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## Environmental Significance

PFAS are persistent contaminants increasingly detected in soils, yet urban community garden soils—where direct human exposure can occur—remain poorly studied. Understanding PFAS distribution in these systems is critical because gardeners and consumers may be exposed through soil contact and consumption of produce. This study reveals that community garden soils can contain higher PFAS concentrations than nearby agricultural soils, with PFAS strongly retained in surface layers due to soil organic matter composition, dissolved organic matter fractions, and cation exchange capacity. By demonstrating matrix-specific and depth-dependent PFAS behavior, this work advances predictive understanding of PFAS fate in soils and informs exposure assessment, soil management practices, and mitigation strategies for urban and agricultural environments.



## Depth Profiles of PFAS in Community Garden and Agricultural Soils

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### ABSTRACT

Despite growing evidence of PFAS contamination in agricultural soils, urban community gardens have received little attention. However, these gardens may receive PFAS through various inputs and amendments, depending on local site conditions, surrounding land use, and environmental exposure pathways. In this study, thirteen out of forty targeted PFAS, representing three PFAS classes, were detected in a community garden soil, with a total PFAS concentration of  $3,987 \pm 146$  ng kg<sup>-1</sup>. Interestingly, total PFAS in the community garden soil was 2.6 times of the investigated agricultural soils ( $1,523 \pm 25$  ng kg<sup>-1</sup>). The dominant PFAS classes, PFCAs and PFSAs, were highest in the surface soil (0–5 cm) and decreased with increasing depth in both soils. This distribution pattern varied among PFAS classes and was influenced by soil physicochemical properties, including dissolved organic matter characteristics, humic- and



fulvic-like components, and cation exchange capacity. Furthermore, PFAS distribution patterns in soils, when compared with those in compost and landfill organics from our previous studies, exhibited distinct, matrix-specific depth profiles. Distinct distribution patterns of PFAS may offer useful insights to guide risk assessment and develop mitigation strategies in future studies.

**Keywords:** Garden soil; Vertical distribution; Organic matter; DOM components; Cation bridging.

## 1. INTRODUCTION

The persistent presence of PFAS in the environment is alarming due to their adverse effects on humans, animals, and other living organisms [1, 2]. Among various environmental matrices, soil functions as a major sink for PFAS [3, 4]. Agricultural soils can become contaminated with PFAS through multiple pathways, including the application of biosolids and compost, surface runoff from contaminated sites, irrigation with treated wastewater, use of fluorinated agrochemicals, and atmospheric deposition [5, 6]. For instance, agricultural lands amended with municipal biosolids for over ten years were found to contain 320–920  $\mu\text{g kg}^{-1}$  of perfluoroalkyl acids (PFAAs) [7]. It has been well studied that both legacy PFAS, such as perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), and emerging PFAS, including perfluorobutanoic acid (PFBA), perfluorohexanoic acid (PFHxA), and fluorotelomers are persistent in soil [8, 9]. PFAS can alter microbial communities and potentially affecting soil fertility and nutrient cycling [10, 11]. They can be taken up by plants, leading to bioaccumulation in crops and vegetables [12, 13]. As a result, PFAS can enter the food chain, posing risks to livestock and humans through dietary exposure [14, 15]. Furthermore, PFAS-contaminated soil can act as long-term sources of groundwater pollution, especially in regions with high rainfall or intensive irrigation, leading to long-term contamination of water resources [16, 17].

The fate and transport of PFAS in soil is strongly influenced by PFAS chemical properties and the physicochemical characteristics of soil. PFAS can sorb to organic matter, clay minerals, and metal oxides through hydrophobic interactions, electrostatic attraction, and hydrogen bonding, which control their vertical movement [18-21]. Soil organic matter (SOM) has been widely recognized as a major factor controlling PFAS retention at topsoil, yet the role of dissolved organic matter (DOM) components in influencing PFAS transport and accumulation in soil is still evolving [22, 23]. Additionally, most field-scale studies have focused on agricultural soils, where intensive practices such as repeated biosolid applications, uncontrolled surface runoff, and agrochemical use increase the likelihood of PFAS accumulation [21, 24-26]. In contrast, soils in urban settings, such as community gardens, have received little attention, despite their potential for human exposure through fresh produce. Community gardens have become common, with over 29,000 such gardens across the United States in 2024, engaging millions of people nationwide through shared urban green spaces [27, 28].



Previous studies have reported elevated levels of heavy metals in U.S. urban community garden soils, including cadmium (0.5–11 mg kg<sup>-1</sup>) and lead (102–950 mg kg<sup>-1</sup>), primarily resulting from soil amendments and surface runoff [29, 30]. In addition, soils from New York City community gardens contained polycyclic aromatic hydrocarbons (PAHs) ranging from 6 to 159.2 mg kg<sup>-1</sup>, accumulated due to atmospheric deposition and road runoff [31]. Like heavy metals and PAHs, PFAS can accumulate in community garden soils via different pathways. PFAS fate can also be influenced by gardening practices such as mixing, tilling, planting, and the soil's physicochemical properties. PFAS accumulation and transport processes highlight the need for effective management approaches that can reduce PFAS mobility and potential exposure in garden soils. PFAS mitigation strategies such as soil washing, stabilization, excavation, and sorbent-based immobilization are used to reduce PFAS mobility and exposure [32, 33]. Among these approaches, sorbent-based immobilization has gained attention, especially using metal–organic frameworks (MOFs) and covalent organic frameworks (COFs) because of their tunable pore structures and surface functionalities [32, 33]. Thus, understanding the factors influencing PFAS persistence and distribution in community garden soils is critical for assessing exposure risks and developing effective management strategies.

To address these concerns, we investigated forty PFAS from seven different PFAS classes in a community garden soil and their vertical distribution across soil depths. For comparison, the same PFAS were analyzed in agricultural soils, also considering their depth-wise distribution. DOM characterization was performed to assess the influence of specific DOM fractions on the vertical distribution and fate of PFAS in both community garden and agricultural soils. Moreover, the effect of PFAS chemical properties and the key physicochemical properties of the soils such as SOM content, moisture content, pH, conductivity, cation exchange capacity, anion concentrations, and soil texture, on the depth profiles were studied. To evaluate matrix-specific PFAS depth profiles, we compared the PFAS profiles in compost and landfill organics from our previous studies [34, 35] with the depth-wise PFAS profiles observed in soils in this study. The findings of this study offer critical insights into PFAS fate in soils, informing the development of targeted mitigation strategies.

## 2. MATERIALS AND METHODS

### 2.1. Community garden and agricultural soil

Community garden and agricultural soil samples were collected from locations in Fargo, North Dakota, during June and July 2024. No known PFAS-producing facilities or major point sources were located near the sampling areas. A community garden is a shared green space in an urban setting where residents cultivate fruits, vegetables, and flowers on individual plots, promoting urban sustainability, food security, and community engagement [36, 37]. Garden soil samples were collected from the Yunker Farms Community Garden at Fargo, which consists of approximately 200 plots of 20 ft by 30 ft each [36]. Soil samples were collected from six randomly selected plots at three depths—surface soil (0 ft), 1 ft, and 2 ft. At each depth, six individual samples were composited to prepare one representative sample. Agricultural soil samples were collected separately from corn (*Zea mays*), soybean (*Glycine max*), and



sunflower (*Helianthus annuus*) fields. In each crop field, samples were collected from six randomly selected locations at three depths—surface soil (0 ft), 0.5 ft, and 1 ft. Samples from the same depth within each crop field were composited to prepare one representative sample for that depth, and samples from different crop fields were not mixed. All garden and agricultural soil samples were collected using methanol-cleaned stainless-steel samplers. For both soil types, approximately 500 g of composite soil sample from each depth was retained for analysis. These composite soil samples were placed in methanol-rinsed polyethylene zip-lock bags, transported to the laboratory, and stored at 4 °C before extraction. Specific information on tillage/ploughing practices at the agricultural sites was not available. However, intensive tillage is generally not recommended in the Fargo, North Dakota region due to soil erosion concerns; therefore, substantial mechanical mixing of the soil profile is not expected. The latitude and longitude coordinates of the community garden soil sampling locations are provided in Table S1. Exact coordinates were not available for the agricultural and background soil sampling locations. However, a study area map is provided to show the approximate locations, directions, and distances of the background and agricultural soil sampling sites relative to the community garden soil location (Figure S1).

## 2.2. PFAS extraction and analysis

All soil samples were dried at 60 °C and grinded using a methanol-cleaned mortar and pestle. Homogenized soil samples were sieved twice through a 1.5 mm pore-sized methanol-cleaned aluminum net. Ten grams of soil were taken in duplicate from each depth-specific composite sample and transferred into pre-cleaned 50 mL polypropylene tubes for PFAS extraction. Then, 100 µL mass-labeled PFAS internal standard (MPFAC-HIF-ES, Wellington Laboratories) was added to the sample followed by proper mixing (Table S2). The targeted analytes, extracted internal standards, and non-extracted internal standards were similar to the EPA method 1633 (Table S3). PFAS extraction was conducted using a multi-step solvent extraction process with LC-MS grade methanol containing 0.3% ammonium hydroxide (Text S1). After extraction, supernatants were combined into pre-cleaned 15 mL polypropylene tubes. Then, the combined supernatant was concentrated to 1 mL using a nitrogen blowdown evaporator and kept at 4 °C until PFAS analysis. We also prepared procedural blank samples to assess background contamination during extraction (Text S2). The extraction recovery for the mass-labeled PFAS internal standards was 58–126%.

We targeted forty PFAS (demonstrated in the EPA method 1633 [38]) spanning from seven PFAS classes. These include eleven perfluorocarboxylic acids (PFCAs), eight perfluorosulfonic acids (PFSAs), three fluorotelomer carboxylic acids (FTCAs), three fluorotelomer sulfonic acids (FTSs), seven perfluoroalkane sulfonamides (PASFs), five perfluoroether carboxylic acids (PFECAs), and three perfluoroether sulfonic acids (PFESAs) (Table S4). The PFAS were quantified using an ultra-performance liquid chromatograph (UPLC) coupled with a Triple Quadrupole mass spectrometer (Vanqish UPLC-Altis MS, Thermo Scientific, USA) in the negative ionization mode. UPLC separation was carried out using a Phenomenex Luna Omega C18 column (2.1 × 100 mm) following our previous method (Text S3, Table S5) [34]. The measured PFAS concentrations in the soil samples are reported on a



dry weight basis. Moreover, we compared the PFAS depth profiles observed across soil layers in this study with those previously reported for compost and landfills in our recent studies [34, 35] to develop matrix-specific insights.

### 2.3. Physicochemical characteristics of soil

A certain mass of the dried homogenized soil was extracted using the ASTM D3987-12 protocol (Text S4) [39]. The extracts were filtered through 0.45  $\mu\text{m}$  Whatman filter papers (Sigma Aldrich, MO, USA), and the DOM content of the extracts were quantified using a Shimadzu TOC Analyzer (Table S6). The ultraviolet absorbance of the extracts was measured at 254 nm using a GENESYS 150 UV-Visible Spectrophotometer (Thermo Scientific, USA), and the specific UV absorbance (SUVA) was calculated (Text S5) [40]. The three-dimensional excitation-emission matrix (EEM) spectroscopy was conducted for the extracts using an FP-8350 Spectrofluorometer (Jasco Inc., Japan). The three-dimensional spectral data were collected for emission wavelengths ranging from 250 to 550 nm and excitation wavelengths ranging from 200 to 500 nm (Text S5 and Table S7). The three-dimensional data were viewed as two-dimensional contour plots to elucidate different organic fractions in soil DOM. The highest fluorescence intensities of these DOM fractions were considered to infer the relative levels of DOM components, providing an indirect measure [41]. The excitation and emission absorbance data were used to calculate the fluorescence index (FI), biological index (BIX), and humification index (HIX), to explain the DOM characteristics (Text S5 and Tables S7, S8) [42, 43]. The pH and conductivity of the soil extracts were measured using benchtop meters (Mettler Toledo, Fisher Scientific, USA) (Text S4). Additionally, we measured the anions, including chloride, nitrite, nitrate, sulfate, and phosphate in soil extracts, using ion chromatography (Thermo Scientific, DIONEX ICS 6000, USA) (Table S6). The cation exchange capacity (CEC) of the soil at each depth was measured in centimoles of positively charged ions per kilogram of soil ( $\text{cmol}_c \text{kg}^{-1}$ ), reflecting the soil's capacity to retain cations (Text S6). Moreover, we measured the moisture content of all soil samples and then combusted the soils to measure soil organic matter (SOM) content by following the ASTM D2974 method (Text S4) [44]. Furthermore, soil texture was tested using the hydrometer method to elucidate the composition of sand, silt, and clay in all analyzed soil samples [45].

### 2.4. Statistical analyses

Statistical analyses and data visualization were performed using JMP Pro (version 17.2) and OriginPro (version 2025, 10.05). The normality of PFAS concentration in soils at different depths was assessed using the Shapiro-Wilk test (Table S9). Since the data were not normally distributed, PFAS values were logarithmically transformed (Log-PFAS) prior to multivariate statistical analysis. Differences in PFAS concentrations among depths were evaluated using Kruskal-Wallis one-way ANOVA. We conducted polynomial regression analyses to investigate the relationships between PFAS classes, total PFAS, and soil properties such as SOM, DOM, and CEC, providing insights into PFAS fate in soils. Spearman correlation analysis was used to examine the relationship between PFAS distribution across depths and the measured soil physicochemical properties. All statistical analyses were conducted at a significance level of



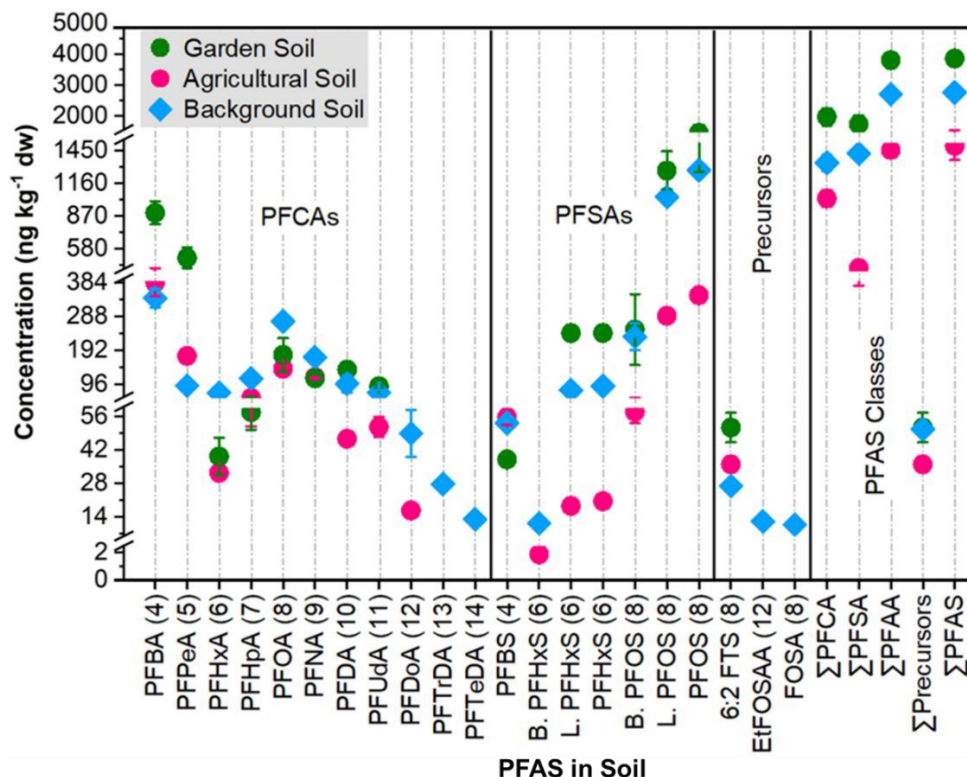
$\alpha=0.05$ , and results with  $p<0.05$  were considered statistically significant. Details on statistical analyses are provided in Text S7.

### 3. RESULTS AND DISCUSSIONS

#### 3.1. PFAS in community garden and agricultural soils

Thirteen PFAS compounds from three classes were detected in the community garden and agricultural soils (Figure 1), with concentrations significantly differing between the soil types (Table S10). Total PFAS concentration in the community garden soil ( $3,987 \pm 146 \text{ ng kg}^{-1}$ ) was 2.6 times of the agricultural soil ( $1,523 \pm 25 \text{ ng kg}^{-1}$ ) (Figure 1). PFAS concentrations ranging from 2 to 9,700  $\text{ng g}^{-1}$  have been reported in agricultural soils from China and Germany, due to the application of treated wastewater, biosolids, surface runoff, and PFAS-containing agrochemicals [46, 47]. PFAS levels in private garden soils near fluorochemical plants in Belgium ( $21.0\text{--}24.1 \mu\text{g kg}^{-1}$ ) [48] and in commercial garden soils in Australia ( $2.32\text{--}3.61 \mu\text{g kg}^{-1}$ ) [49] have been reported, but our study is the first to examine PFAS in community garden soils in urban areas. The higher concentrations in private gardens near fluorochemical plants are likely driven by direct point-source inputs, including atmospheric deposition, contaminated dust, and runoff [48]. In commercial garden soils, PFAS may originate from PFAS-containing biosolid-based soil amendments, compost, and potting mix components [49]. Similarly, PFAS accumulation in community garden soils can be attributed to compost application, agricultural pest control, and urban runoff [17, 34, 50]. Notably, our previous study measured elevated total PFAS ( $18,526 \pm 1,500 \text{ ng kg}^{-1}$ ) in yard waste compost, a common soil amendment in community gardens [34]. Additionally, we measured high total PFAS levels in background soils ( $2,878 \pm 110 \text{ ng kg}^{-1}$ ), which are neither agricultural nor garden soils, indicating that PFAS contamination is widespread and can be transported to surrounding areas (Figure 1). Runoff from sites such as airports, military bases, or firefighting training areas, driven by rainfall or snowmelt, can transport PFAS and PFAS containing particulates into low-lying lands [51, 52], contributing to widespread PFAS accumulation. PFAS in community garden soils may pose potential exposure risks to residents through inhalation of resuspended dust and consumption of garden produce [2, 53]. Children and older adults may be more vulnerable due to frequent soil/dust contact, cumulative exposure, and pre-existing health conditions [2, 54]. Exposure to certain PFAS has been linked to developmental effects, immune disruption, and thyroid dysfunction [55, 56].





**Figure 1.** Concentrations of PFAS in community garden, agricultural, and background soils. Thirteen PFAS spanned from three PFAS classes- perfluorocarboxylic acids (PFCAs), perfluorosulfonic acids (PFSA), and fluorotelomer sulfonic acids (FTS) were detected in community garden and agricultural soils. Concentrations are presented as mean  $\pm$  standard deviation of duplicate samples. The detected PFAS species and their concentrations are presented in Table S11. Extended forms of the PFAS are available in Table S4.

Significant differences between the analyzed PFAS classes, such as PFCAs and PFSA, were observed between the community garden and agricultural soil (Table S12). Approximately 98.7% of the total PFAS detected in the community garden soil were perfluoroalkyl acids (PFAAs), comprising 52.4% perfluorocarboxylic acids (PFCAs) and 46.3% perfluorosulfonic acids (PFSA). In contrast, PFCAs comprised 69.2% and PFSA comprised 28.4% of the total PFAS in agricultural soils. Among the PFCAs in the community garden soil, short-chain PFAS ( $C \leq 6$ ) such as perfluorobutanoic acid (PFBA;  $924 \pm 103$  ng kg<sup>-1</sup>) and perfluoropentanoic acid (PFPeA;  $523 \pm 91$  ng kg<sup>-1</sup>) were dominant (Figure 1). Additionally, the concentration of long-chain PFAS such as perfluorooctanoic acids (PFOA) and perfluorosulfonic acids (PFOS) was  $185 \pm 47$  ng kg<sup>-1</sup> and  $1,561 \pm 271$  ng kg<sup>-1</sup> (Figure 1). In agricultural soil, the dominant PFAS were PFBA ( $392 \pm 39$  ng kg<sup>-1</sup>), PFOA ( $146 \pm 7$  ng kg<sup>-1</sup>), and PFOS ( $355 \pm 22$  ng kg<sup>-1</sup>), which are lower than previously reported concentrations of PFOA ( $3,400$ – $47,500$  ng kg<sup>-1</sup>) and PFOS ( $1,700$ – $59,000$  ng kg<sup>-1</sup>) in agricultural soils [26, 46]. This could be attributed to the voluntary phase-out of PFOS and PFOA by chemical manufacturers in the United States in 2002 and 2015 [57].

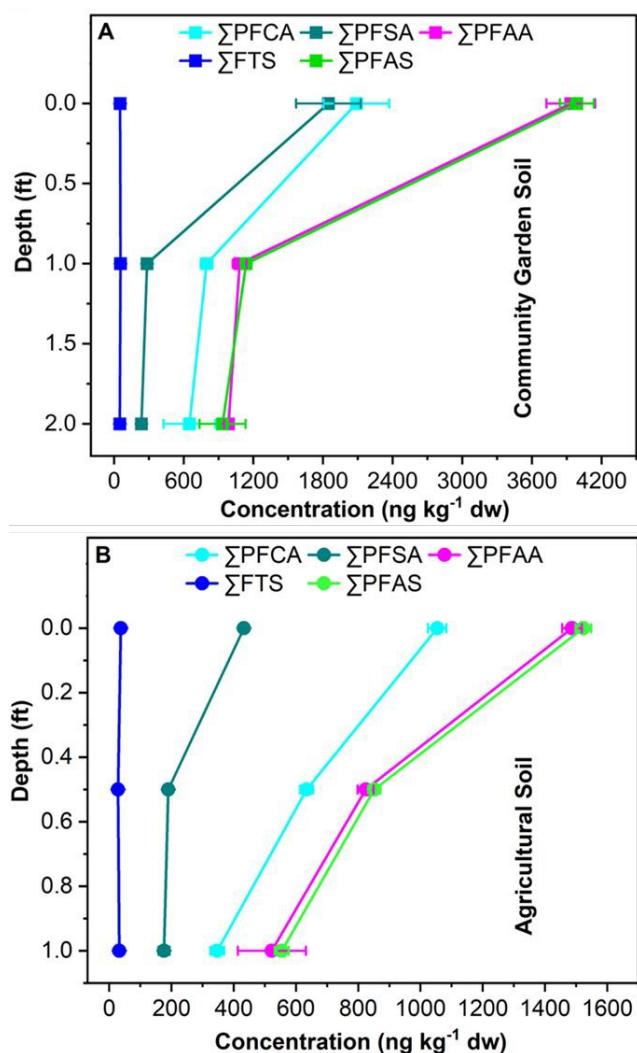


Long-chain PFAS ( $C > 6$ ) comprised 55% of the total PFAAs in community garden soils compared to 63% in agricultural soils, while short-chain PFAS accounted for 45% and 37%, respectively. The high concentration of long-chain PFAS in both soils is likely due to their lower water solubility, greater hydrophobicity, and stronger sorption to organic matter, which enhances their persistence in soil [58-60]. Interestingly, linear PFOS and linear PFHxS dominated over their branched isomers in both soils, likely due to their stronger sorption onto organic matrices (Figure 1). Among the targeted PFAA precursors, only 6:2 fluorotelomer sulfonic acids (6:2 FTS) were measured in both soils (Figure 1). Interestingly, perfluoroalkane sulfonamides such as EtFOSAA and FOSA were detected in background soils but not found in garden and agricultural soil (Figure 1). This indicates that greater microbial activity in agricultural and garden soils can facilitate the biotransformation of fluorotelomers and sulfonamides into PFAAs [61, 62]. Soil microbes can bio-transform fluorotelomer- and sulfonamide-based PFAS, leading to the formation of PFAAs [63, 64]. Consequently, some of the PFAAs detected in soils may result from the microbial transformation of precursors.

### 3.2. PFAS vertical distribution in soil as a function of organic matter

PFAS concentrations in the community garden and agricultural soils were significantly greater at the surface compared to the deeper layers (Figure 2, S2, and S3). The total PFAS concentration in the garden surface soil (0 ft,  $3,987 \pm 146 \text{ ng kg}^{-1}$ ) was 4.3 times of that at 2 ft ( $934 \pm 198 \text{ ng kg}^{-1}$ ), while in agricultural soils, the surface PFAS concentration ( $1,523 \pm 25 \text{ ng kg}^{-1}$ ) was 2.7 times of that at 1 ft ( $554 \pm 22 \text{ ng kg}^{-1}$ ) (Figure 2). The higher concentration of PFAS in the surface levels can be attributed to the greater SOM content in these layers than in subsurface soil layers [65-67]. A significant (Spearman's  $\rho=0.86$ ,  $p<0.05$ ) positive correlation between total PFAS and SOM across different depths was observed (Table S13). In the garden soil, SOM was 3.5% at the surface, decreasing to 2.6% at 1 ft, and 2.1% at 2 ft (Table S6). In agricultural soils, surface-level SOM was 3.1%, declining to 2.4% at 1 ft (Table S6). In addition to total PFAS, PFCAs and PFSA concentrations were higher in surface soils of both garden and agricultural sites (Figure 2). Thus, the strong positive correlation between total PFAS, PFAS classes, and SOM in this study highlights SOM's key role in PFAS retention in surface soils [68-70]. As a soluble component of SOM, DOM can provide additional binding sites that promote stronger molecular-level interactions with PFAS, affecting their retention and distribution beyond the influence of SOM.



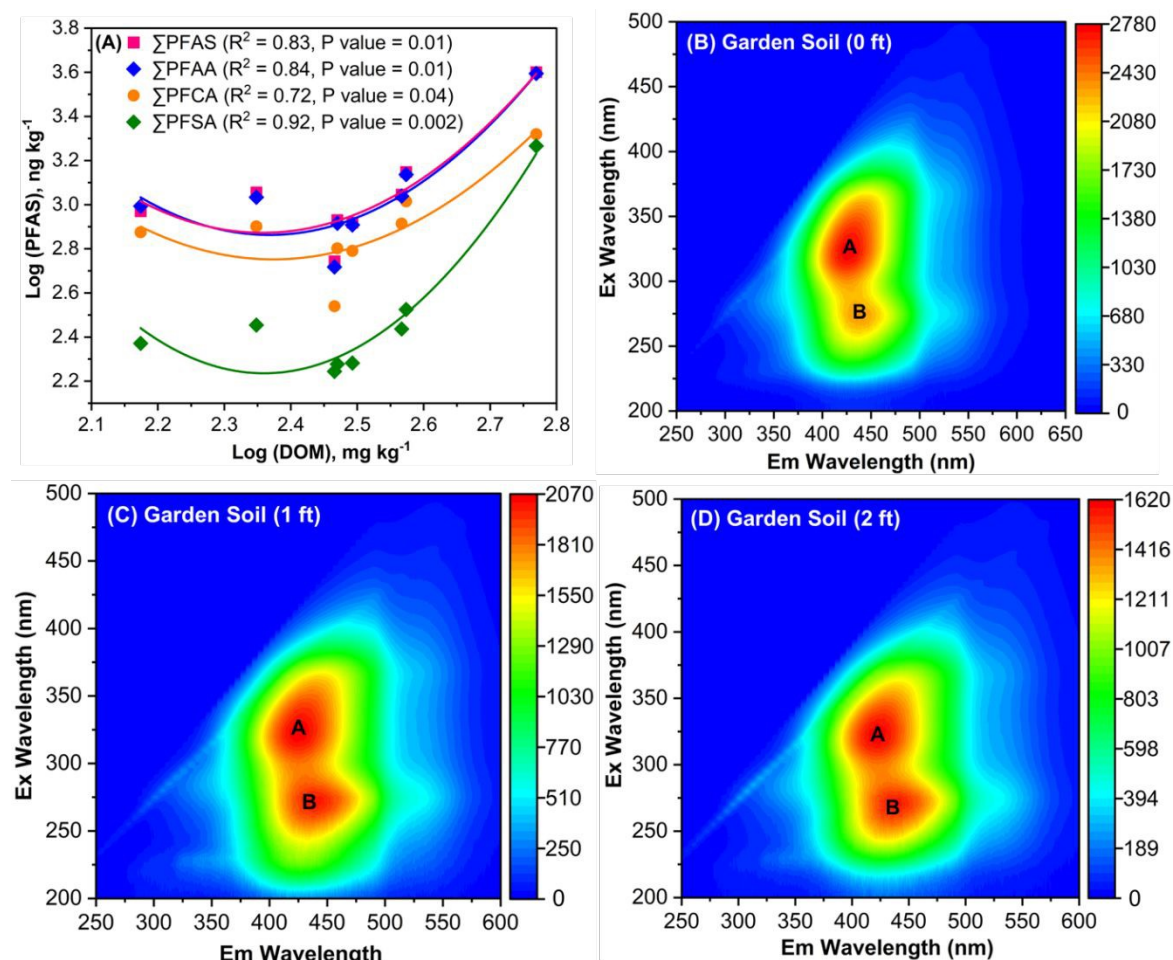


**Figure 2.** Concentrations of different PFAS classes and total PFAS in the community garden soil (A) and agricultural soil (B) at different depths. Three PFAS classes were detected, such as perfluorocarboxylic acids (PFCAs), perfluorosulfonic acids (PFSAs), and fluorotelomer sulfonic acids (FTSs). PFCAs and PFSAs are collectively known as perfluoroalkyl acids (PFAAs). Only 6:2 fluorotelomer sulfonic acid (6:2 FTS) was detected among the FTSs in soil across depths. Extended forms of PFAS are available in Table S4.

The total PFAS and the DOM content of the soils at different depths demonstrated a significant (Spearman's  $\rho=0.82$ ,  $p<0.05$ ) positive correlation (Figure 3 and Table S14). Besides total PFAS, DOM demonstrated a strong positive correlation with PFCAs and PFSAs. Community garden soil contained significantly higher DOM at the surface ( $588\pm 65$  mg kg<sup>-1</sup>) compared to 1 ft ( $223\pm 44$  mg kg<sup>-1</sup>) and 2 ft ( $149\pm 27$  mg kg<sup>-1</sup>) (Table S6). Similarly, in agricultural soil, DOM was  $375\pm 7$  mg kg<sup>-1</sup> in surface soil and declined with depth to  $295\pm 3$  mg kg<sup>-1</sup> at 0.5 ft and  $292\pm 7$  mg kg<sup>-1</sup> at 1 ft (Table S6). The elevated concentrations of long-chain PFAS (e.g., PFOA, PFOS) at the surface level of the garden and agricultural soils, coupled with high DOM levels, demonstrate the strong influence of DOM on long-chain PFAS retention (Figure S2-S4 and Table S6). Interestingly, perfluoroundecanoic acid (PFUdA) and



perfluorododecanoic acid (PFDoA) were detected in the surface of garden soil but were not detected at 1-ft and 2-ft depths, highlighting that PFAS with longer C–F chains tend to accumulate more in surface layers (Figure S2). Several studies reported that long-chain PFAS are retained in the soil surface layer, whereas short-chain PFAS typically move downward due to their lower affinity for soil organic matter [71–73].



**Figure 3.** Polynomial regression analysis between Log-DOM and Log-PFAS in soil across depths (A). Regression analyses of Log-transformed PFAS and DOM data from the community garden and agricultural soils illustrate the strong positive correlation between PFAS and DOM. Fluorescence three-dimensional excitation–emission matrix (3D EEM) spectra of DOM in the community garden soil (B–D). Peaks A and B represent humic-like and fulvic-like substances, respectively. The highest fluorescence intensities of these peaks were used to estimate the relative abundance of humic- and fulvic-like substances in soils at different depths. Similar fluorescence features for agricultural soils are provided in the Supplementary Information (Figure S5). Details of the excitation and emission wavelengths corresponding to the identified peaks are listed in Table S7.

DOM in the garden and agricultural soils mainly consists of humic- and fulvic-like substances, with higher fluorescence intensity at surface soils than subsurface soils (Figure 3,



S5 and Table S7). There is a significant positive correlation between the intensity of humic- and fulvic-like substances and PFAS concentrations across depths (Figure S6, S7). In the community garden soil, the surface layer was enriched with humic-like (2,770 au) and fulvic-like (2,330 au) substances, whereas their intensity decreased to 1,610 au and 1,570 au, respectively, at 2 ft depth (Figure 3 and Table S7). A similar depth-wise distribution pattern of humic- and fulvic-like substances was observed in agricultural soils (Figure S5 and Table S7). Moreover, the surface layers of the garden soil (humification index 1.4,  $SUVA_{254}$  0.7 L mg<sup>-1</sup> m<sup>-1</sup>) and agricultural soil (humification index 1.3,  $SUVA_{254}$  0.6 L mg<sup>-1</sup> m<sup>-1</sup>) showed higher humification and aromaticity than deeper layers, indicating more mature and stable DOM in surface layer soils [74, 75] (Table S6, S8). The stable DOM across soil depths is primarily derived from microbial and terrestrial sources, as indicated by FI values ranging from 1.4–1.6 in garden soil and 1.5–1.7 in agricultural soil across depths (Table S6, S8). Higher BIX values in the surface soils of both garden (biological index 1.2) and agricultural soil (biological index 0.9) compared to their deeper layers indicate a greater proportion of DOM derived from microbial activities in the surface soils (Table S6, S8). This enhanced microbial activity, along with greater oxygen availability at surface soils, can promote the transformation of PFAS precursors into terminal PFAS (i.e., both long- and short-chain PFAAs) [76, 77]. Several studies reported that biotransformation of PFAS is generally higher in topsoil (0-10 cm) due to high microbial activity and oxygen availability [47, 78].

### 3.3. Impacts of additional factors on PFAS vertical distribution in soil

Polynomial regression and Spearman correlation analyses showed that soil CEC strongly influenced PFAS distribution across soil depths, with higher CEC values linked to greater PFAS levels (Figure 4, S8 and Table S15). In garden soils, CEC was highest at the surface (37 cmol<sub>c</sub> kg<sup>-1</sup>), decreasing to 19 cmol<sub>c</sub> kg<sup>-1</sup> at 1 ft and 17 cmol<sub>c</sub> kg<sup>-1</sup> at 2 ft (Table S6). A similar trend was observed in agricultural soils, where surface soil exhibited a higher CEC (36 cmol<sub>c</sub> kg<sup>-1</sup>) compared to 0.5-ft (17 cmol<sub>c</sub> kg<sup>-1</sup>) and 1-ft (15 cmol<sub>c</sub> kg<sup>-1</sup>) depths (Table S6). The strong (Spearman's  $\rho=0.81$ ,  $p<0.05$ ) positive correlation between CEC and total PFAS can be attributed to the combined effects of soil texture, cation bridging, and ionic interactions. The clay and silt content in the soil may reduce hydraulic conductivity (Table S6), limiting the downward flow of dissolved PFAS and favoring their retention in surface soils [79, 80]. The higher CEC in the surface layer suggests a greater abundance of exchangeable multivalent cations capable of mediating cation bridging between negatively charged PFAS functional groups (carboxylate or sulfonate) and soil particles [81-83]. These cations act as electrostatic links, reducing repulsion between anionic PFAS and the negative charge sites of the soil matrix, and thereby enhancing PFAS sorption and retention [84, 85].

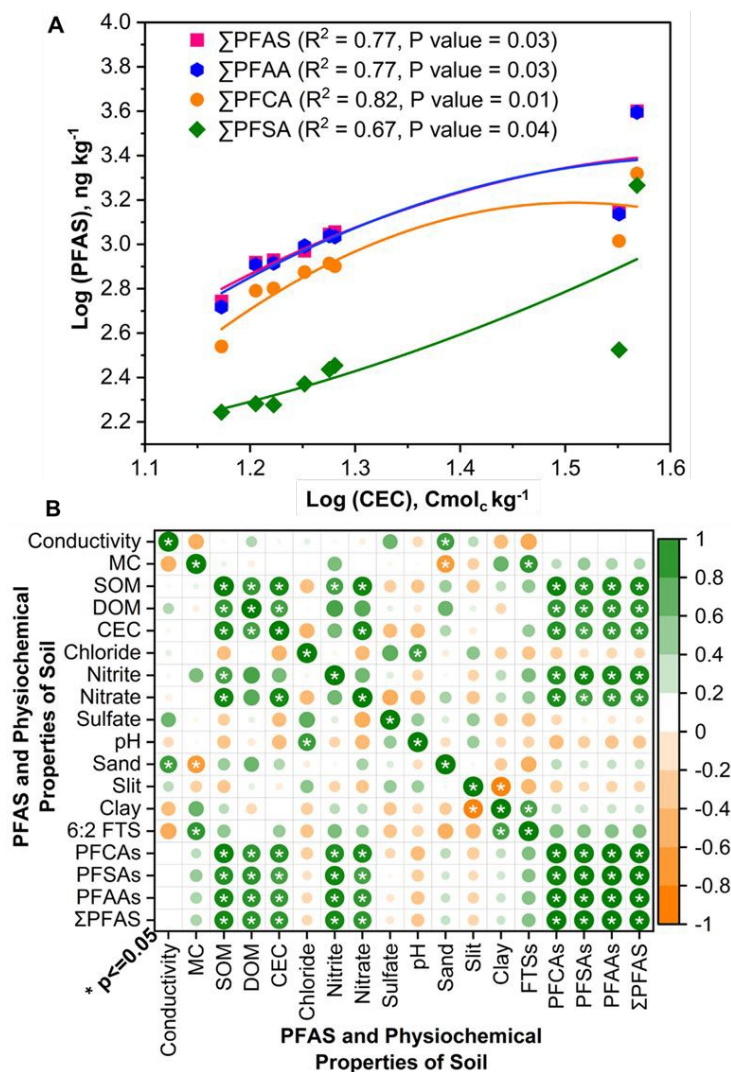
A significant positive correlation was observed between short-chain PFAS, such as PFBA, PFPeA, and PFHxA, and the CEC of soils at different depths (Figure S8). The concentrations of short-chain PFAS in garden surface soil, including perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), and perfluorohexane sulfonic acid (PFHxS) were 61%, 72%, and 89% higher than the soil at 2 ft depth. Similarly, PFBA, PFPeA, and PFHxA in



agricultural surface soil were 2.2, 6.6, and 1.7 times of those in soil at 1 ft. This likely reflects enhanced sorption of short-chain PFAS through cation-mediated electrostatic interactions and bridging with available surface binding sites in soil [86, 87]. Strong positive correlations were observed between total PFAS, PFCAs, and PFSAAs and the concentrations of nitrite ( $\text{NO}_2^-$ ) and nitrate ( $\text{NO}_3^-$ ) across soil depths (Figure 4). This relationship likely reflects biogeochemical conditions in surface soils, with high organic matter enhancing PFAS retention and active microbial processes driving nitrogen transformations, resulting in elevated nitrite and nitrate levels [88, 89]. In contrast, PFAS showed no significant correlations with chloride ( $\text{Cl}^-$ ) and sulfate ( $\text{SO}_4^{2-}$ ), likely due to their distinct geochemical behaviors (Figure 4, S8). Chloride is highly mobile and weakly sorbed, while sulfate preferentially adsorb to Fe and Al oxides, limiting their interaction with PFAS [90, 91].

Furthermore, soil pH, electrical conductivity, and moisture can influence PFAS fate by affecting the speciation and interactions with DOM in soils. The soils in this study were slightly acidic, with pH ranging from 6.6 to 6.8 (Table S6). Under mildly acidic conditions, PFAS remain predominantly in their anionic form, which can interact with DOM mainly through hydrophobic interactions [92]. However, we did not find a significant correlation between pH and PFAS distribution across depths, likely because pH did not vary substantially with depth (Table S6). Garden surface ( $1,566 \mu\text{S cm}^{-1}$ ) and agricultural surface soil ( $1,380 \mu\text{S cm}^{-1}$ ) had higher electrical conductivity than soil in deeper layers (Table S6). This higher conductivity reflects greater ionic strength of the analyzed soils due to the presence of soluble ions [93]. High ionic strength can enhance electrostatic interactions, increasing retention and reducing the movement of anionic PFAS [84]. Soil moisture increased with depth in both community garden and agricultural soils (Table S6), but PFAS distribution was not significantly impacted by moisture (Figure 4, S4). Deeper soil layers are relatively wetter because surface soil may lose water through evaporation and plant uptake [94]. Thus, no significant correlations with pH, moisture, or conductivity were observed in this study (Figure 4, S4), suggesting that CEC, along with DOM components, play a strong role in controlling PFAS distribution in these soils.

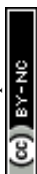




**Figure 4.** A. Polynomial regression analysis between Log-CEC and Log-PFAS in soil across depths. Regression analysis of Log-transformed cation exchange capacity and PFAS in community garden and agricultural soils illustrates the strong positive correlation between the soil's CEC and PFAS. B. Spearman correlation analysis between soil physicochemical properties and total PFAS, as well as PFAS classes, at different depths in the community garden and agricultural soil. The green circle with '\*' denotes a significant correlation between the pairs. The green color in the right-side bar indicates the intensity of a strong correlation, and the orange color denotes the intensity of a weak correlation among the pairs. Extended forms of the PFAS are provided in Table S4.

### 3.4. Matrix-specific PFAS vertical distribution patterns

PFAS vertical distribution varies across environmental matrices. In the community garden and agricultural soils, total PFAS was highest in surface layers compared to deeper depths (Figure 2). PFAS retention in the surface soil is driven by high organic matter content, including humic- and fulvic-like components, and cation exchange capacity of soil, which



collectively enhance sorption of both long- and short-chain PFAS. In contrast, our recent study on an idle mature compost pile exhibited the opposite trend, with surface PFAS concentrations approximately 59–63% lower than at 2 ft (Figure 9A). This pattern likely reflected the dominant downward transport of PFAS and DOM with moisture in compost. Landfill organics, however, demonstrated a markedly different vertical pattern, with total PFAS concentrations peaking at 20–35 ft—approximately 2.5 times higher than those in the 10–20 ft and 35–50 ft layers (Figure 9B). In landfills, PFAS distribution was strongly correlated with total carbon and DOM, highlighting the role of organic matter in controlling PFAS accumulation. The vertical distribution of PFAS in landfill organics may also be influenced by waste heterogeneity, cover status, and the age of landfilled waste, which collectively affect PFAS release, mobility, and retention within landfills. These observations demonstrate that PFAS vertical distribution is highly matrix-dependent. Understanding these matrix-specific distribution patterns is critical for predicting PFAS fate, transport, and potential exposure risks in different environmental settings.

#### 4. Conclusions

In this study, elevated total PFAS concentrations were detected in community garden soils in an urban setting. These concentrations exceeded those found in agricultural soils, suggesting a potentially higher exposure risk for gardeners and consumers of garden produce. Short-chain PFCAs, which are more readily taken up by plants, were also higher in community garden soils than in agricultural and background soils. Unlike agricultural soils, which have been extensively studied, community garden soils often receive inputs from compost, landscaping materials, and urban runoff, creating complex contamination scenarios. Monitoring PFAS levels in these soils is essential for understanding dietary and dermal exposure, particularly for vulnerable populations such as children and senior citizens who spend extended periods working in gardens. Even trace levels of PFAS in edible plants can contaminate the food chain, emphasizing the need for targeted and non-targeted monitoring and risk assessment in urban gardens. Additionally, understanding the depth profiles of PFAS in soil and other environmental matrices is critical for predicting their fate, transport, and potential exposure risks in the environment. Thus, the findings of this study are crucial for developing effective soil management, remediation strategies, and risk assessment frameworks to protect human health and environmental quality.

#### Supplementary Information

Supporting texts on PFAS extraction, quality assurance and quality control (QA/QC), UPLC–MS/MS analysis for PFAS quantification, pH and dissolved organic matter analysis, ultraviolet and fluorescence 3D excitation-emission matrix spectroscopy, anions and cation exchange capability measurement, and statistical analyses; supporting tables on sampling locations coordinates, mass-labeled PFAS internal standards, UPLC–MS/MS operating conditions, extended forms of PFAS, soil physiochemical characteristics, fluorescence spectroscopy parameters, excitation-emission matrix indices, normality test, Kruskal-Wallis one-



way ANOVA, detected PFAS and their concentration in soils, and polynomial regression analysis; supporting figures of concentrations of the investigated PFAS in soil at different depths, Fluorescence three-dimensional excitation–emission matrix (3D EEM) spectra of DOM, Spearman correlation analysis, and matrix specific PFAS distribution pattern.

### Author Contributions

Biraj Saha: Methodology, Data Curation, Formal Analysis, Visualization, Writing; Mohamed Ateia: Methodology, Writing, Discussion, Editing; Sujan Fernando: Data Curation, Discussion, Editing; Thomas DeSutter: Writing, Discussion; Syeed Md Iskander: Conceptualization, Writing, Editing, Overall Management.

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### Conflicts of Interest

There are no conflicts to declare.

### Data Availability

Data will be made available upon request.

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## Data Availability

Data will be made available upon request.

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