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Efficient and fast remediation of soil contaminated by per- and polyfluoroalkyl substances (PFAS) by high-frequency heating

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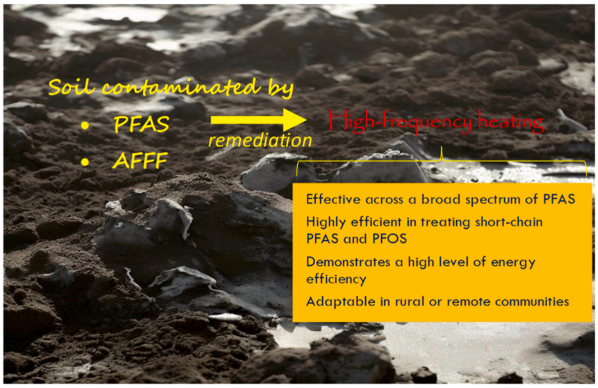
HIGHLIGHTS

- A new and energy efficient thermal technology for PFAS remediation.
- Rapid PFAS degradation achieved with this technology.
- It is effective regardless of the types of PFAS and soil.
- Minimal PFAS loss to water vapor during water evaporation.

GRAPHICAL ABSTRACT



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ABSTRACT

This study presents a novel thermal technology (high-frequency heating, HFH) for the decontamination of soil containing per- and polyfluoroalkyl substances (PFAS) and aqueous film-forming foams (AFFFs). Ultra-fast degradation of short-chain PFAS, long-chain homologs, precursors, legacy PFAS, emerging PFAS was achieved in a matter of minutes. The concentrations of PFAS and the soil type had a negligible impact on degradation efficiency, possibly due to the ultra-fast degradation rate overwhelming potential differences. Under the current HFH experiment setup, we achieved near-complete degradation (e.g., >99.9%) after 1 min for perfluoroalkyl carboxylic acids and perfluoroalkyl ether carboxylic acids and 2 min for perfluoroalkanesulfonic acids. Polyfluoroalkyl precursors in AFFFs were found to degrade completely within 1 min of HFH; no residual cationic, zwitterionic, anionic, or non-ionic intermediate products were detected following the treatment. The gaseous byproducts were considered. Most of gaseous organofluorine products of PFAS at low-and-moderate temperatures disappeared when temperatures reached 890 °C, which is in the temperature zone of HFH. For the first time, we demonstrated minimal loss of PFAS in water during the boiling process, indicating a low risk of PFAS entering the atmosphere with the water vapor. The findings highlight HFH its potential as a promising remediation tool for PFAS-contaminated soils.

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1. Introduction

1.1. Soil contamination of per- and polyfluoroalkyl substances (PFAS)

PFAS are a broad group of synthetic chemicals known for their unique properties, including resistance to both water and oil and strong carbon—fluorine bonds [1–5]. Since their introduction in the 1940 s, PFAS have been used extensively in various applications: non-stick cookware, stain-resistant fabrics, firefighting foams, and food packaging [6,7]. Despite their widespread use and valuable properties, PFAS have emerged as a significant environmental concern due to their persistent, bioaccumulative, and potentially toxic nature [8–12].

There are two primary categories within PFAS: perfluoroalkyl substances, with perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) being the most studied, and polyfluoroalkyl counterparts. PFOA and PFOS have been found to cause adverse effects in laboratory animals, including developmental, reproductive, and liver toxicity [10, 13–15]. Epidemiological studies in humans have also suggested potential links between PFOA and PFOS exposure and various health effects, such as kidney and testicular cancer [16–18], thyroid disruption [19], and immunotoxicity [20]. In addition to perfluoroalkyl substances, numerous polyfluorinated species, or so-called *precursors*, have been detected in aqueous film-forming foams (AFFFs) [6, 21, 22].

On April 13, 2023, the US EPA started the process to regulate PFAS as hazardous substances under the Superfund Act [23], including seven PFAS besides PFOA and PFOS, [2] precursors to PFOA, PFOS, and seven other PFAS; and [3] categories of PFAS. This regulatory move underscores the growing emphasis on addressing PFAS-contaminated sites, particularly soil and groundwater.

PFAS contamination in soil can occur through several pathways, including direct release from industrial sites [24–28], landfills [29,30], biosolids [31,32], and firefighting training areas [33–35] where PFAS-containing AFFFs are used. Due to the use of AFFFs for fire training alone, more than 400 locations in the United States have been identified where known or suspected releases of PFAS to the soil have occurred [36]. Other sources include atmospheric deposition, wastewater irrigation, and the agricultural use of PFAS-containing biosolids [5, 37, 38]. Once in the soil, PFAS can persist for long periods due to their high stability and resistance to degradation. This persistence poses a risk to both human health and the environment, as PFAS can enter the food chain through the uptake by plants [39,40] or consumption of contaminated water [41,42]. Therefore, managing PFAS contamination in soil is crucial to prevent further exposure and ensure the safety of groundwater and food supply.

1.2. Current PFAS remediation methods and challenges

PFAS, distinguished by their carbon chains where some or all hydrogen atoms are replaced by fluorine, possess strong C–F bonds, granting them remarkable chemical stability and resistance to degradation. While perfluoroalkyl substances (e.g., PFOA and PFOS) contain fully fluorinated carbon chains, polyfluoroalkyl substances have carbon chains that are only partially fluorinated. This difference in chemistry ultimately affects their transport, persistence, and health effects. Under conditions like heating [43]{Xiao, 2023 #6681}, oxidation [44,45], and biological processes [46–48], polyfluoroalkyl substances can transform into their fully fluorinated variants, possibly elevating their toxicity.

Various technologies have been developed in the past 30 years to remediate organic-contaminated soils; however, not all these techniques are appropriate for addressing PFAS contamination in soil. One method, for example, is excavation and disposal at secure landfills or other containment facilities. This method involves physically removing and transporting the contaminated soil to a disposal facility, then replacing it with clean soil. While it effectively reduces onsite PFAS concentrations [41,49], it has limitations like high costs, potential secondary contamination during transit, and the problem of merely relocating the contamination.

An alternative is soil washing, where the soil undergoes treatment using water or solvents to separate organic compounds. The soil interacts with the washing solution, mobilizing PFAS into the liquid phase [37, 50, 51]. After separating the PFAS-containing liquid from the soil, the latter is returned to its origin, while the former undergoes further processing to remove the PFAS. Though effective in certain conditions [37,51], soil washing can be expensive and energy-demanding. It requires significant quantities of water or solvents, and subsequent treatment of the contaminated solution is necessary. Its efficacy can also be influenced by factors such as soil type, organic content, and the distribution of contaminants. Additionally, soil washing might not be effective in completely removing PFAS from soil, as some residual contamination may remain [51,52].

In situ immobilization is a widely researched soil remediation technique. It employs adsorbent materials, notably activated carbon, biochar, and clay minerals, to capture and secure contaminants within soil [37, 53–56]. These materials can be added directly to the contaminated soil or used in ex situ treatment systems, such as permeable reactive barriers or filtration units. While adsorption can effectively reduce the bioavailability of PFAS in the soil environment, it is generally considered a containment strategy rather than a degradation method. Moderately hydrophobic PFAS can potentially be released back into the environment. Additionally, adsorbent materials may require periodic replacement or regeneration after being saturated with PFAS. The effectiveness of the adsorption method can vary depending on factors such as the soil's composition, the type of PFAS, and prevailing environmental conditions, making it challenging to develop a one-size-fits-all solution.

Chemical oxidation techniques, such as supercritical water oxidation [57], electrochemical oxidation [58,59], and photocatalytic decontamination [45, 60, 61], can break down PFAS in water under specific conditions. However, these methods may be ineffective for degrading PFAS in soil without water, and they may require carefully controlled conditions to be effective.

Biological treatment methods, such as bioremediation or phytoremediation, involve using microorganisms or plants to degrade or immobilize PFAS in the soil. Some studies have reported the successful degradation of certain PFAS by certain microbial strains or consortia [47, 48, 62, 63], but the overall effectiveness of biological treatment for PFAS remains limited due to the recalcitrant nature of these compounds. Furthermore, biological treatments of polyfluorinated compounds may lead to the formation of more problematic perfluorinated species [64–66]. Phytoremediation, the use of plants for remediation, has shown potential in laboratory settings [9, 67–69], but its large-scale applicability and efficiency are yet to be conclusively demonstrated.

Previous reports have highlighted the nonlinear sorption of AFFF-related polyfluoroalkyl substances to soil, characterized by concave-down isotherms and irreversible (hysteretic) behaviors [70]. Consequently, PFAS molecules that are irreversibly bound can resist physical (e.g., washing), chemical, or biological treatments, resulting in prolonged periods needed to flush out PFAS plumes from an aquifer than initially anticipated [27, 70, 71]. For instance, numerous studies have shown that soil and groundwater contamination by PFAS persists for several years, and in some cases, decades, even after contamination activities have ceased [6, 27, 35, 72–76].

Thermal treatment methods, encompassing techniques like pyrolysis [77,78], thermal air oxidation [78], thermal desorption [55,79] and smoldering [80], subject contaminated soil to high temperatures to break down PFAS. These methods have shown promise, particularly against recalcitrant short-chain PFAS and perfluoroalkanesulfonic acids (PFSAs), such as PFOS. While thermal techniques can be effective for treating PFAS-contaminated soils [78–80], they are generally energy intensive. Alternative innovative thermal-related strategies, such as ultrasound remediation [81], gas fractionation [82], and ball milling [83], have also been explored. However, these methods necessitate additional research to enhance their effectiveness against specific types of PFAS.

The limitations of the existing methods for PFAS remediation in soil highlight the need for the development of innovative, energy-efficient, and sustainable solutions to address the challenges posed by PFAS contamination.

1.3. Introduction to high-frequency heating (HFH) as an alternative approach for remediation of PFAS in soil

An optimal approach to treating solid waste containing PFAS would be effective for a broad range of PFAS compounds while achieving rapid degradation. Additionally, the treatment method should be adaptable in rural or remote communities lacking centralized waste treatment facilities.

The aim of this study is to investigate the feasibility of using HFH for the remediation of soil contaminated by PFAS. As far as we know, this study represents the first instance of using HFH to remediate soils contaminated with PFAS. HFH employs electromagnetic fields to produce heat in conductive mediums like soil, eliminating the need for direct contact with the heat source. It operates on electromagnetic induction; an alternating current in a coil creates an oscillating magnetic field, inducing eddy currents in any conductive material within, generating heat [84]. HFH is a highly energy-efficient process, as heat is generated directly within the target material, minimizing heat loss to the surroundings. HFH sets itself apart from conventional slow heating methods by rapidly increasing the temperature, allowing it to pass quickly through the low to moderate temperature ranges where many fluorinated PFAS species are likely to be formed [77, 85, 86]. We hypothesize that HFH may offer a more efficient and effective remediation option for PFAS-contaminated soils than other remediation methods, particularly for soils contaminated with PFAS compounds.

To test this hypothesis, we conducted a series of HFH treatments of soil contaminated by a wide range of PFAS, including short-chain PFAS, perfluoroalkyl carboxylic acids (PFCAs), PFSAs, perfluoroalkyl ether carboxylic acids (PFECAs), and polyfluoroalkyl substances. In addition to legacy PFAS, emerging PFAS such as perfluoro-2,5-dimethyl-3,6-dioxanonanoic acid (HFPO-TA) [87,88] were included as well. In addition, we also included PFAS-containing AFFFs and surfactant concentrates containing various cationic, zwitterionic, and anionic polyfluorinated compounds in this study. To the best of our knowledge, this represents one of the most comprehensive studies on PFAS remediation, encompassing a wide array of PFAS classes and their presence in relevant commercial products. Ultimately, our goal is to provide a foundation for further research in this area and contribute to the development of a more sustainable and eco-friendly approach to PFAS-contaminated soil remediation.

Furthermore, in this study, we conducted the Monte Carlo estimation of the energy consumption by this innovative method in comparison with mainstream thermal remediation technologies.

Lastly, the potential loss of PFAS to water vapor is a potentially important factor to consider while studying their thermal degradation in moist soil. Because many PFAS are surfactants, they may attach or adsorb to water vapor or aerosols generated during the thermal treatment. Therefore, we also have assessed the possible loss of PFAS in water in boiling processes, which is pertinent to grasping their possible mobilization during the early stages of heating. Furthermore, this information is vital for effective and safe removal of PFAS from contaminated soil via thermal approaches. It also aids in estimating the potential PFAS inhalation risks that may occur due to their attachment to water vapor or aerosols in other relevant heating processes (e.g., cooking, firefighting, and baking).

2. Methods and materials

2.1. PFAS and HFH device

This study included six PFCAs (e.g., PFOA), three PFSAs (e.g., PFOS), and three PFECAs that have been produced as alternatives to PFOA and PFOS (Table S1 of the Supplementary document). These chemicals were purchased from Sigma—Aldrich. This study also included two 3 M AFFF samples (3% v/v) (#1 and #2) containing a mixture of anionic perfluorinated compounds and anionic, zwitterionic, and cationic polyfluoroalkyl substances (Table S2). Lastly, two “Fluorad” brand fluorosurfactant concentrates containing cationic, zwitterionic, and non-ionic polyfluoroalkyl substances (Tables S3 and S4) were included in the HFH experiment. The structures of PFAS can be found in the Supplementary document (Tables S1−S4).

The HFH device was a handheld induction-heating tool (Bolt Buster™) procured from LACE Technologies, Inc (Addison, IL, USA). The maximum operating time of this HFH device is 2 min. Stainless steel reactors (7 mL; 45 mm in height and 19 mm in outside diameter) with a stainless-steel screw lid were obtained from the QAQC Lab Inc. (White Stone, VA, USA) for HFH experiments. The reactor temperature during HFH was recorded using a Digi-Sense dual-laser infrared thermometer (Cole Parmer, IL, USA) in a continuous scan mode connected to a computer with an infrared thermometer software package (Fig. S1).

In our experiments, we examined two potential scenarios, namely Scenario #1 and Scenario #2, to demonstrate the impact of four variables. These variables encompass thermal attributes (i.e., HFH duration), characteristics of PFAS, the texture of the soil, and the concentration of PFAS within the soil.

2.2. Scenario #1: HFH of PFAS pre-adsorbed on soil

The pre-adsorption of perfluorinated chemicals (Table S1) in soil was performed in batch sorption experiments following the previous procedure [70]. The liquid phase was a landfill leachate sample provided by Waste Management Inc. No measurable PFAS (Table S1) were detected in microfiltered landfill leachate samples. The soil was a clay loam with an organic matter content of 9.8% and a cation-exchange capacity of 41.0 cmol/kg [46]. Leachate samples were spiked with PFAS (Table 1) to $\sim 2 \times 10^{-6}$ mol/L in the laboratory to facilitate detection. The apparent sorption equilibrium was reached after two days [89]. After sorption, the supernatant fluid was decanted. The remaining PFAS-laden soil particles were freeze-dried, stored in a desiccator to reach room temperature, and thermally treated in a sealed steel reactor by HFH.

2.3. Scenario #2: HFH experiments of AFFF and surfactant concentrate in soil

In the case of the second scenario, we conducted experiments by directly integrating PFAS substances or AFFF with soil particles. This was done to emulate the unrestricted presence of PFAS molecules in the soil and those molecules that have only a weak association with soil particles. In brief, AFFF or surfactant methanol stock solutions were prepared by adding 80 μL AFFF or surfactant concentrate solution into 200 mL of HPLC-grade methanol (Thermo Fisher Scientific, Pittsburgh, USA). An aliquot (1 mL) of the AFFF or surfactant concentrate methanol stock solution was added to the stainless-steel reactor (without a lid) and dried in a forced-air oven at 25 °C. Then, a known amount (0.1 g) of dry natural soil was added to the reactor, which was then screwed tight using clamps and heated by the induction heater for 1 or 2 min

In addition to the natural soil (a clay loam), this study also included a reference clay (KGa-1b kaolinite) purchased form the Clay Minerals Society (GA, USA) and the Pahokee peat, a high-organic (56% organic carbon) reference material (IHSS; St. Paul, MN).

Table 1
Comparison of conventional heating and HFH for decomposition of PFAS.

	Conventional heating (slow heating)	HFH (fast heating)
Temperature (°C) at which PFAS degradation was observed	150 – 900 (PFOA and HFPO-DA) [78, 85, 86, 90, 92] 450 – 900 (PFOS) [78, 86, 92]	500 – 845
Typical heating rate	10°C/min	16°C/sec
Residence time	Minutes to hours	Seconds
Reactors and heating method	Furnace or oven Heat is transferred through conduction, convection, and radiation	Metallic reactor Heat is generated within the metallic reactor by electromagnetic induction
Advantages	A mature heating technology Relatively easy to scale up [105]	Rapid heating High heating efficiency (80 –90%) [98] High energy conversion efficiency Precise temperature control and fast heat-up times HFH reactors are easy to install and maintain
Disadvantages	Low heating efficiency (e. g., 20 –40% for coil heaters [98]), and thus requiring a longer heating time to achieve PFAS degradation High energy consumption Long startup, processing, and cooling times Generation of various fluorinated intermediates at low to moderate temperatures [77]	Corrosion of the metallic reactor An emerging PFAS treatment technology, which lacks generalized information

2.4. *Possible PFAS mass loss due to water evaporation*

We also carried out an investigation into the potential loss of PFAS mass during the process of water boiling. Our setup consisted of a 500-mL round-bottom evaporation flask, on a heater, containing 100 mL of distilled water spiked with a PFAS mixture. To determine the initial PFAS concentration prior to heating, we collected three samples from this solution. This flask was then linked to a 250 mL round-bottom flask via a distillation column condenser, designed to collect the evaporated solution. The column condenser was maintained at a low temperature by circulating cold tap water through it, which facilitated the cooling and condensation of vapors originating from the evaporation flask. The solution was allowed to boil for 25 min, after which we collected three samples each from both the evaporation and receiving flasks, once the solutions had sufficiently cooled.

2.5. *Thermal desorption—pyrolysis—gas chromatography—MS (TD—Pyr—GC—MS) experiments*

Our team has expended considerable effort in profiling the gaseous emissions from PFAS and their heat-induced degradation by-products. We have accumulated a substantial amount of data, including findings that have yet to be published. In one study, we explored the gaseous products resulting from the thermal decomposition of AFFF samples (Tables S2) [77] [77]. This very same system has been deployed in earlier research efforts, where it was used to examine the gaseous by-products formed during the thermal treatment of long-chain [78] and short-chain [90] PFAS. For this study, we utilized unpublished findings from our prior work [77] to shed light on the gaseous emissions resulting from the rapid heating of AFFF samples. The analyses were conducted by using a Frontier 3030D thermal desorption-pyrolysis system (Frontier Labs Inc., Japan) coupled with a gas chromatography–mass spectrometry (GC–MS) system (Agilent GC 7890 and 5975 C MS; Santa Clara, CA). We utilized a Frontier 30-m Ultra Alloy capillary column from Frontier Labs Inc. (Japan), which had an inner diameter of 0.25 mm and was equipped with a 5% diphenyldimethyl polysiloxane stationary phase with a 0.25 µm film thickness. The MS analysis was carried out using electron ionization, surveying the mass range of 35–850 *m/z*. Ultra-pure helium with a purity level of 99.999% served as the carrier gas, employed at a constant flow rate of 1.1 mL/min. In the experiment, an AFFF sample was subjected to heating at 400 °C/min (or 6.7 °C/sec), starting from 50 °C and escalating to various temperatures [77]. The data processing stage incorporated the evaluation of the MS fragmentation pattern of potential pyrolyzates, leveraging the extensive 2005 National Institute of Standards and Technology library, which houses 190,825 spectra. Any compounds were considered provisionally identified if they demonstrated a library match quality exceeding 70% [77].

2.6. *Energy consumption estimation using Monte Carlo simulation*

Consider a reactor with a total surface area (A) of 600 cm^2 containing 100 g GAC (density: 700 kg/m^3) through which the heat is being conducted. We estimated the energy consumed by heating this reactor using Monte Carlo simulations that consider all possible combinations of key independent variables [27,91]. Three thermal treatment approaches were compared for achieving 99% degradation/removal of PFAS from solid materials, including HFH (T = 500–845 °C; t = 20–120 s) (this study), thermal desorption (T = 200–500 °C; t = 5–20 d) [79], and smoldering (T = 600–1100 °C; t = 1–24 h) [80]. The heat transfer rate (Φ) caused by heat convection was calculated using the Newton's law of cooling:

$$\Phi_{\text{convection}} = hA\Delta T \tag{1}$$

where Φ is the heat transfer rate (W); h is the surface heat transfer coefficient (3.42 $W/m^2{\cdot}K$ for air and estimated at 0.25 $W/m^2{\cdot}K$ for soil); ΔT is the temperature difference (K). The transfer rate of heat radiation was estimated by the Stefan Boltzmann equation:

$$\Phi_{\text{radiation}} = \varepsilon_1 A\sigma\left(T_1^4 - T_2^4\right) \tag{2}$$

where ε_1 is the emissivity (0.9); σ is Stefan Boltzmann constant (5.67×10^{-8} $W/m^2{\cdot}K^{-4}$); T_1 and T_2 are temperatures (K) of the reactor and the ambient air, respectively.

Combining Eqs. 1 and 2, we can estimate the total heat consumption (E, kW·h) using Monte Carlo simulations:

$$E = \left\{hAF(\Delta T) + \varepsilon_1 A\sigma\left[\left(F(T_1)\right)^4 - T_2^4\right]\right\} \times F(t)/1000 \tag{3}$$

where F represents the frequency in Monte Carlo simulations, and t is the treatment time. The peak of this frequency distribution curve shows the most probable value of E within the ranges of T_1 and t.

3. Results and discussion

3.1. *Degradation efficiency of PFAS of various chain lengths and functional groups in soil under HFH*

Fig. 1 illustrates the degradation of soil-borne PFCAs with varying chain lengths subjected to HFH in soil. The degradation curves exhibited an overall inverted L-shape, indicating that degradation efficiency increased with HFH time. For instance, PFOA degradation was 45.16% after 15 s of HFH, while this efficiency escalated to 99.91% when the heating time was extended to 60 s. The PFAS chain length did not significantly impact the degradation. This observation is exemplified by the comparable degradation efficiencies of PFBA and PFOA, which is also mirrored in PFAS degradation during heating at a regular rate [78, 85, 92].

The type of PFAS, including their functional groups, affects the degradation efficiency under HFH. The degradation of PFSAs was

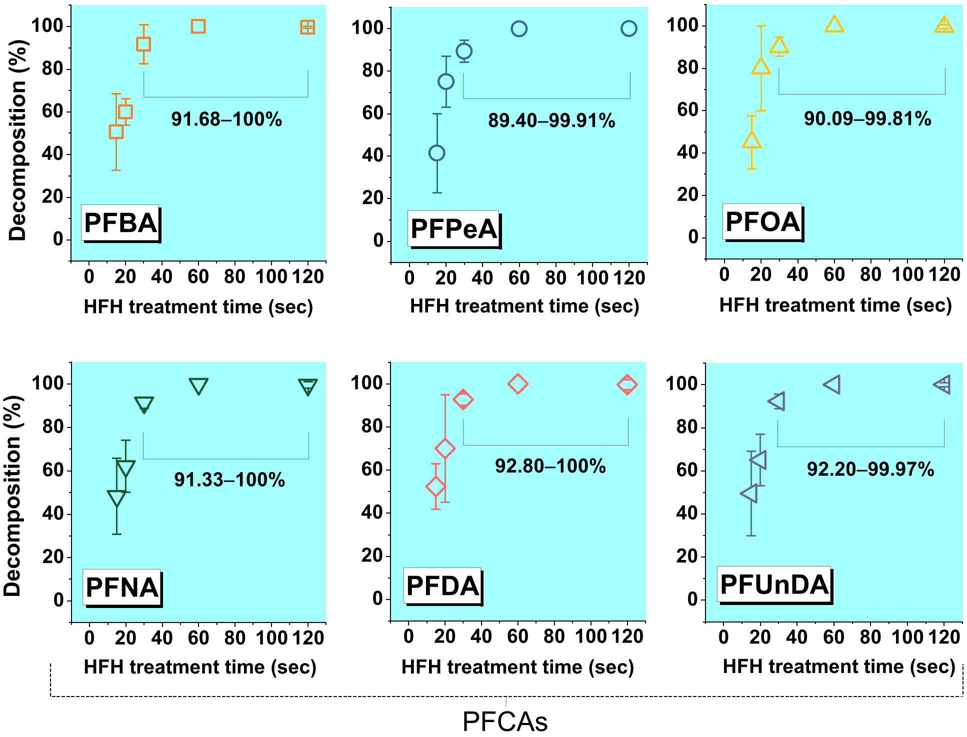

Fig. 1. HFH (up to 2 min) of a mixture of PFCAs added to natural soil (0.2 g) in a sealed steel reactor. The initial mass of each PFCA was 0.02 μmol. The decomposition efficiency was assigned to 100% if no measurable PFCA was found after the thermal treatment. Note that the actual PFCA heating time is longer than the HFH time (e.g., 20 s) as it took approximately 65 s to cool down from ~845 °C to room temperature.

markedly lower than that of PFCAs, particularly in instances of ultra-short treatment time (Figs. 1 and 2). For example, after 30 s of HFH, PFBS degradation reached 37.82% (Fig. 2), while PFBA, its corresponding PFCA counterpart, achieved a 91.68% degradation rate (Fig. 1). Conversely, PFECA degradation appeared more efficient than PFCA degradation (Figs. 1 and 2). After 15 s of HFH, 57% of HFPO-DA had already degraded.

Temperature plays a critical role in the degradation of PFAS under thermal conditions. While PFOA degradation can occur at low temperatures (150 –200 °C) with the presence of granular activated carbon

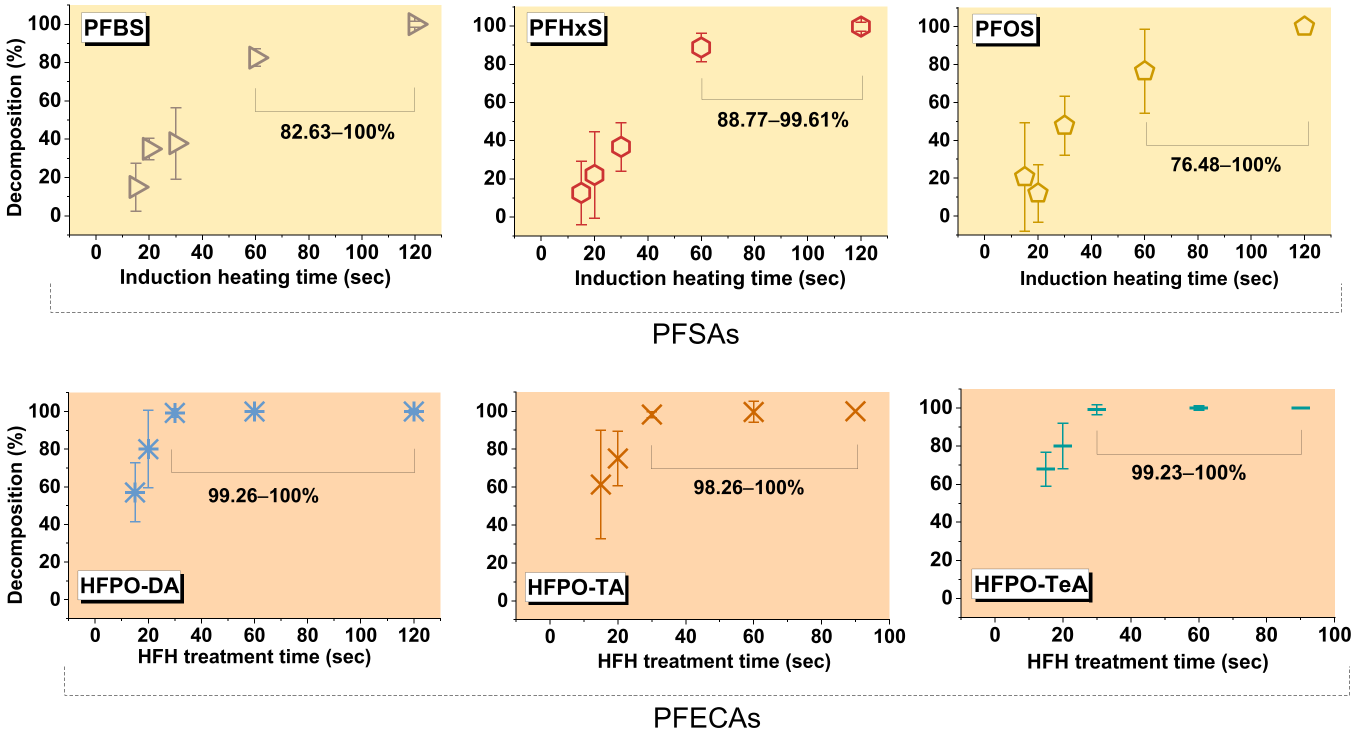

Fig. 2. HFH (up to 2 min) of a mixture of PFSAs and PFECAs added to 0.2 g of natural soil in a sealed steel reactor. The initial mass of each PFAS was 0.02 μmol. The decomposition efficiency was assigned to 100% if no measurable PFAS was found after the thermal treatment. Note that the actual PFAS heating time is longer than the HFH time (e.g., 20 s) as it took approximately 65 s to cool down from ~845 °C to room temperature.

[85], a much higher temperature (>700 °C) would be needed to achieve a mineralization rate of < 80% [92]. In this study, the steel reactor temperature rapidly increased to approximately 500 °C within 30 s at a heating rate of 14.5 °C/sec, and subsequently rose to approximately 845 °C after 1 min (Table S5). The infrared thermometer considerably underreported the temperature of steel, a low-emissivity material (Table S5). The high temperatures induced by the HFH device were the critical factor in achieving the complete degradation of PFAS in such a short period of time (Fig. 1).

Treatment time apparently is an important factor influencing the degradation of PFAS in soil under HFH. Longer treatment times can allow for more complete degradation of PFAS. Based on the aforementioned discussion, it appears that 1 min of HFH is adequate for achieving near-complete degradation (approximately 100%) of PFCAs and HFPO-DA. For PFSAs, 2 min of HFH treatment is necessary to attain a similar degradation rate. Following 2-min HFH, the degradation of all PFAS in soil neared 100% (Figs. 1 and 2).

3.2. Effects of contamination level and soil type on degradation of PFAS in soil by HFH

The contamination level of PFAS in the soil is another potentially important factor affecting the efficiency of chemical and biological treatments. Fig. 3a and b display the degradation of PFAS adsorbed onto soil at varying concentrations subjected to HFH. The figures employ a logarithmic scale for the horizontal axis, as the concentrations of PFAS in the soil span two orders of magnitude. As depicted in the figure, PFAS in the soil exhibited high degradation rates at both low and high concentrations following 1 or 2 min of HFH treatment.

Furthermore, the type of soil in which PFAS contamination occurs can significantly impact the degradation efficiency under HFH. Factors such as organic matter content and mineralogy can influence the heating characteristics, heat distribution, and the interaction between PFAS and soil components. For example, soils with higher clay content may require more energy to heat due to their low thermal conductivity [93, 94], while soils with high organic matter content can potentially enhance the degradation of organic compounds by facilitating the formation of reactive species [95–97]. The presence of certain minerals, such as iron or other metal oxides, in soil has also been reported to enhance PFAS degradation, possibly due to their catalytic activity at high temperatures [98].

Our study found that soil type had a negligible effect on the degradation of PFAS compounds during HFH. The degradation rate of PFAS in different soils approached 100% (Fig. 3c). This is likely because the ultra-fast degradation rate of PFAS under high temperatures (as induced by the HFH device) overwhelmed any potential differences in soil properties. Specifically, our results demonstrated that PFAS compounds were rapidly degraded within a matter of minutes in all three types of soils with different amounts of minerals and organic matter (as shown in Fig. 3). Therefore, we conclude that soil type does not play a significant role in the efficacy of HFH as a remediation technique for PFAS-contaminated soils.

3.3. Thermal decomposition of polyfluoroalkyl substances

In addition to perfluoroalkyl substances, numerous structurally similar compounds known as polyfluoroalkyl substances have been identified, which contain a non-fluorinated branched chain [99–101]. We also investigated the degradation of AFFF and polyfluoroalkyl substances in surfactant concentrates during HFH. Our previous study revealed that polyfluoroalkyl substances are more susceptible to pyrolysis than perfluoroalkyl substances [85,92]. In this paper, we discovered that 1 min of HFH resulted in complete degradation (100%) of polyfluoroalkyl substances (Fig. 4). After 1 min, no residual cationic, zwitterionic, anionic, and non-ionic polyfluoroalkyl substances (Tables S1–S4) were detected. This finding further underscores the effectiveness of HFH in treating PFAS-contaminated soil.

When exposed to low to moderate heat, polyfluoroalkyl substances—or precursors—within these AFFF samples can convert into perfluorinated compounds [86]. The generation of intermediate products from precursors follows a bell-shaped curve; they are produced at low to moderate heat levels and then degrade when subjected to higher temperatures [86]. In this study, there were no detected perfluorinated transformation products after subjecting the polyfluoroalkyl substances

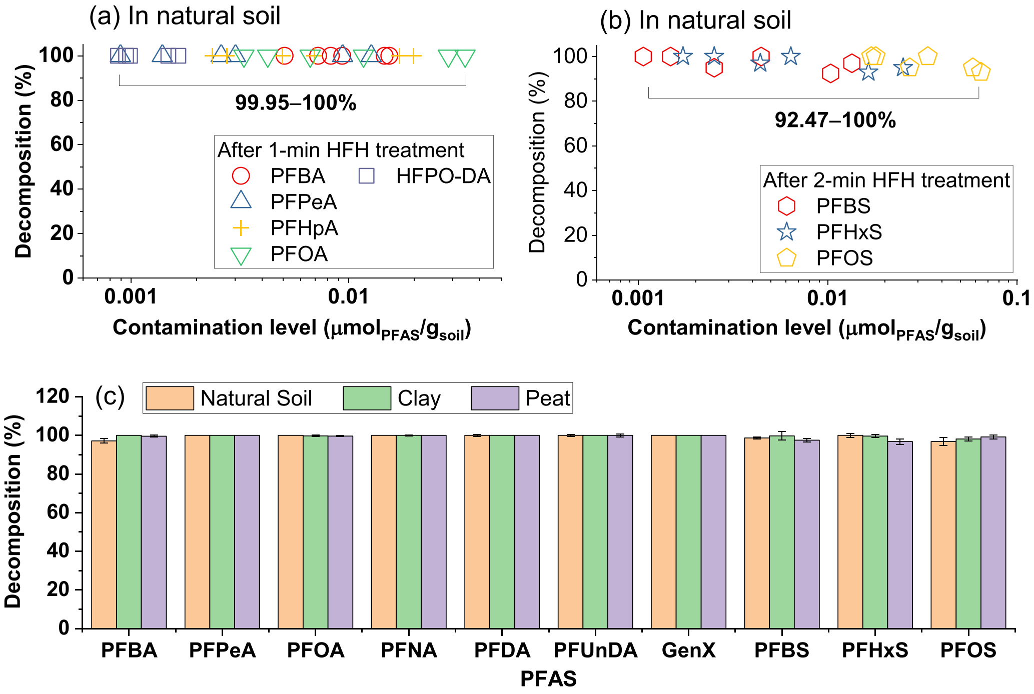

Fig. 3. (a) and (b): Degradation of PFAS preadsorbed to natural soil by 1 or 2-min HFH. (c): 2-min HFH treatments of PFAS (0.02 μmol) added to different types of soils (0.2 g).

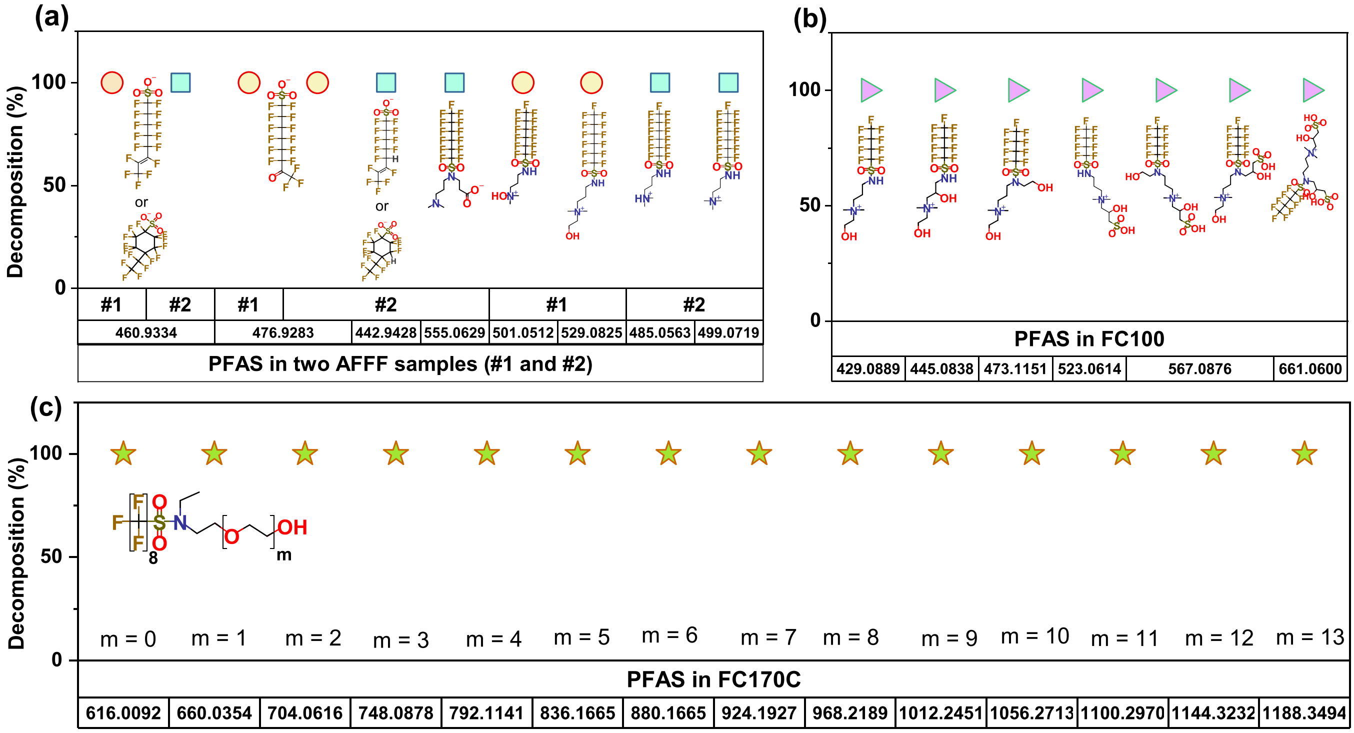

Fig. 4. HFH (1 min) of *poly*fluoroalkyl substances present in AFFFs (#1 and #2) and surfactant concentrates (FC-100 and FC-170 C) with the presence of natural soil (0.1 g) in a sealed reactor. The decomposition efficiency was assigned to 100% if no measurable PFAS was found after the thermal treatment. The numbers (e.g., 460.9334) are the *m/z* values of these polyfluoroalkyl substances (e.g., Table S3).

in AFFFs to HFH for 1 min. This implies a swift degradation of the precursors and intermediate products when formed at low and moderate temperatures [86].

3.4. Gaseous degradation products

It is also important to consider the potential formation of gaseous products from PFAS under thermal conditions. Careful temperature control can help minimize the risk of generating harmful gaseous products, promoting PFAS mineralization. When heated at low and moderate temperatures and in the absence of granular activated carbon, a significant portion of the thermal degradation products of PFAS consists of gaseous compounds [77, 92, 102–104]. One potential advantage of HFH is its ability to rapidly increase the temperature, allowing it to pass quickly through the low to moderate temperature range where many fluorinated PFAS species are likely to be generated [77, 92, 102].

As depicted in Fig. 5, AFFFs generated numerous volatile pyrolysis products at temperatures ranging from 300 – 500 °C. When the temperature increased to 890 °C, most of these gaseous products vanished, indicating their decomposition at elevated temperatures. The overall product distribution exhibited a bell-shaped curve with respect to temperature. Due to the absence of standards and effective detection methods, the quantities of these products were characterized by their MS peak intensities.

3.5. Loss of PFAS in water during water boiling

Understanding the behavior of PFAS during thermal soil remediation processes is of significant importance, particularly considering that soils contain varied moisture levels. As surfactants, PFAS could potentially adhere to water vapor or aerosols produced during thermal treatment. However, our recent investigations have indicated minimal PFAS adsorption to water vapor during the boiling process (Fig. 6). No PFAS were detected in the solution of the receiving flask. Except for PFDA, the loss of the studied PFAS in the evaporation flask solution was less than 0.5 mol% after boiling water for 25 min (Fig. 6). This indicates a minimal rate of PFAS loss during the water boiling process. These findings hold significance as they offer the initial understanding of the potential reduction of PFAS in water vapor.

3.6. Key advantages of HFH

Thermal remediation techniques, such as thermal desorption [79] and smoldering [80], have shown promise for the treatment of PFAS-contaminated soil. Thermal desorption at low and moderate temperatures is a widely used remediation method that involves heating contaminated soil to evaporate and separate volatile contaminants, which are then collected and treated in an off-gas treatment system. The goal of thermal desorption is not to decompose the contaminants, but rather mobilize them from soil. Smoldering is a thermal technology used for soil remediation, which employs low-temperature, self-sustaining combustion processes to degrade, immobilize, or remove contaminants. Through harnessing the inherent heat of reaction, this technique effectively treats a wide range of organic pollutants. However, their energy efficiency, environmental impact, and overall performance can differ significantly.

We compared the energy consumption in the HFH process to thermal desorption and smoldering. As illustrated in Fig. 7, while thermal desorption can be effective in treating a range of volatile and semi-volatile organics, its energy efficiency is low; the heat loss to the surroundings can be significant, especially in large-scale or in situ applications, leading to higher energy consumption (Fig. 7). Smoldering can be energy-efficient in comparison with thermal desorption, as the heat generated by the combustion process is used to sustain the reaction and treat the soil (Fig. 7). As illustrated in Fig. 7, HFH is a much more energy-efficient method for the remediation of PFAS-contaminated soils than thermal desorption and smoldering. The heat consumption by HFH is markedly lower, by several orders of magnitude, than that of thermal desorption and smoldering. These results suggest that HFH has the potential to become a preferred method for energy-efficient remediation of

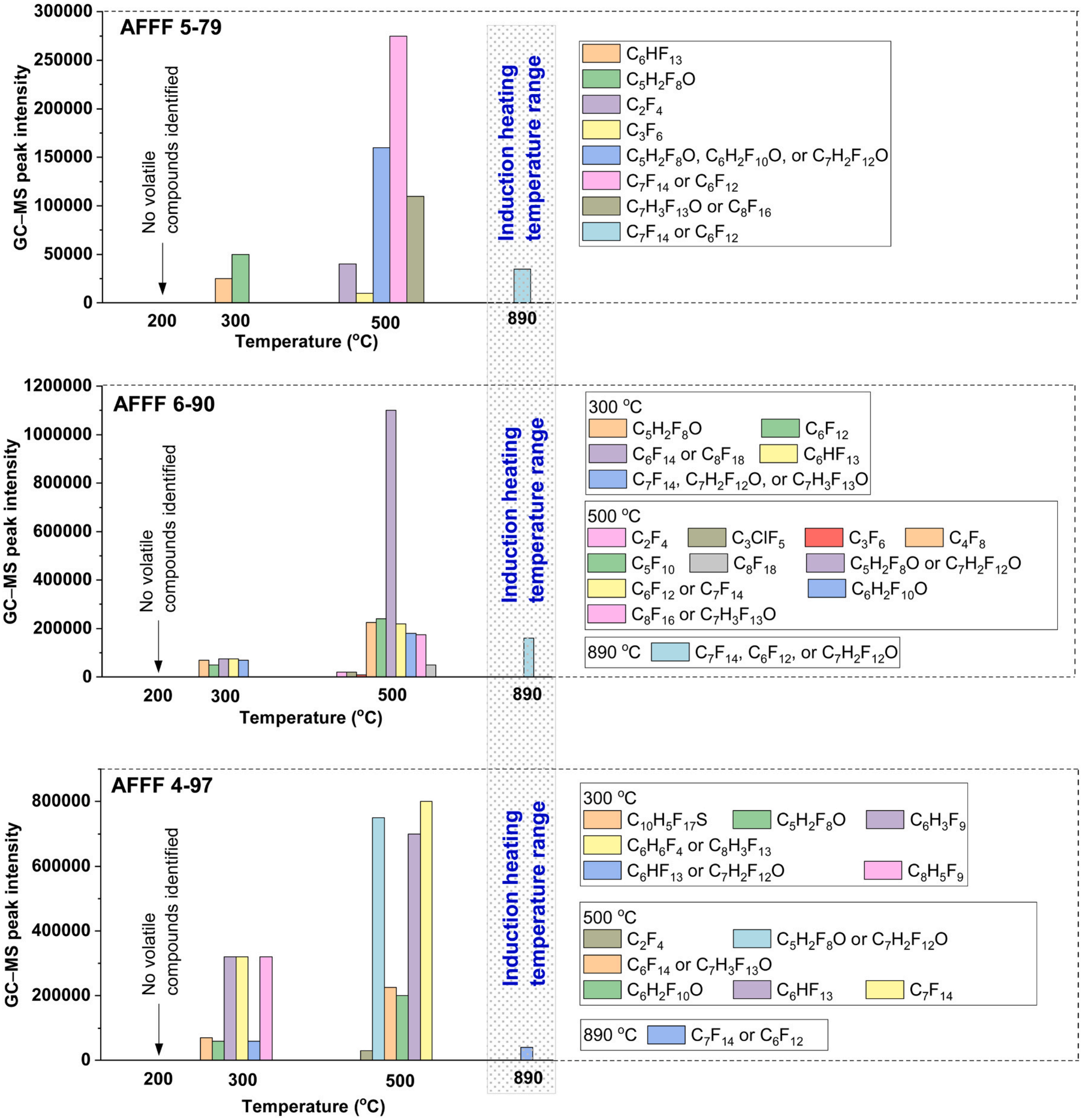

Fig. 5. Formation of fluorinated species from PFAS in three AFFF samples, including AFFF#1 (i.e., AFFF 5–79), when subjected to fast heating rates (6.7 °C/sec) from 50 °C to varying temperatures. PFAS identified in these AFFF samples can be found in a previous study [86].

organic-contaminated soils.

In addition to the low energy consumption, HFH allows for rapid and uniform heating of soil matrices in contrast to conventional thermal methods (e.g., thermal desorption, ovens and furnaces) (Table 1). This not only results in efficient removal and degradation of PFAS in soil (Figs. 1–4), but also reduces the risk of harmful gaseous products formed at low and moderate temperatures (Fig. 5). Additionally, the non-contact nature of induction heating reduces equipment wear, providing enhanced control over the remediation parameters (Table 1).

3.7. Potential Challenges and Limitations of HFH

Although HFH has demonstrated potential as an efficient and effective method for remediating PFAS-contaminated soil, there are several challenges and limitations that need to be addressed for its successful large-scale application (Table 1).

First, the scale-up of HFH technology from laboratory-scale to field-scale applications can be challenging due to the complexity of soil properties, contamination levels, and spatial heterogeneity at larger scales. Effective large-scale application of HFH requires careful consideration of factors such as heat distribution, temperature control, and treatment time to ensure uniform and efficient PFAS degradation across the contaminated area. Additionally, the design of the HFH system, including the choice of coil configuration and power supply, needs to be optimized for field-scale applications to maximize efficiency and minimize energy consumption.

Furthermore, the potential environmental impact of HFH, such as the formation of harmful gaseous products from PFAS, needs to be carefully assessed and managed for large-scale applications. The release of reactive F species (e.g., F radicals and HF) from PFAS can lead to corrosion in the steel reactor, which can be costly to repair and limit its large-scale application. Although not tested in this study, our previous research indicates that the addition of kaolinite is effective in quenching reactive F species released from PFAS upon heating [78]. Careful temperature control and monitoring, as well as the development of appropriate off-gas treatment systems, can help mitigate these environmental concerns.

Lastly, like several intensive remediation techniques, thermal

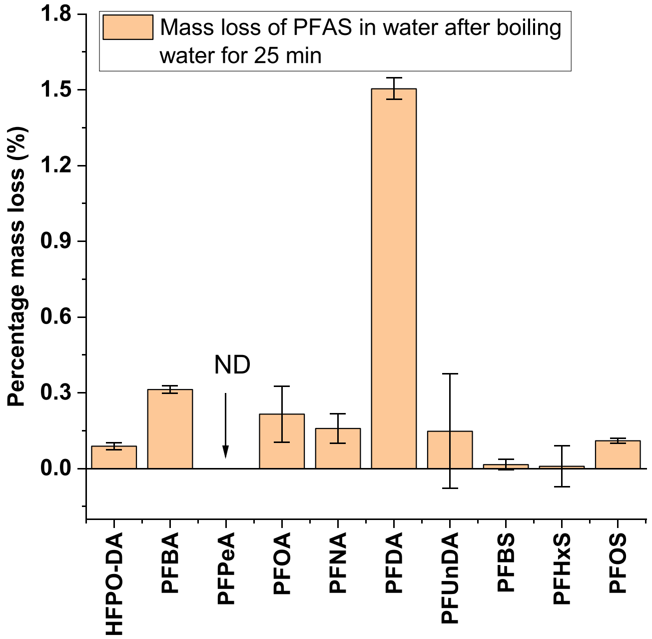

Fig. 6. Mass loss of PFAS from water after boiling the water at 100 °C for 25 min.

treatment has its drawbacks. Among these, one notable limitation is its potential to affect soil texture adversely. The short heating duration associated with high-frequency heating might offer a slight reprieve, potentially causing less harm to the soil texture than other prolonged thermal procedures or aggressive chemical and physical treatments.

3.8. Mechanisms of PFAS degradation by HFH

Understanding the mechanisms of PFAS degradation by HFH is critical for optimizing the remediation process and developing efficient strategies for treating PFAS-contaminated soil. The mechanisms of PFAS degradation by HFH are potentially complex and can involve a combination of thermal degradation and reactive species formation. Xiao and co-workers believed that PFAS can be thermally decomposed through initial, chain propagation, termination, and recombination mechanisms [78, 85, 86, 90, 92, 106]. The extent of thermal degradation is strongly influenced by the temperature, with higher temperatures generally leading to more efficient PFAS degradation. HFH can rapidly generate high temperatures within the soil, which can overcome the thermal stability of PFAS and promote their degradation.

Another potential mechanism of PFAS thermal degradation, which has been overlooked in the literature, is the formation of reactive species, such as radicals or reactive species, which can attack and break the carbon-fluorine bonds in PFAS. High temperatures generated by HFH can promote the formation of reactive species, either through the thermal decomposition of soil components, such as organic matter, or through C—F bond dissociation releasing reactive F species from PFAS at elevated temperatures [78, 85, 90]. F radicals are highly reactive, and the presence of these reactive species can contribute to the fast PFAS degradation as observed during HFH.

These mechanisms can be influenced by factors such as temperature, soil composition, and PFAS type, highlighting the need for a comprehensive understanding of these factors to optimize the HFH process and develop efficient remediation strategies. Further research is needed to elucidate the detailed mechanisms of PFAS degradation by HFH and to identify the optimal conditions for achieving efficient and environmentally friendly remediation of PFAS-contaminated soil.

4. Conclusions

PFAS contamination in soil poses significant risks to human health and the environment due to their persistence, bioaccumulative nature, and potential toxicity. Existing remediation methods for PFAS-contaminated soil include excavation and disposal in secure landfills, soil washing, and adsorption onto activated carbon or other materials. However, these techniques often have limitations in terms of efficiency, cost, and environmental impact. Therefore, there is a growing need for innovative and sustainable solutions to address PFAS contamination in soil. HFH is a highly energy-efficient thermal remediation method, as it generates heat directly within the target material, minimizing heat loss to the surroundings. The use of electromagnetic fields to generate heat within the soil allows for rapid and uniform heating, which can contribute to shorter treatment times and lower overall energy consumption. In addition, the data of this study showed varying degradation rates for PFAS in different classes during HFH. For example, the degradation of PFAS varies in the following order: PFECAs > PFCAs > PFSAs. This observation may be attributed to the differences in thermal stability and the energy required to break the O–F, C–F, and S–F bonds in PFECAs, PFCAs, and PFSAs. HFH can be tailored to target specific PFAS class by adjusting the treatment time (Fig. 1), potentially reducing the energy requirements of the process. These features make HFH a viable alternative for PFAS-contaminated soil remediation and are expected to attract increasing attention from researchers and practitioners in the field. Furthermore, this study pioneers in providing the first exploration into the potential reduction of PFAS in water vapor, marking its critical importance in this field of research.

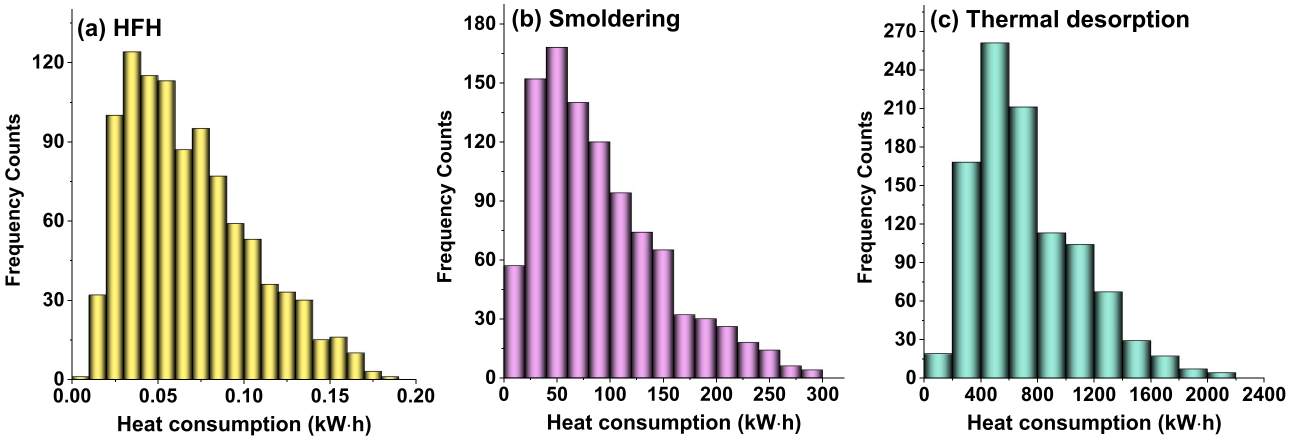

Fig. 7. Estimation of heat consumption by means of Monte Carlo simulation for three thermal treatment processes (HFH, thermal desorption, and smoldering) for removing PFAS from soil.

When comparing HFH with other thermal remediation methods, such as thermal desorption and smoldering, in terms of energy efficiency, HFH appears to offer several advantages. Its ability to generate heat directly within the target material, rapidly achieve high temperatures, and selectively target specific PFAS contributes to its energy-efficient nature. While each thermal remediation method has its merits and limitations, HFH may present a more energy-efficient and environmentally friendly alternative for the remediation of PFAS-contaminated soil. Further research and development are needed to optimize and scale-up HFH technology for the successful treatment of PFAS contamination and to fully realize its potential as a sustainable and cost-effective remediation solution.

The development and application of HFH for PFAS remediation have significant implications for the field. As an energy-efficient and potentially more cost-effective method, HFH can address some of the limitations of existing remediation technologies. Its ability to rapidly generate high temperatures and target specific contaminants offers the possibility of more efficient and environmentally friendly PFAS degradation. By focusing on these key areas of research and development, the environmental community can work to advance the field of HFH for PFAS remediation and contribute to the development of more sustainable and effective solutions for managing PFAS-contaminated sites globally.

Environmental implication

PFAS contamination in soil is a major environmental concern due to their persistent, bioaccumulative nature, and potential toxicity. This study introduces high-frequency heating (HFH), an innovative thermal remediation method, as a more sustainable and energy-efficient solution. With its ability to rapidly degrade PFAS in soil irrespective of their concentrations, HFH emerges as a potentially cost-effective alternative to current remediation methods. The widespread adoption of HFH can significantly alleviate the environmental and health risks associated with PFAS contamination. Ongoing research into optimizing HFH could revolutionize this approach to managing PFAS-contaminated sites, promoting more effective and sustainable soil remediation strategies.

CRediT authorship contribution statement

Runze Sun: Methodology, Visualization, Validation, Writing – original draft preparation. **Samuel Babalol**: Writing – review & editing. **Ruichong Ni**: Methodology, Visualization. **Alireza Arhami Dolatabad**: Writing – review & editing. **Jiefei Cao**: Writing – review & editing. **Feng Xiao**: Conceptualization, Methodology, Visualization, Writing – review & editing.

Declaration of Competing Interest

The authors have disclosed the HFH or induction heating technique as an invention to the University of Missouri.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.132660.

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