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Bioaccumulation and Translocation of 6:2 Fluorotelomer Sulfonate, GenX, and Perfluoroalkyl Acids by Urban Spontaneous Plants

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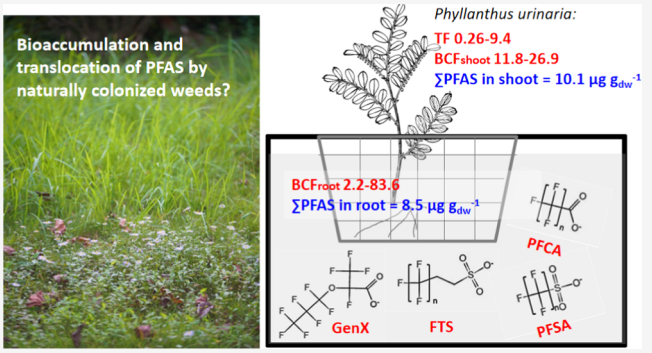
ABSTRACT: There is limited information available regarding the bioaccumulation potential of polyfluoroalkyl substances (PFAS) in urban vegetation. Using a controlled greenhouse exposure setting, we investigated the bioaccumulation and translocation of select PFAS in four common urban spontaneous plants. Target compounds included legacy PFAS (perfluoroalkyl carboxylic and sulfonic acids, PFCA/PFSA), a fluorotelomer sulfonate (6:2 FTS), and an emerging fluorinated ether (i.e., hexafluoropropylene oxide dimer acid (HFPO-DA), or GenX). Results from this study showed that bioaccumulation factors in root and shoot (BCF_{root} and BCF_{shoot}) ranged from 0.7 to 83.6 and 0.95 to 26.9, respectively. *Phyllanthus urinaria* harbored the highest PFAS bioaccumulation capacity among the four urban weed species. The log BCF_{root} of PFCA homologues showed a concave shape as a function of chain length, while log BCF_{root} of PFSA increased with chain length. The BCF_{root} of GenX was lower than that of PFOA; likewise, 6:2 FTS bioaccumulated to a less extent than PFOS. Root uptake seemed to be the dominant accumulation mechanism for the shorter-chain compounds, whereas adsorption was the dominant mechanism for longer-chain compounds such as PFOA. BCF_{root} and BCF_{shoot} showed consistent trends in response to foliar and root characteristics. Leaf area and average root diameter were the most correlated traits with PFAS bioaccumulation factors, with higher BCF values for plants with smaller leaves and finer roots. This study also provides an important basis for the role and selection of urban weeds in future PFAS bioaccumulation and translocation studies within urban settings.



KEYWORDS: *GenX (HFPO-DA), 6:2 FTS, urban spontaneous plants, bioaccumulation, phytoremediation*

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) have been extensively used in industrial and consumer products, and many of these chemicals are detected in the global environment and biota.[1−7] Perfluorooctane sulfonic (PFOS) and carboxylic acids (PFOA) are examples of PFAS currently subject to regulatory and scientific scrutiny.[8,9] To specifically address concerns related to PFOS and PFOA toxicity, PFAS alternative products with various structures have emerged.[7,8,10] Among various "new" or current-use PFAS, the main alternative strategy is to reduce the "effective length" of the perfluorinated segment. For instance, shorter fluoroalkyl chains were used to replace C8-PFAS (e.g., PFOA and PFOS), such as C2−C6 perfluoroalkyl acids (e.g., perfluorobutanoic acid, PFBA, perfluorohexanoic acid, PFHxA, and perfluorobutane sulfonate, PFBS) and fluorotelomer sulfonates (e.g., 6:2 FTS). Inserting one or more oxygen atoms in the $-CF_2-$ perfluoroalkyl chain is another strategy, as exemplified by perfluoro-2-propoxypropanoic acid, also termed hexafluoropropylene oxide dimer acid (HFPO-DA) or the trade name "GenX."[7,8,10]

Recently, new alternatives such as GenX have been more frequently detected in various aquatic and terrestrial environments, including many chemicals that are not explicitly used commercially.[4,11−16] Among various remediation techniques, uptake by plants may serve as a potential solution to remedy low-level PFAS-contaminated sites by sequestrating and immobilizing PFAS (through phytoremediation) where other methods of treatment are too expensive or impractical, and only "polishing treatment" is required over long periods of time. Further, phytoremediation has other advantages in terms of aesthetics, cost-effectiveness, and long-term applicability.[17]

Recent studies have demonstrated that plants are capable of PFAS uptake via the vascular system. After uptake, PFAS can be translocated to stems, shoots, leaves, and fruits.[18−24] Studies

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also found that the extent of PFAS accumulation and translocation inside the plant (manifested by bioaccumulation factors, BCFs, and translocation factors, TFs, respectively) tends to decrease with increasing carbon-chain length.[25−27] Hydrophobicity or the PFAS chain length seems to have a greater impact on the root uptake and translocation than differences resulting from the functional head group.[25−27] However, target pollutants in past studies were mainly PFOS and PFOA, and their uptake and translocation patterns were only investigated among agricultural crops,[18−21] wetland plants,[22,23] and trees.[24] Since the behaviors of accumulation vary greatly among plant species, clarifying accumulation patterns across different plant taxa is crucial to better understand plant uptake and translocation of PFAS, particularly in terms of identifying suitable candidates for phytoremediation in future work. There is also a strong need to better understand plants' responses to new PFAS alternatives that have different structures compared with traditional PFOS and PFOA.

Here, we conducted hydroponic greenhouse experiments using four urban spontaneous plants that are commonly distributed worldwide. These weeds require no human assistance to assert and maintain themselves in extreme, often volatile urban conditions. Furthermore, they have been shown to have promising applications in urban restoration practices. In fact, many of them can grow well under postmining site conditions, and they can serve as a good option in urban greening initiatives within polluted areas to develop species-rich plant communities and accumulate heavy metal pollutants.[28−30]

With this background information, the objectives of the current study are to (1) assess the concentration and distribution of PFAS in various plant tissues and calculate the bioaccumulation factors in root and shoot (i.e., BCF_{root} and BCF_{shoot}), translocation factor (TF), and total burden for PFAS for four weed species; (2) determine the species that has the most significant bioaccumulation of PFAS and identify the key physiological characteristics (e.g., root and leaf macroscopic morphology) influencing a plant's PFAS accumulation potential; and (3) compare the behavioral differences between emerging alternatives (e.g., GenX and 6:2 FTS) and legacy PFOS and PFOA in terms of bioaccumulation. The results from this study are also important to better assess the ability of urban weeds to be used in phytoremediation initiatives of PFAS in future work.

2. MATERIALS AND METHODS

2.1. Chemical Reagents and Lab Materials. Ten PFAS of four classes were targeted for this work on the basis of the availability of analytical standards (Table S1 and Figure S1). Analytes in this study include 6:2 fluorotelomer sulfonate (6:2 FTS), hexafluoropropylene oxide dimer acid (HFPO-DA, or the parent acid of "GenX"), three perfluoroalkyl sulfonates [PFSA, i.e., perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), and PFOS], and five perfluorinated carboxylic acids [PFCA, i.e., perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), and PFOA]. Certified standards as well as stable isotope-labeled standards were obtained from Wellington Laboratories (Guelph, ON, Canada). HFPO-DA was purchased from SynQuest Laboratories, Inc. (Alachua, FL). Upon receiving, a stock solution was prepared for each compound in methanol with a concentration of 1 g L^{-1}. Before the exposure experiments, all PFAS were prepared in one aqueous working solution (each compound at 0.01 g L^{-1}) and investigated as a mixture. Ultrapure water (UPW) used in the tests was obtained from a Milli-Q system (Millipore, Billerica, MA). HPLC-grade acetonitrile, certified sodium hydroxide, LC-MS-grade methanol, water, and acetic acid were purchased from Sigma-Aldrich (St. Louis, MO). For extraction cleanup, chromabond diamino from Macherey-Nagel Inc. (Bethlehem,PA) and Supelclean ENVI-Carb from Sigma-Aldrich were used. A description of the PFAS compounds, both directly purchased from chemical vendors and the custom-synthesized ones, is provided in the Supporting Information, as well as the analytical standards and other reagents.

2.2. Plant Culture and Exposure Experiments. Bioaccumulation was studied with four common urban weeds, i.e., *Phyllanthus urinaria* (*Pu*), *Justicia procumbens* (*Jp*), *Eleusine indica* (*Ei*), and *Aster indicus* (*Ai*). Representative photographs of plants are provided in Figure S2A−J in the Supporting Information. These four plants are common urban weeds in China, and plants of the same families span a wide geographic range across global urban spaces. In addition, they possess significantly different morphological and physiological traits (Table S2 and Figure S2A−J) and have demonstrated superenrichment of heavy metals in previous studies.[28−31] The experiments were conducted in five replicates in a greenhouse (25 °C, 14 h light). Plants were collected from the campus of Chongqing University (Shapingba District, Chongqing, China), and no source of industrial pollution (e.g., factories) within 10 km was confirmed. Upon collection, the plants were first carefully removed from the soil, soil was washed off the roots with UPW, and plants were transferred to an experimental hydroponic system. PFAS-spiked UPW was prepared by spiking the aqueous working solution to UPW in a large polypropylene (PP) jar, and then a 300 mL aliquot of solution was removed and added to each experimental unit. Methanol content in the feed solution was <0.1‰ v/v. Because all weeds are spontaneous, ruderal species, thus no additional nutrient solution was added to the culture solution. The hydroponic system was used to avoid sorption of the dosed chemicals to soil and to help ensure that the spiked PFAS were completely bioavailable. The weeds were set in mesh pots, which were inserted into the lid of 1 L PP buckets. The buckets were wrapped in plastic wrap and aluminum foil to keep the root zone dark. Control experiments without PFAS were also carried out on four plants in triplicates. In addition, another group of buckets (n = 3) was set up without weeds to estimate the evaporation loss. The buckets were randomly arranged daily to account for any spatial variations in light and temperature within the greenhouse.

Weeds were harvested after 21 days, gently rinsed with UPW three times, and dried superficially. Root and shoot were separated and then frozen at −20 °C in sealed plastic bags until extraction. After the 21 day exposure, the volumes of water in the pots of the control group were 86.7−96.9% of the initial volume, and the concentrations of PFAS were 82.4−119.3% of their initial doses, suggesting that the losses of PFAS due to evaporation, degradation, and adsorption to the reactors were negligible.

2.3. Plant Characterization. For the four urban weeds, we measured a list of plant foliar and root traits prior to cultivation. Foliar traits measured in this study included leaf chlorophyll content, average leaf area and mass, specific leaf

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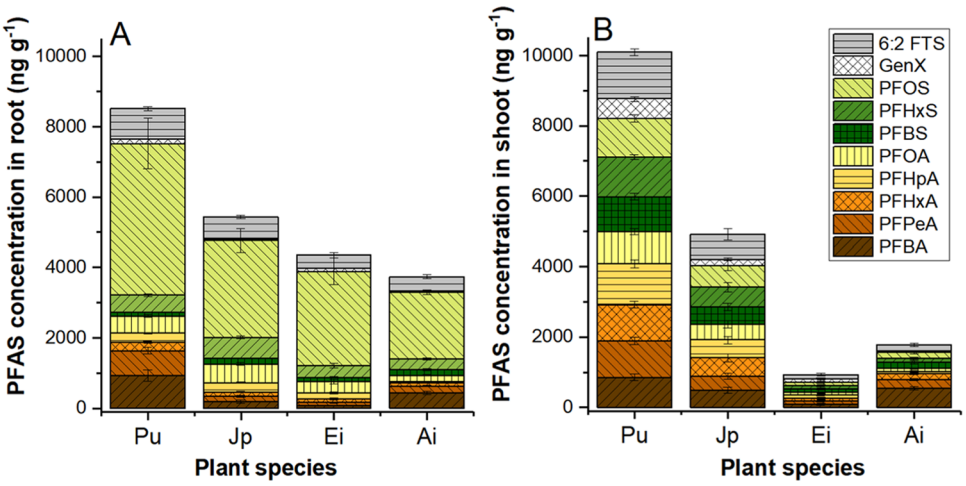

Figure 1. Bioconcentrations (ng g^{-1}, based on dry weight) of 10 PFAS in (A) root and (B) shoot of four urban spontaneous plants, i.e., *E. indica* (*Ei*), *J. procumbens* (*Jp*), *P. urinaria* (*Pu*), and *A. indicus* (*Ai*) after 21 day exposure. Error bar represents the standard deviation of five replicates.

area, leaf mass-based carbon (C), nitrogen (N), and phosphorus (P) content, and C/N, C/P, and N/P ratios. We selected this set of traits as they can represent weed's biochemical and physiognomic characteristics that control photosynthesis, nutrient, and water cycling. For root traits, we measured total root length, project area, surface area, surface area to length ratio, average diameter, and length per volume. These root morphological traits are recognized as important functional facets for a plant's nutrient uptake in soil. Detailed protocols for the measurements of plant foliar and root traits are provided in Supporting Information Text S1.

2.4. Sample Pretreatment. Freeze-dried plant samples were homogenized prior to extraction using a handheld food grinder. The crushing blade and container of the food processor were cleaned with UPW and chromatographically pure methanol three times between each sample. An aliquot of the homogenized plant tissue (0.5 g) was transferred to a 50 mL polypropylene tube and amended with a mixture of surrogate isotope-labeled internal standards. Solvent extraction proceeded as follows: after the addition of 4 mL of methanol with 2.0% ammonium hydroxide, samples were submitted to high-speed vortexing for 2 min, sonicated for 15 min at 30 °C, and placed on a shaker for 1 h. The tubes were then centrifuged (5000 rpm, 30 min), and the supernatant was transferred out to another 15 mL tube. The extraction procedure was repeated twice for a total of three cycles. 1.2 mL of the combined supernatants were transferred into a 1.5 mL microcentrifuge tube containing 15 mg of ENVI-Carb adsorbent. The samples were then vortexed for 1 min and centrifuged at 13 000 rpm for 10 min. The supernatant was filtered through a 0.45 μm glass fiber filter into an HPLC vial and stored at −20 °C prior to LC-MS/MS analysis. For analysis of the exposure medium (culture solution), aqueous samples were diluted in methanol and amended with internal standards prior to LC-MS/MS analysis.

2.5. PFAS Analysis. Quantitative analyses of PFAS were performed using ultraperformance liquid chromatography coupled to tandem mass spectrometry via a negative electrospray ionization source. The LCMS-8060 (Shimadzu, Japan) was operated in multiple reaction monitoring (MRM) mode. Calibration standards were prepared from 0.05 to 50 μg L^{-1}, with eight calibration points in the regression line. The limit of quantification (LOQ) was defined as the lowest point of the standard curve, provided that the regression equation yielded a calculated value within less than ±30% error. Additional details on the quantitative analytical method, including chromatographic gradient, source parameters, and MS/MS transitions, are provided in Supporting Information Table S3.

2.6. Quality Assurance and Control. The instrumental limit of detection (LOD) was 0.025 ng mL^{-1} for PFOS and 0.01 ng mL^{-1} for other PFAS. A blank matrix was obtained from the control group, where concentrations of ambient PFAS in root and shoot of four weed species grown in tap water were quantified. Measurable ambient blank levels were only observed for PFOA, close to the LOD. PFAS concentrations were below the LOD in laboratory extraction blanks (n = 3). Matrix effects were evaluated at low and high spike levels using *E. indica* (*Ei*); root and shoot tissues were separately tested. The PFAS response in plant extracts was within ±30% that in neat reagent reference, suggesting suitable accuracy and moderate matrix effects (Table S4). Spike/recovery experiments were performed in triplicate at four concentration levels in control *Ei* tissues; both roots and shoots were included for testing (Table S5). PFAS recoveries were within 70−130% for most analytes/matrix/spike level combinations (Table S5).

2.7. Data Analysis. Bioaccumulation factors of root (BCF_{root}) and shoot (BCF_{shoot}) were calculated by dividing each PFAS concentration in root or shoot by the concentration in culture solution (eqs 1 and 2). Translocation factor (TF) was calculated as the ratio of shoot to root concentration using eq 3.

$$BCF_{root} = \frac{C_{root}}{C_w} \quad (1)$$

$$BCF_{shoot} = \frac{C_{shoot}}{C_w} \quad (2)$$

$$TF = \frac{C_{shoot}}{C_{root}} \quad (3)$$

where C_w is the PFAS concentration in culture solution (ng mL^{-1}) and C_{root} and C_{shoot} are the PFAS concentrations in plant roots and shoots (ng g_{dw}^{-1}), respectively.

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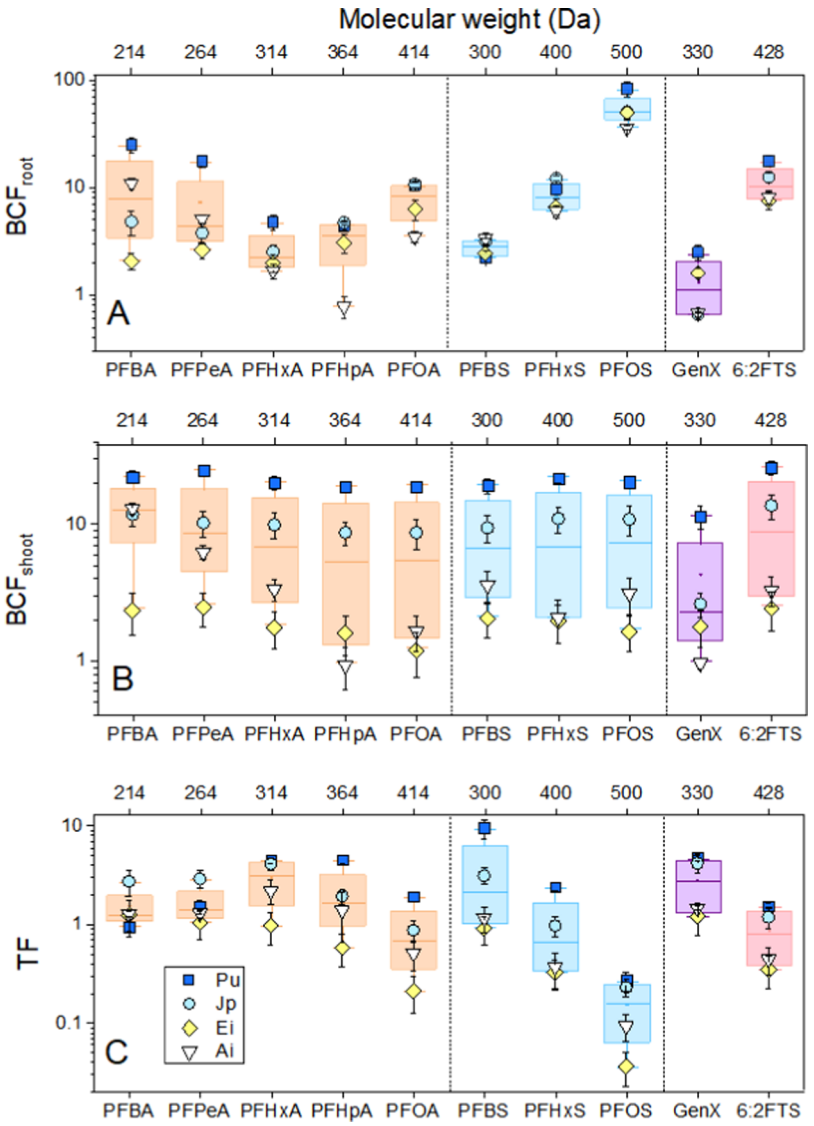

Figure 2. Bioaccumulation factor of (A) root (BCF_{root}) and (B) shoot (BCF_{shoot}) derived from the concentrations of PFAS in roots and shoot divided by their concentrations in the spiked culture solution and (C) translocation factors (TF) of plants calculated from concentrations in foliage divided by concentrations in roots. Four plants were investigated in this study, i.e., *E. indica* (*Ei*), *J. procumbens* (*Jp*), *P. urinaria* (*Pu*), and *A. indicus* (*Ai*). Error bar represents the standard deviation of five replicates.

Bioaccumulation factors were averaged at the species level, and their correlations with plants' morphological characteristics were checked using correlation analysis. We computed correlation matrices using BCF values and plant traits measured for the four species and then selected plant traits that were most correlated with BCF and TF based on the values of correlation coefficients, in order to identify key plant traits in influencing plant's bioaccumulation potentials (Figures S5−S8). Statistical analyses were performed using SPSS 22.0 software for Windows (IBM Corp., Armonk, NY) and the R statistical software (Version 4.1.1, R Core Team 2021).

3. RESULTS AND DISCUSSION

3.1. Bioaccumulation of PFAS. Figure 1 presents the bioconcentration of 10 PFAS compounds in plant roots (Figure 1A) and shoots (Figure 1B) after 21-day exposure. The concentration of PFAS in the culture solution at harvest is listed in Table S6. When the water phase concentration ranged from 116 to 356 $\mu g\ L^{-1}$, all 10 compounds were taken up by plant roots, translocated to shoots, and accumulated in plant tissues in the four plant species (Figure 1). Under the same culture conditions (e.g., temperature, humidity, light, similar aqueous PFAS concentration in the culture solution, etc.), the four plants showed different PFAS bioaccumulation behavior. The total concentration in root decreases successively as *Pu* > *Jp* > *Ei* > *Ai*; and that in shoot decreases successively as *Pu* > *Jp* > *Ai* > *Ei* (Figure 1). The highest concentrations of PFAS were detected in the roots and shoots of *Pu* (8512 and 10092 ng g_{dw}^{-1}, respectively; Figure 1).

The ability of plants to bioaccumulate PFAS is also related to their biomass. By multiplying the concentration of PFAS by the biomass of the corresponding tissue, we obtained the accumulated mass of PFAS in roots and shoots (see Figure S3). Across the four plants, *Pu* enriched the most mass of summed PFAS (∑PFAS: 36.0 μg). PFAS taken up by *Pu* are mainly stored in the shoot, attributed to the higher biomass of shoot (3.4 g per plant) than root (0.2 g per plant). As shown in Figure S3, *Ei*'s ability to bioaccumulate PFAS ranks second

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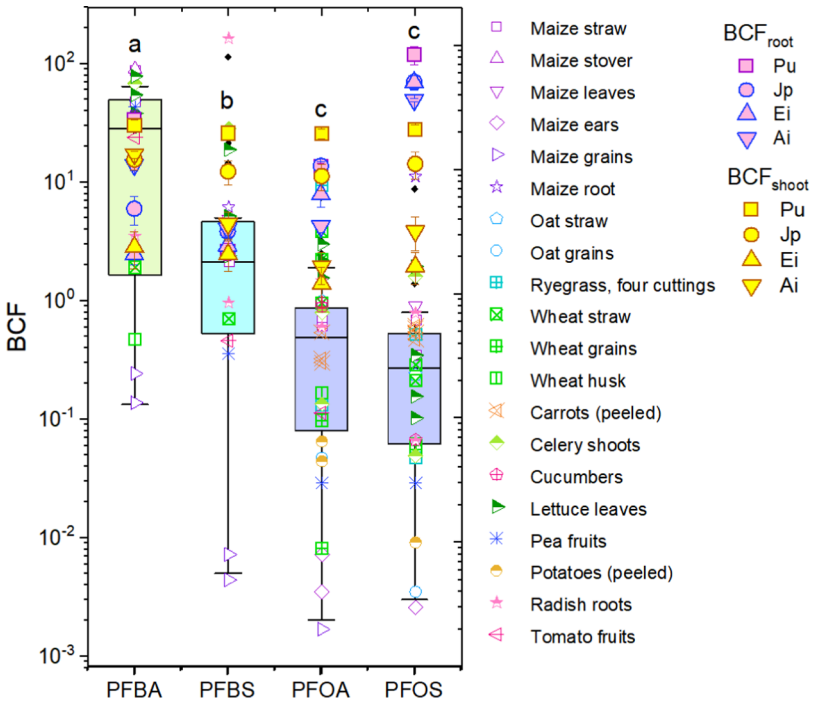

Figure 3. Summary of bioaccumulation factors (BCFs) of PFBA, PFBS, PFOA, and PFOS in various plant species and tissues, including data from the present study and literature (data cited from refs 18, 19, 21, 32−34, 42−44). Different letters above the boxes indicate significant differences at $p < 0.05$ using the least significant difference (LSD) test.

among the four plants (21.0 μg per plant), which is due to its greater biomass (14.7 g of shoot and 1.9 g of root per plant).

Our data are in line with the results of previous greenhouse exposure experiments that have shown that plants and crops can take up considerable amounts of PFAS.[18−20,23,32−36] For instance, Stahl et al. showed that perennial ryegrass accumulated PFOS and PFOA to different extents at four successive cuttings, which ranged from 408 to 7520 ng g_{dw}^{-1}.[32] In addition, Krippner et al. demonstrated maize having a high accumulation capacity for 10 perfluoroalkyl acids (PFAA) mixture, as maize straw had a total PFAS content of around 52 000 ng g_{dw}^{-1}.[33] These previous studies documented the bioaccumulation ability of plants for PFAS. However, the concentration of PFAS detected in plants can also differ by several orders of magnitude, and a direct correlation between culture medium and plant tissue concentrations was found. Blaine et al. found that all PFAA measured in lettuce leaves showed predominately linear concentration−response relationships.[19] In the logarithmic coordinate, this linear relationship held true from PFAS concentrations 50−10^5 ng L^{-1} in irrigation water. Within this range, the log concentration of PFAA detected in the plant tissues increased linearly with the PFAS level in the irrigation water without saturation.

3.2. Distribution of PFAS in Roots and Shoots. Our data reflect that plant roots were more selective for PFAS than shoots. Of the 10 compounds investigated, the accumulation of PFAS in roots of the four plants was dominated by PFOS (1900−4309 ng g_{dw}^{-1}, Figures 1A and S4A), while the distribution of various PFAS in shoots was relatively even (Figures 1B and S4B). For the PFSA series, concentrations of PFSA in roots increased with increasing chain length, which is consistent with a past hydroponic study.[20] For the PFCA series, a linear bioconcentration-chain length trend was only found in *Ei*, where the concentration of PFCA in root increased with the chain length. This trend was absent for *Pu*, *Jp*, and *Ai*, as a concave shape was seen in the bioconcentration-chain length plot, and the concentration of short-chain PFBA in root of *Pu* and *Ai* (934.6 and 438.6 ng g_{dw}^{-1}) is higher than that of PFOA (491.4 and 171.7 ng g_{dw}^{-1}) (Figure S4). The nonlinearity can be attributed to preferential adsorption of longer-chain compounds to root solids and prioritized intake of a small compound by root. Briggs et al. state that the uptake by roots of rather lipophilic chemicals is dominated by physical sorption, although they did not discriminate between adsorption and absorption.[37]

The distribution of PFAS in the shoot compartment was more evenly spread than within roots. Only the foliage of *Ai* showed that the PFCA concentrations decreased with the chain length, while this trend is absent in the foliage of the other three plants. In addition, no trend was observed for the PFSA, which was similar to past studies with hydroponic systems.[20]

3.3. Root Bioaccumulation Factors (BCF_{root}). A more comprehensive parameter with which to compare different experiments and plant species with respect to a plant's ability to accumulate pollutants could be BCF_{root} and BCF_{shoot} factors (see eqs 1 and 2). In the present study, the compounds had BCF_{root} ranges from 0.66 to 83.6 (Figure 2A). These factors indicate the tendency of plants to accumulate PFAS by root uptake. As a substitute for PFOA, the BCF_{root} of GenX was between 0.66 and 2.5, which was lower than that of PFOA (BCF_{root} of 3.5−10.5). Likewise, BCF_{root} of GenX was lower than that of PFHxA (BCF_{root} of 1.7−4.7), which has the same perfluorocarbon chain length as GenX; in addition, the BCF_{root} of 6:2 FTS was between 7.6 and 17.8, which was also lower than that of PFOS (BCF_{root} of 36.0−83.6). This indicates that GenX and 6:2 FTS are less likely to migrate to plant roots than legacy C8 PFOA and PFOS.

As shown in Figure 2A, median BCF_{root} of PFCA [PFBA(C3)-PFOA(C7)] homologues of the four plants

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show a concave shape with the chain length. In contrast, the BCF_{root} increases with the chain length for PFSA homologues (C4, C6, and C8) ($F = 23.5$, $p < 0.01$). This is consistent with previous plant uptake studies where increased BCF_{root} with increasing chain length for PFSA and a U-shaped dependency with minima for PFHxA (C5) or PFHpA (C6) for PFCA homologues were observed.[20,38,39] These findings indicate that uptake into root tissue is increasingly inhibited with increasing carbon-chain length, e.g., due to increasing molecule size, while adsorption becomes stronger with increasing carbon-chain length.[20,38,39] Therefore, root uptake seems to be the dominant accumulation process for PFBA and PFPeA, while adsorption seems to be dominant for longer-chain compounds such as PFHpA to PFOA.[20] In addition, Guelfo and Higgins reported that short-chained PFAS such as PFBA have also shown a strong sorption tendency to organic matters in soils, which is comparable to that of PFOA.[40] It was explained by either a different sorption mechanism relatively more important for short-chained PFCA, such as ion exchange, or a subset of sorption sites that are only approachable for smaller molecules due to steric effects.[40]

3.4. Shoot Bioaccumulation Factor (BCF_{shoot}). For the PFCA series, values of BCF_{shoot} varied from 0.95 to 25.7 across the four plant species, and the values decreased with the increase of perfluorocarbon chain length (Figure 2B). However, a one-way analysis of variance (ANOVA) showed that the BCF_{shoot} values of PFCA series (C3−C7) were not significantly different ($F = 0.24$, $p = 0.91$). Among the PFSA series, only *Ei* showed a downward trend in the BCF_{shoot}-chain length, while in the other three plants, PFBS, PFHxS, and PFOS had similar BCF_{shoot} values (Figure 2B) ($F = 0.0058$, $p = 0.99$). Based on the reviewed literature for plants (Figure 3), BCF values differed significantly among PFAS substances ($F = 12.67$, $p < 0.001$); the overall median BCF (various plant species and tissues) is higher for short-chain perfluoroalkyl acids (i.e., PFBA and PFBS) compared to the long-chain analogs (i.e., PFOA and PFOS), although a statistically significant difference in BCF values between PFOA and PFOS was not detected (Figure 3; $t = 0.548$, $p = 0.29$).

GenX, a current-use alternative to PFOA, showed a lower BCF_{shoot} compared to that of PFOA for *Jp* and *Pu* species ($\Delta BCF_{shoot} = -7.9$ and -6.3, respectively, $p < 0.05$). The BCF_{shoot} of GenX is similar to that of PFOA in *Ai* and *Ei* species ($\Delta BCF_{shoot} = -0.7$ and 0.6, respectively, $p = 0.21$). Chen et al. compared the accumulation and toxicity of GenX and PFOA in the model plants *Arabidopsis thaliana* and *Nicotiana benthamiana*. Both plants showed a reduction in biomass and root growth following PFOA or GenX exposure. Higher BCF (based on biomass of the whole plant) of PFOA than that of GenX was noticed in both plants.[45] Moreover, in our study, BCF_{shoot} was slightly lower for GenX than PFHxA in *Pu*, *Jp*, and *Ai* ($t = -2.84$, $p < 0.05$). On the other end, BCF_{shoot} values for 6:2 FTS were higher than PFOS across all of the four plant species ($t = 1.62$, $p < 0.05$). While 6:2 FTS was found to be less bioaccumulative than PFOS in many higher-order biotic organisms, this is not necessarily the case for vegetation samples. Gobelius et al. reported field-based soil-to-vegetation BCFs higher (by 17×) for 6:2 FTS compared with PFOS.[24] 6:2 FTS also showed higher (3−6×) bioaccumulative potential than PFHxS/PFOS in a hydroponic culture study with pumpkin plants (*Cucurbita maxima*).[46]

Compared with existing studies (Figure 3), both BCF_{shoot} and BCF_{root} of the four urban spontaneous plants are high compared to the accumulation factors reported elsewhere (mostly cereals, vegetables, and fruits). Species like *J. procumbens* (commonly known as water willow) from the Acanthaceae family showed a high PFAS bioaccumulation potential; more species of the same family or genus that are currently widespread around the globe are also expected to have similar potentials. This could contribute to an increase in global awareness of the phytoremediation values of urban spontaneous plants. However, it should be noted that previous studies were conducted in the soil culture medium, and no selective retardation occurred before PFAS reached the root surface. Comparing hydroponic cultures to soil cultures, the interaction between PFAS and soil may lead to a decreasing availability of plants. Therefore, BCF derived from hydroponic cultures may not be directly comparable with BCF from soil or pore water. PFAS uptake from spike experiments (either to soil or hydroponics) may also differ from real contaminated soils aged in the field.[19]

Felizeter et al. evaluated whether mechanistic understanding of plant uptake of 13 PFAS derived from hydroponic experiments can be applied to soil systems.[47] It was found that the foliage/pore water concentration ratios in the lysimeter were similar to the foliage/water concentration ratios from the hydroponic experiment. In contrast, the root/pore water concentration ratios in the lysimeter were 1−2 orders of magnitude lower than in the hydroponic study for PFAS with C6 or more perfluorinated carbons. Hydroponic studies can provide a good quantitative measure of PFAS transfer from soil to foliage if one accounts for soil/pore water partitioning and differences in transpiration rate.[47]

3.5. Foliage to Root Bioaccumulation Factors (TF). As shown in Figure 2C, median log TF (box plots) of PFCA exhibits an arch trend with chain length (from 0.8 to 2.7). Specifically, in *Ei*, the TF values decreased monotonously with the chain length, while in the other three plants, the TF values increased from PFBA to PFHxA and then declined from PFHpA to PFOA (Figure 2C). Similarly, Zhao et al. (2014) explored how wheat (*Triticum aestivum L.*) accumulated 11 PFAS from soil, the highest TF was noticed for PFHxA (0.60−1.9) instead of the shortest investigated PFCA, PFPeA (0.39−0.58) in wheat.[48] In contrast, the TF values of PFSA decreased exponentially with increasing chain length (Figure 2C) ($F = 4.67$, $p < 0.05$).

For the alternative PFAS, the TF value of GenX across the four study plants was significantly higher than that of PFOA ($\Delta TF = 0.9-3.2$, $p < 0.05$). Likewise, the TF value of 6:2 FTS was significantly higher than that of PFOS ($\Delta TF = 0.3-1.3$, $p < 0.05$). Similar patterns of TF decreasing with increasing chain length are also observed in a previous study.[20] Such behavior can be attributed to sorption of the chemicals to plant tissue, which impedes the translation of PFAS from root to shoot. It is documented that sorption of PFAS from water to organic material generally increases with chain length;[49,50] this could lead to a stronger retention of the compounds in the root section and thereby reduce transport of the longer-chain chemicals. Further, passing across the Casparian strip, which is an obstacle for the translocation between roots and shoots via vascular tissue, was thought to be the main pathway of PFAS uptake.[41−43] It was reported that the ability of PFAS to cross the Casparian strip may decrease with chain length.[26] More specifically, even though higher concentrations of the longer-chain PFAS are transferred to roots (Figure 1), the majority of these chemicals may not cross the Casparian strip and

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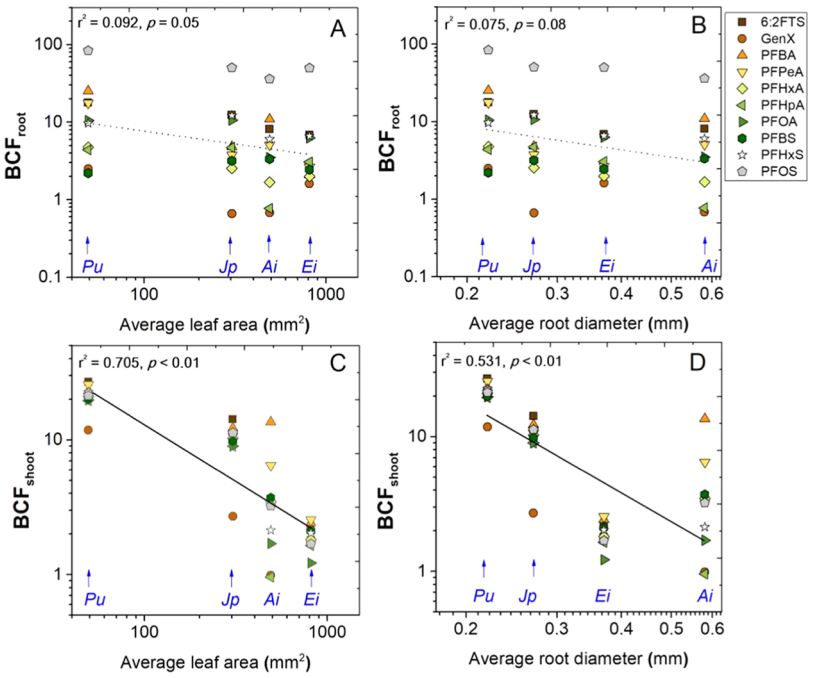

Figure 4. Bioaccumulation factors of root (BCF_{root}) as a function of (A) plant leaf area (mm^2) and (B) average root diameter (mm); bioaccumulation factors of shoot (BCF_{shoot}) as a function of (C) plant leaf area (mm^2) and (D) average root diameter (mm). Four urban spontaneous plants, i.e., *E. indica* (*Ei*), *J. procumbens* (*Jp*), *P. urinaria* (*Pu*), and *A. indicus* (*Ai*) were investigated. The solid lines indicate a significant relationship at $p < 0.05$, and the dashed lines show marginal significance at $p < 0.10$. The solid and dashed lines and regression coefficients were determined using the average values of BCF for all PFAS ($n = 40$). Regression analysis was performed using *R* (Version 4.1.2; R Core Team).

therefore would not be available for translocation to stems and foliage.

3.6. Correlation with Leaf Area and Root Diameter. Bioaccumulation factors (BCF_{root} and BCF_{shoot}) showed consistent trends in response to plants' foliar and root characteristics for 10 PFAS species (Figure 4). Among the list of plant morphological traits, average leaf area and root diameter were among the most correlated traits with BCFs (Figures S5−S8). Inspection of Figure 4 shows that the correlation between BCF_{root} and plant morphological traits varied across PFAS types. In contrast, the correlation between BCF_{shoot} and plant morphological traits was more consistent of all PFAS investigated. In general, urban plant species with smaller leaves and finer roots (i.e., *Pu* and *Jp*; Table S2) had higher BCF_{root} and BCF_{shoot} values than species with larger leaves and coarse roots. While this correlation was strong for BCF_{shoot} (Figure 4C,D), the BCF_{root} values may be influenced by more factors other than plants' foliar and root traits given the relatively weak trends detected (Figure 4A,B). In particular, the correlations between BCF_{root} values and plant morphological traits showed a clear deviation for PFOS compared with other PFAS. This could be due to the longer-chain length of PFOS (C8) that leads to greater sorption on the root surfaces.

Plant species with small leaves often have shorter leaf lifespans and higher relative growth rates.[51] Because for many plants, relative growth rates are highly correlated with rates of photosynthesis, respiration, and nutrient uptake from environments,[52] it could result in a faster and higher level of PFAS accumulation in weeds via transport in the vascular system. This could be true for the annual herb *Pu* as suggested by many densely distributed small pores on its leaves (Figure S2J). In the meantime, plants with abundant fine roots could benefit from a larger surface area for resource uptake, especially when it comes to the case of hydroponic cultivation as in this study. Further, the consistent trends in bioaccumulation factors observed for both traditional PFAS and alternatives in response to plants' foliar and root characteristics could provide knowledge to plant selection criteria for phytoremediation of PFAS-contaminated sites and may serve as an interesting area for further investigations.

4. SIGNIFICANCE

This study assessed the bioaccumulation potential of legacy PFAS, a fluorotelomer sulfonate, and an emerging fluorinated ether (GenX) in spontaneous urban plants. All 10 compounds were taken up by plant roots, translocated to shoots, and accumulated in the tissues of the study plants. Among the four investigated species of plants, *P. urinaria* demonstrated the highest concentrations of ∑PFAS in the roots and shoots and also enriched the greatest mass of ∑PFAS. Bioaccumulation factors (BCF_{root} and BCF_{shoot}) showed consistent trends in response to plants' foliar and root characteristics for PFAS. Leaf area and average root diameter were the most correlated traits with BCF. Plant species with smaller leaves and finer roots thus displayed higher BCF values than species with larger leaves and coarse roots.

We also provided bioaccumulation and translocation factors under a controlled greenhouse exposure setting using hydroponic experiments. This facilitated comparisons across compounds compared to field conditions where different exposure concentrations may confound results due to PFAS concentration-dependent relationships—observed for biofilm[53] and other biological models.[54] Legacy C8 PFAS, such as PFOS, display higher BCF than short-chain alternatives (such as PFBS) in higher-order biota organisms (e.g., fish, mammals), while a reverse trend was generally found for

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vegetation, in that short-chain PFAS were found to translocate to a greater extent.[55] Short-chain PFAS alternatives are also problematic as they are more mobile and difficult to remove by conventional water treatment systems.[56] As a major PFAS present in technical cleaning products[57] and contemporary aqueous film-forming foam (AFFF) (either as component or major degradation intermediate),[58] 6:2 FTS also shows higher accumulation and translocation potential in vegetation than PFOS. The findings derived here from a controlled greenhouse study using hydroponic experiments are in agreement with field monitoring data at an AFFF-contaminated site, where 6:2 FTS was dominant in vegetation despite high PFOS levels in soil and groundwater.[24] A previous monitoring study found high levels of GenX in/on grass and leaves in the vicinity of a fluoropolymer manufacturing plant[15] but could not clarify whether internal bioaccumulation had occurred or reflected superficial deposits from atmospheric industrial emissions. Furthermore, we demonstrate that the PFOA replacement HFPO-DA (GenX) can bioaccumulate at significant levels in urban spontaneous vegetation. Future studies could explore other potential factors in absorbing PFAS, such as root protein and lipid content; field studies could also evaluate the role of phytoremediation involving urban weeds, constructed wetlands,[59] and trees[24] as valuable options to alleviate contamination with current-use PFAS.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestengg.1c00423.

> Plant foliar and root traits characterization (Text S1); details of the PFAS investigated (Table S1); characteristics of urban spontaneous plants (Table S2); quantitative analytical method (Table S3); matrix effect and spike recovery of extraction method (Tables S4 and S5); concentration of PFAS in the culture solution (Table S6); molecular structure of PFAS investigated (Figure S1); photos of the individuals of weeds (Figure S2); mass and concentration of PFAS accumulated in weeds (Figures S3 and S4); and correlation matrix showing the correlation coefficients between bioaccumulation factor and plant root traits (Figures S5−S8) (PDF)

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Notes

The authors declare no competing financial interest.

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