CAN1 (n = 3).

Sorbent dose **wt% of soil weight**	**Contact time** **Day**	**Avg. Conc.** **ppb**	**STD** **ppb**	**% of PFHxS Conc.** **decreased**
0% (No NaN_3)	0	0.00	0.00	
	1	70.2	2.42	
	3	73.0	1.73	
	5	75.0	4.21	
	8	71.7	1.78	
0%	0	0.00	0.00	
	1	62.4	20.0	
	3	71.4	1.15	
	5	75.0	7.28	
	8	72.4	2.93	
0.1%	0	0.00	0.00	
	1	34.1	8.88	45.32%
	3	31.2	1.04	56.38%
	5	25.8	0.62	65.65%
	8	21.5	1.49	70.28%
0.5%	0	0.00	0.00	
	1	4.87	2.72	92.19%
	3	1.59	0.52	97.78%
	5	0.86	0.39	98.85%
	8	0.44	0.27	99.39%
1%	0	0.00	0.00	
	1	0.59	0.32	99.05%
	3	0.28	0.11	99.60%
	5	0.15	0.10	99.80%
	8	0.07	0.03	99.90%
3%	0	0.00	0.00	
	1	0.16	0.11	99.75%
	3	0.13	0.06	99.81%
	5	0.12	0.18	99.84%
	8	0.00	0.00	99.99%
5%	0	0.00	0.00	
	1	0.08	0.05	99.88%
	3	0.18	0.12	99.74%
	5	0.00	0.00	99.99%
	8	0.00	0.00	99.99%

Table S13. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing 6:2 FTSA leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of 6:2 FTSA Conc. decreased
0% (No NaN_3)	0	0.77	0.07	
	1	63.0	3.79	
	3	66.1	0.91	
	5	68.0	4.22	
	8	63.6	1.58	
0%	0	0.77	0.07	
	1	57.0	13.2	
	3	67.6	2.59	
	5	68.0	5.33	
	8	66.2	3.33	
0.1%	0	0.77	0.07	
	1	37.6	7.22	34.11%
	3	40.4	2.71	40.28%
	5	36.0	1.84	46.99%
	8	36.2	1.51	45.26%
0.5%	0	0.77	0.07	
	1	9.91	4.35	82.61%
	3	5.67	2.30	91.62%
	5	4.25	1.70	93.74%
	8	3.45	1.18	94.79%
1%	0	0.77	0.07	
	1	2.10	0.27	96.31%
	3	1.72	0.08	97.45%
	5	1.41	0.29	97.93%
	8	1.27	0.05	98.07%
3%	0	0.77	0.07	
	1	1.07	0.29	98.13%
	3	1.03	0.13	98.47%
	5	0.79	0.03	98.83%
	8	0.57	0.03	99.14%
5%	0	0.77	0.07	
	1	0.88	0.00	98.45%
	3	0.98	0.14	98.55%
	5	0.65	0.14	99.04%
	8	0.53	0.04	99.20%

Table S14. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing 8:2 FTSA leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% 8:2 FTSA Conc. decreased
0% (No NaN_3)	0	0.00	0.00	
	1	37.0	1.49	
	3	51.3	0.92	
	5	57.9	1.09	
	8	57.0	2.79	
0%	0	0.00	0.00	
	1	36.4	9.67	
	3	51.9	4.32	
	5	55.1	4.35	
	8	59.1	0.93	
0.1%	0	0.00	0.00	
	1	21.9	4.03	39.83%
	3	28.0	0.80	46.00%
	5	27.1	1.43	50.82%
	8	25.0	1.00	57.72%
0.5%	0	0.00	0.00	
	1	4.89	2.24	86.56%
	3	2.56	0.60	95.07%
	5	1.43	0.71	97.40%
	8	0.92	0.56	98.45%
1%	0	0.00	0.00	
	1	0.80	0.29	97.80%
	3	0.45	0.15	99.14%
	5	0.35	0.25	99.37%
	8	0.16	0.06	99.73%
3%	0	0.00	0.00	
	1	0.22	0.12	99.41%
	3	0.18	0.16	99.66%
	5	0.27	0.36	99.52%
	8	0.00	0.00	99.99%
5%	0	0.00	0.00	
	1	0.11	0.16	99.69%
	3	0.21	0.18	99.59%
	5	0.00	0.00	99.99%
	8	0.00	0.00	99.99%

Table S15. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing 6:2 FTAB leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of 6:2 FTAB Conc. decreased
0% (No NaN_3)	0	0.00	0.00	
	1	21.0	1.42	
	3	28.1	1.41	
	5	33.5	2.95	
	8	35.8	6.00	
0%	0	0.00	0.00	
	1	25.1	1.79	
	3	33.0	4.76	
	5	30.1	2.01	
	8	34.4	1.44	
0.1%	0	0.00	0.00	
	1	26.9	3.55	-6.85%
	3	32.6	0.44	1.44%
	5	41.6	5.45	-38.19%
	8	42.8	1.25	-24.39%
0.5%	0	0.00	0.00	
	1	34.6	2.38	-37.74%
	3	44.0	3.00	-33.25%
	5	48.0	1.26	-59.60%
	8	55.8	4.19	-62.24%
1%	0	0.00	0.00	
	1	35.0	1.97	-39.41%
	3	45.5	1.58	-37.78%
	5	52.5	0.93	-74.49%
	8	60.6	5.76	-76.30%
3%	0	0.00	0.00	
	1	25.0	1.65	0.50%
	3	36.2	1.72	-9.60%
	5	40.2	2.19	-33.40%
	8	43.4	1.64	-26.15%
5%	0	0.00	0.00	
	1	19.2	3.64	23.66%
	3	28.0	2.37	15.18%
	5	30.8	0.68	-2.46%
	8	34.5	1.20	-0.35%

Table S16. Leaching kinetics, the influence of dosages, and the impact of potential microbial activities on the decreasing PFOA leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of PFOA Conc. decreased
0% (No NaN_3)	0	0.31	0.04	
	1	30.8	1.18	
	3	32.6	0.83	
	5	33.8	1.06	
	8	32.0	0.56	
0%	0	0.31	0.04	
	1	27.9	7.54	
	3	33.3	0.78	
	5	33.2	3.22	
	8	32.6	1.38	
0.1%	0	0.31	0.04	
	1	17.1	3.86	38.70%
	3	17.7	0.58	46.70%
	5	16.5	0.63	50.21%
	8	15.0	0.95	54.19%
0.5%	0	0.31	0.04	
	1	4.05	1.82	85.49%
	3	1.78	0.91	94.65%
	5	1.38	0.61	95.86%
	8	0.97	0.29	97.03%
1%	0	0.31	0.04	
	1	0.81	0.12	97.10%
	3	0.64	0.09	98.08%
	5	0.59	0.11	98.22%
	8	0.48	0.13	98.54%
3%	0	0.31	0.04	
	1	1.01	0.77	96.37%
	3	0.46	0.01	98.60%
	5	0.28	0.28	99.14%
	8	0.41	0.11	98.73%
5%	0	0.31	0.04	
	1	0.49	0.12	98.23%
	3	0.54	0.24	98.37%
	5	0.19	0.18	99.41%
	8	0.24	0.21	99.26%

Table S17. Leaching kinetics, the influence of dosages, and microbial activities on the decreasing PFHxA leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	**Contact time Day**	**Avg. Conc. ppb**	**STD ppb**	**% of PFHxA Conc. decreased**
0% (No NaN_3)	0	0.27	0.03	
	1	30.4	0.73	
	3	31.9	0.71	
	5	31.2	1.18	
	8	31.1	0.39	
0%	0	0.27	0.03	
	1	27.4	4.51	
	3	30.6	1.91	
	5	30.8	2.92	
	8	30.6	0.66	
0.1%	0	0.27	0.03	
	1	20.2	3.56	26.44%
	3	21.8	0.58	28.76%
	5	21.4	0.83	30.52%
	8	21.9	0.69	28.46%
0.5%	0	0.27	0.03	
	1	7.99	2.20	70.88%
	3	5.65	1.71	81.52%
	5	5.29	1.44	82.83%
	8	4.70	0.96	84.65%
1%	0	0.27	0.03	
	1	2.12	0.45	92.26%
	3	1.81	0.28	94.09%
	5	1.73	0.38	94.40%
	8	1.65	0.28	94.61%
3%	0	0.27	0.03	
	1	0.55	0.20	97.99%
	3	0.48	0.21	98.44%
	5	0.52	0.03	98.31%
	8	0.47	0.02	98.46%
5%	0	0.27	0.03	
	1	0.37	0.04	98.64%
	3	0.53	0.13	98.27%
	5	0.33	0.07	98.92%
	8	0.33	0.02	98.93%

Table S18. Leaching kinetics, the influence of dosages, and microbial activities on the decreasing PFNS leachability from Soil CAN1 (n = 3).

Sorbent dose wt% of soil weight	Contact time Day	Avg. Conc. ppb	STD ppb	% of PFNS Conc. decrease
0% (No NaN_3)	0	0.00	0.00	
	1	15.8	0.85	
	3	23.6	0.66	
	5	26.7	1.69	
	8	25.6	1.35	
0%	0	0.00	0.00	
	1	14.9	6.48	
	3	30.4	3.22	
	5	25.6	1.48	
	8	27.6	15.91	
0.1%	0	0.00	0.00	
	1	0.00	0.00	100.00%
	3	6.80	0.79	77.60%
	5	5.80	0.97	77.33%
	8	0.00	0.00	100.00%
0.5%	0	0.00	0.00	
	1	0.87	0.36	94.16%
	3	0.45	0.37	98.50%
	5	0.14	0.02	99.45%
	8	0.04	0.06	99.87%
1%	0	0.00	0.00	
	1	0.20	0.15	98.68%
	3	0.13	0.05	99.58%
	5	0.10	0.09	99.62%
	8	0.00	0.00	100.00%
3%	0	0.00	0.00	
	1	0.06	0.03	99.62%
	3	0.03	0.03	99.89%
	5	0.10	0.14	99.60%
	8	0.00	0.00	100.00%
5%	0	0.00	0.00	
	1	0.03	0.04	99.82%
	3	0.05	0.04	99.84%
	5	0.00	0.00	100.00%
	8	0.00	0.00	100.00%

S.2.4 Leaching PFAS with solutions of different pHs

Table S19. Profiles of PFASs in soil leachate at different pH values. Control was extracted using deionized water without any pH adjustment.

PFAS	pH = 4.00		pH = 5.72		pH = 6.97		pH = 8.02		Control	
	Avg. Conc. ppb	STD ppb	Avg. Conc. ppb	STD ppb	Avg. Conc. ppb	STD ppb	Avg. Conc. ppb	STD ppb	Avg. Conc. ppb	STD ppb
PFPrA	0.0	0.0	7.5	13.0	0.0	0.0	0.0	0.0	0.0	0.0
PFBA	60.8	9.5	84.8	9.8	62.1	15.0	71.4	12.2	90.6	7.2
PFPeA	0.0	0.0	0.0	0.0	0.0	0.0	4.2	7.2	8.8	7.6
PFHxA	26.4	0.4	31.1	2.2	25.0	2.4	26.1	3.1	29.7	3.1
PFHpA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOA	6.2	0.5	10.0	2.3	6.7	0.9	7.9	2.2	11.2	1.9
PFNA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFDA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFUnA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFDoA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFTrDA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFTeDA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFPrS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFBS	2610	10.4	2630	18.5	2614	50.8	2592	34.2	2607	71.9
PFPeS	0.0	0.0	1.4	2.4	0.9	1.5	0.0	0.0	0.0	0.0
PFHxS	53.9	3.1	56.3	2.1	55.2	0.8	51.4	0.7	54.1	1.7
PFHpS	2.2	3.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOS	1194	8.5	1224	66.9	1309	70.7	1152	51.2	1277	60.9
PFNS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.9	3.3
PFDS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFECHS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
FBSA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
FHxSA	47.4	1.4	47.3	1.7	50.0	2.6	47.2	2.1	50.1	1.7

FOSA	3.4	0.6	2.9	0.1	3.3	0.4	3.1	0.3	2.9	0.5
MeFOSA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
EtFOSA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
FOSAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MeFOSAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
EtFOSAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
5:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
7:3 Acid	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4:2 FTS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
6:2 FTS	49.5	3.8	53.9	4.3	49.7	3.9	50.8	9.2	48.7	4.0
8:2 FTS	24.3	0.6	27.1	3.3	28.9	3.2	25.2	1.7	31.9	0.9
10:2 FTS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
6:2 FTUA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
8:2 FTUA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFHxSAm	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOSAm	112.2	1.7	115.4	11.5	130.7	12.7	113.1	6.8	122.8	16.6
PFHxSAmS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOSAmS	93.5	3.9	99.8	8.8	110.3	11.4	97.8	7.6	103.1	15.1
6:2 FTAB	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOAB	26.4	2.7	47.3	47.2	33.3	4.8	24.6	5.5	28.8	0.9
PFOSB	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOANO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PFOSNO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
∑PFAS	1640	30	1724	166	1803	115	1603	98	1771	118

The samples were 500 times diluted as the PFOS concentration was very high; without the dilution, PFOS would oversaturate the signal of the instruments and may also cause carryover issues. The data for PFBS and PFBA (in pink bands) were excluded because they were regarded as the instrumental background, as the peak areas of PFBS and PFBA were at a similar level before and after dilution. Because of the high dilution factor, some PFAS analytes were below the detection limits.

S.2.5 Comparison of FS100, GAC, Biochar, and FS100/Bentonite on PFAS reduction

Table S20. Comparative assessment of FS100, GAC, Biochar and FS100/bentonite system in lowering the leachability of PFAS using Soil CAN1 (n = 3)

Analytes	**No sorbent**		**% of PFAS Conc. decreased with 0.5% sorbent**			
	Avg. Conc. ng/g	**STD ng/g**	**FS100**	**GAC**	**Biochar**	**FS100/Bentonite**
PFPrA	0.8	0.2	100%	100%	100%	100%
PFBA	2.6	0.2	80.2%	80.8%	8.3%	80.6%
PFPeA	6.1	0.3	88.2%	89.8%	17.4%	86.5%
PFHxA	30.6	0.7	87.2%	81.6%	-12.9%	85.5%
PFHpA	2.5	0.1	92.9%	95.6%	14.8%	93.7%
PFOA	32.6	1.4	93.6%	81.7%	-18.5%	92.9%
PFNA	1.4	0.1	100%	100%	33.6%	100%
PFDA	0.7	0.0	100%	100%	100%	100%
PFUnA	0.0	0.0	N/A	N/A	N/A	N/A
PFDoA	0.0	0.0	N/A	N/A	N/A	N/A
PFTrDA	0.0	0.0	N/A	N/A	N/A	N/A
PFTeDA	0.0	0.0	N/A	N/A	N/A	N/A
PFPrS	0.0	0.0	N/A	N/A	N/A	N/A
PFBS	1.7	0.1	80.0%	79.7%	-1.9%	79.9%
PFPeS	3.3	0.1	97.1%	91.4%	18.3%	100%
PFHxS	72.4	2.9	98.9%	82.7%	-30.8%	99.0%
PFHpS	4.1	0.2	98.0%	91.8%	14.7%	100.0%
PFOS	3005.0	94.7	99.9%	74.1%	-37.7%	99.8%
PFNS	27.6	15.9	99.1%	67.9%	-24.5%	100.0%
PFDS	0.0	0.0	N/A	N/A	N/A	N/A
PFECHS	0.4	0.0	100%	100%	79.4%	100%
FBSA	1.1	0.1	100%	100%	100%	100%
FHxSA	86.0	2.8	97.4%	78.7%	-39.6%	96.6%
FOSA	6.4	0.1	98.1%	80.5%	5.1%	100%
MeFOSA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSA	0.0	0.0	N/A	N/A	N/A	N/A
FOSAA	0.0	0.0	N/A	N/A	N/A	N/A
MeFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
3:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A

4:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
5:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
7:3 Acid	0.4	0.1	100%	100%	100%	100%
4:2 FTS	0.2	0.0	100%	100%	100%	100%
6:2 FTS	66.2	3.3	90.5%	81.2%	11.5%	93.4%
8:2 FTS	59.1	0.9	99.4%	63.8%	-32.4%	100.0%
10:2 FTS	0.1	0.1	100%	100%	100%	100%
6:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
8:2 FTUA	0.4	0.0	100%	100%	100%	100%
10:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAm	272.9	10.3	69.7%	74.4%	-34.3%	71.9%
PFOSAm	0.1	0.0	100%	100%	100%	100%
PFHxSAmS	203.1	4.9	4.6%	60.3%	-44.4%	33.0%
PFOSAmS	0.1	0.1	100%	100%	100%	100%
6:2 FTAB	34.4	1.4	-21.4%	24.5%	-29.3%	3.0%
PFOAB	0.3	0.1	100%	100%	100%	100%
PFOSB	1.0	0.2	100%	100%	100%	100%
PFOANO	0.0	0.0	N/A	N/A	N/A	N/A
PFOSNO	0.0	0.0	N/A	N/A	N/A	N/A
∑PFAS	3923.6	141.2	89.74%	74.75%	-28.91%	91.55%

N/A means initial PFAS concentration was below the detection limit.

Table S21. Comparative assessment of FS100, GAC, Biochar and FS100/bentonite system in lowering the leachability of PFAS using Soil CAN2 (n = 3).

PFAS	No sorbent		% of PFAS Conc. decreased with 0.5% sorbent			
	Avg. Conc. ppb	STD ppb	FS100	GAC	Biochar	FS100/Bentonite
PFPrA	0.0	0.0	N/A	N/A	N/A	N/A
PFBA	1.4	0.5	14.2%	-18.9%	6.7%	8.7%
PFPeA	0.4	0.0	100%	100%	-1.8%	100%
PFHxA	1.4	0.4	71.4%	42.1%	-3.5%	76.8%
PFHpA	0.2	0.0	100%	100%	-1.4%	100%
PFOA	0.9	0.0	79.7%	74.5%	-15.3%	63.9%
PFNA	0.1	0.0	100.0%	100.0%	21.2%	100.0%
PFDA	0.1	0.0	100.0%	100.0%	22.4%	100.0%
PFUnA	0.0	0.0	N/A	N/A	N/A	N/A
PFDoA	0.0	0.0	N/A	N/A	N/A	N/A
PFTrDA	0.0	0.0	N/A	N/A	N/A	N/A
PFTeDA	0.0	0.0	N/A	N/A	N/A	N/A
PFPrS	0.0	0.0	N/A	N/A	N/A	N/A
PFBS	2.6	4.3	-14.2%	93.8%	61.2%	82.6%
PFPeS	0.1	0.0	100.0%	100.0%	2.3%	100.0%
PFHxS	1.8	0.0	97.4%	96.7%	3.6%	97.2%
PFHpS	0.2	0.0	100.0%	100.0%	4.4%	100.0%
PFOS	174.0	2.1	99.6%	99.3%	10.5%	99.8%
PFNS	0.7	0.0	100%	100%	20.2%	100%
PFDS	0.1	0.0	100%	100%	40.1%	100%
PFECHS	0.0	0.0	N/A	N/A	N/A	N/A
FBSA	0.7	0.0	100%	100%	6.6%	100%
FHxSA	21.3	1.5	99.4%	99.5%	18.5%	99.3%
FOSA	1.3	0.3	100%	94.2%	17.4%	100%
MeFOSA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSA	0.0	0.0	N/A	N/A	N/A	N/A
FOSAA	0.0	0.0	N/A	N/A	N/A	N/A
MeFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
3:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A

4:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
5:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
7:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
4:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTS	19.7	0.5	95.4%	94.9%	3.5%	95.4%
8:2 FTS	15.7	0.3	99.7%	98.9%	13.1%	100%
10:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
8:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
10:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAm	0.2	0.0	-15.5%	100%	49.4%	100.0%
PFOSAm	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAmS	0.5	0.0	-36.1%	100%	100%	66.9%
PFOSAmS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTAB	0.3	0.0	-19.8%	100%	-15.5%	100%
PFOAB	0.0	0.0	N/A	N/A	N/A	N/A
PFOSB	0.0	0.0	N/A	N/A	N/A	N/A
PFOANO	0.0	0.0	N/A	N/A	N/A	N/A
PFOSNO	0.0	0.0	N/A	N/A	N/A	N/A
∑PFAS	243.6	8.6	96.8%	97.8%	11.3%	98.4%

N/A means initial PFAS concentration was below the detection limit.

Table S22. Comparative assessment of FS100, GAC, Biochar and FS100/bentonite system in lowering the leachability of PFAS using Soil US1 (n = 3)

PFAS	No sorbent		% of PFAS Conc. decreased with 0.5% sorbent			
	Avg. Conc. ppb	STD ppb	FS100	GAC	Biochar	FS100/Bentonite
PFPrA	0.0	0.0	N/A	N/A	N/A	N/A
PFBA	1.5	0.2	0.1%	-19.6%	15.9%	-8.4%
PFPeA	0.5	0.0	100%	100%	11.5%	100%
PFHxA	3.3	0.1	80.0%	67.9%	13.3%	70.7%
PFHpA	0.5	0.0	100%	100%	12.3%	100%
PFOA	2.5	0.1	81.6%	83.5%	11.5%	82.6%
PFNA	0.0	0.0	N/A	N/A	N/A	N/A
PFDA	0.0	0.0	N/A	N/A	N/A	N/A
PFUnA	0.0	0.0	N/A	N/A	N/A	N/A
PFDoA	0.0	0.0	N/A	N/A	N/A	N/A
PFTrDA	0.0	0.0	N/A	N/A	N/A	N/A
PFTeDA	0.0	0.0	N/A	N/A	N/A	N/A
PFPrS	0.0	0.0	N/A	N/A	N/A	N/A
PFBS	3.4	3.1	69.3%	69.7%	70.5%	77.1%
PFPeS	0.2	0.0	100%	100%	18.6%	100%
PFHxS	5.9	0.1	98.4%	97.7%	12.4%	97.8%
PFHpS	0.1	0.0	100%	100%	19.5%	100%
PFOS	62.1	0.6	99.3%	97.9%	11.9%	99.1%
PFNS	1.5	0.0	100%	93.5%	13.6%	100%
PFDS	0.3	0.0	100%	73.7%	17.0%	100%
PFECHS	0.0	0.0	N/A	N/A	N/A	N/A
FBSA	0.1	0.0	100%	100%	20.9%	100%
FHxSA	9.9	0.1	99.3%	97.4%	15.0%	99.0%
FOSA	15.6	0.2	99.8%	97.2%	30.1%	99.8%
MeFOSA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSA	0.0	0.0	N/A	N/A	N/A	N/A
FOSAA	0.2	0.0	100%	100%	13.8%	100%
MeFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
3:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
4:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A

5:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
7:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
4:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTS	2.5	0.1	66.9%	64.1%	6.9%	66.9%
8:2 FTS	1.3	0.0	100%	100%	16.1%	100%
10:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
8:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
10:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAm	5.3	0.3	95.0%	100%	40.9%	95.0%
PFOSAm	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAmS	43.5	1.3	63.0%	99.1%	13.8%	79.4%
PFOSAmS	3.6	0.2	82.8%	86.3%	7.9%	97.6%
6:2 FTAB	0.4	0.1	22.7%	100%	23.3%	100%
PFOAB	0.0	0.0	N/A	N/A	N/A	N/A
PFOSB	0.0	0.0	N/A	N/A	N/A	N/A
PFOANO	4.2	0.1	61.2%	100%	13.8%	87.3%
PFOSNO	0.0	0.0	N/A	N/A	N/A	N/A
∑PFAS	168.0	0.9	85.7%	95.0%	16.4%	90.9%

N/A means initial PFAS concentration was below the detection limit.

Table S23. Comparative assessment of FS100, GAC, Biochar and FS100/bentonite system in lowering the leachability of PFAS using Soil US2 (n = 3)

PFAS	**No sorbent**		**% of PFAS Conc. decreased with 0.5% sorbent**			
	Avg. Conc. ppb	**STD ppb**	**FS100**	**GAC**	**Biochar**	**FS100/Bentonite**
PFPrA	0.0	0.0	N/A	N/A	N/A	N/A
PFBA	1.3	0.1	-15.6%	-4.3%	-17.4%	3.7%
PFPeA	0.2	0.0	81.8%	100%	-10.1%	100%
PFHxA	1.4	0.1	45.4%	55.0%	-20.7%	63.6%
PFHpA	0.3	0.0	100%	100%	-20.0%	100%
PFOA	1.8	0.2	71.0%	71.8%	-12.4%	75.0%
PFNA	0.0	0.0	N/A	N/A	N/A	N/A
PFDA	0.0	0.0	100%	100%	-105.3%	100%
PFUnA	0.0	0.0	N/A	N/A	N/A	N/A
PFDoA	0.0	0.0	N/A	N/A	N/A	N/A
PFTrDA	0.0	0.0	N/A	N/A	N/A	N/A
PFTeDA	0.0	0.0	N/A	N/A	N/A	N/A
PFPrS	0.0	0.0	N/A	N/A	N/A	N/A
PFBS	0.8	0.3	-23.4%	-33.8%	-24.2%	-18.3%
PFPeS	0.0	0.0	N/A	N/A	N/A	N/A
PFHxS	1.5	0.1	96.0%	97.2%	-11.1%	97.2%
PFHpS	0.0	0.0	100%	100%	-20.4%	100%
PFOS	80.7	2.4	99.7%	98.7%	-8.9%	99.7%
PFNS	0.3	0.0	100%	100%	-13.5%	100%
PFDS	0.0	0.0	N/A	N/A	N/A	N/A
PFECHS	0.0	0.0	N/A	N/A	N/A	N/A
FBSA	0.1	0.0	100%	100%	17.5%	100%
FHxSA	10.8	2.2	99.7%	98.5%	-0.9%	99.5%
FOSA	34.1	1.3	99.8%	98.5%	1.7%	99.8%
MeFOSA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSA	0.0	0.0	N/A	N/A	N/A	N/A
FOSAA	0.3	0.0	100%	100.0%	-17.8%	100%
MeFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
EtFOSAA	0.0	0.0	N/A	N/A	N/A	N/A
3:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
4:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A

5:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
7:3 Acid	0.0	0.0	N/A	N/A	N/A	N/A
4:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTS	1.5	0.1	50.3%	35.0%	0.1%	46.2%
8:2 FTS	3.9	0.1	100%	98.6%	-2.0%	100%
10:2 FTS	0.0	0.0	N/A	N/A	N/A	N/A
6:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
8:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
10:2 FTUA	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAm	3.8	0.8	100%	100%	29.7%	100%
PFOSAm	0.0	0.0	N/A	N/A	N/A	N/A
PFHxSAmS	80.7	11.9	73.8%	99.6%	-32.9%	84.8%
PFOSAmS	7.6	2.3	92.3%	93.8%	-34.1%	96.6%
6:2 FTAB	2.2	0.3	44.0%	100%	-30.3%	73.2%
PFOAB	0.0	0.0	N/A	N/A	N/A	N/A
PFOSB	0.0	0.0	N/A	N/A	N/A	N/A
PFOANO	22.0	3.6	72.5%	99.8%	-34.4%	88.3%
PFOSNO	0.0	0.0	N/A	N/A	N/A	N/A
∑PFAS	255.4	23.0	86.7%	97.2%	-17.3%	92.2%

N/A means initial PFAS concentration was below the detection limit.

S.2.6 Adsorption of PFOAAmS on natural Na-montmorillonite

A preliminary test that evaluated the adsorption of PFOAAmS on a natural Na-montmorillonite clay (SWy-2) was conducted. Detailed experimental design and the results are presented in the following tables.

Table S24 Experimental design of PFOAAmS adsorption study

PFAS	PFOAAmS (cation)
Initial concentration level of PFOAAmS	ppm level
Sorbent	Natural bentonite clay SWy-2 (Particle size: 0.1-0.5μm)
Solid-to-liquid ratio	0.01g/ 50 mL;
Background solution	0.01M KCl (aq)
Containers	60 mL HDPE bottles
Equilibrium time	96 h
Microcentrifugation	20,000 g, 10 min
pH recorded	5.39

Table S25 Solid-liquid partitioning of PFOAAmS on natural Na^+ saturated bentonite

	Average concentration, ppb	Standard deviation, ppb
0 h	2080	240
96 h_SWy-2	33.5	6.7
96 h_Control	2093.3	80.2
Sorption on clay (aqueous concentration reduction)	98.4%	
logKd	2.5	

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