Science of the Total Environment 906 (2024) 167578

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Bioaccumulation of PFASs in cabbage collected near a landfill site in China: Laboratory and field investigations

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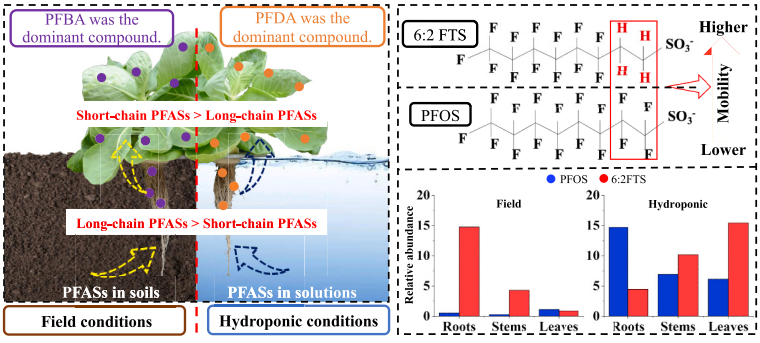
HIGHLIGHTS

- Long-chain PFASs were remained primarily in roots, while short-chain PFASs were readily being translocated.
- PFASs distribution patterns in cabbage organs differed under field and hydroponic conditions.
- PFBA and PFDA were the dominant compounds in field and hydroponics, respectively.
- 6:2 FTS, an alternative of PFOS, had higher translocation factors among cabbage organs.
- Short-chain and emerging alternatives can pose greater risk to human health than legacy PFASs.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Shuzhen Zhang

Keywords:
PFASs
Cabbage root
Bioaccumulation
Bioconcentration factor
Hydroponic

ABSTRACT

Previous studies found that the bioaccumulation of PFASs in vegetables poses potential risks to the health of residents in local areas near landfills in China. Therefore, our study investigated the uptake of perfluoroalkyl and polyfluoroalkyl substances (PFASs) and their accumulation and distribution in cabbage roots, stems, and leaves under both field and laboratory hydroponic conditions. It was found that the sum of concentration of 15 PFASs (designated as Σ_{15}PFASs) in roots, stems, and leaves ranged from 24.8 to 365 ng/g, 49.2 to 204 ng/g, 11.9 to 115 ng/g, respectively, in the order of roots > stems > leaves, which were generally higher than the range in soil samples (6.07–63.91 ng/g). The dominant compounds in cabbage were PFBA and PFDA in field and hydroponic samples, respectively. The hydroponic experimental results revealed that the sum concentration of 10 PFASs (designated as Σ_{10}PFASs) was the highest in roots, and PFDA was the dominant compound in different cabbage fractions. Bioconcentration factors of short-chain PFBA, PFPeA, and PFBS in hydroponics followed the trend of leaves > stems > roots, indicating that they were readily transported from roots to stems, and then to leaves, with the majority stored in leaves at abundance levels of 53 %, 71 %, and 60 %, respectively. Additionally, the

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https://doi.org/10.1016/j.scitotenv.2023.167578
Received 26 July 2023; Received in revised form 29 September 2023; Accepted 2 October 2023
Available online 4 October 2023

much higher concentration factor for 6:2 FTS in leaves suggested a higher potential health risk than PFOS in terms of dietary consumption of cabbage leaves.

1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are a class of synthetic organic compounds that have been widely used for many decades in a myriad of industrial and commercial applications due to their chemical and thermal stability, such as in surfactants, food packages, coatings, cosmetics and aqueous firefighting foams (AFFFs) (Buck et al., 2011; Paul et al., 2009). The widespread use of these products has led to the ubiquitous presence of PFASs in the environment, wildlife, and humans (Dalahmeh et al., 2018; Vuong et al., 2018). PFASs pose severe threat to human health owing to their persistency in the environment, toxicity, and bioaccumulation (Schwanz et al., 2016; Sunderland et al., 2019). As a result, perfluorooctane sulfonic acid (PFOS) and its salts, and perfluorooctanoic acid (PFOA) and its related compounds, were listed as "persistent organic pollutants" under the Stockholm Convention in 2009 and 2019, respectively (UNEP, 2019, 2009). Due to the restrictions on PFOS and PFOA, there has been a shift from traditional PFASs to fluorinated alternatives, such as per- and polyfluoroalkyl ether carboxylic (PFECAs) and sulfonic acids (PFESAs) (UNEP, 2012). These emerging substances, e.g., hexafluoropropylene oxide dimer and trimer acids (HFPO-DA and HFPO-TA), ammonium 4,8-dioxa-3H-perfluorononanoate (ADONA), and chlorinated polyfluorinated ether sulfonic acid (6:2 Cl-PFESA) have been frequently detected in different environmental media (Pan et al., 2018), raising further concerns worldwide.

PFASs can enter the soil through a variety of pathways, including irrigation via PFAS-contaminated water, land application of biosolids, release from industrial manufacturing, leaching from landfill waste and pesticide applications (Blaine et al., 2014; Gallen et al., 2016; Liu et al., 2016; Piva et al., 2023; Taniyasu et al., 2013; Xu et al., 2021a). Landfill has been identified as a major environmental source of PFASs (Tian et al., 2018), and it was suggested that further studies are needed to assess the holistic health risk of PFASs for the local residents around landfills, taking into account food intake (Domingo, 2012; Xu et al., 2021a). The dietary exposure studies have shown that vegetables are one of the most critical food categories for exposure to PFOA and PFHxA, with up to 69 % of total exposure coming from vegetables (Klenow et al., 2013). The uptake of PFASs by plant roots from soil pore water is considered to be the main pathway into the terrestrial food web (He et al., 2023; Krippner et al., 2014; Liu et al., 2016).

Previous studies concluded that the PFASs health risks for local residents through vegetables (e.g. cabbage) consumption were alarming, and the vegetables in local area near a landfill site were suggested not suitable for consumption (Xu et al., 2021b). Furthermore, it has been reported that the emerging alternatives of PFASs, including 6:2 Cl-PFESA, 6:2 fluorotelomer sulfonate (6:2 FTS) and perfluorophosphinates (C6/C6 and C8/C8 PFPiAs), were more easily accumulated in roots and shoots of maize (*Zea mays* L.), wheat (*Triticum aestivum* L.), pumpkin (*Cucurbita maxima* L.) and soybean (*Glycine max* L. Merrill) (Zhou et al., 2020). Both hydroponic and pot experiments showed that translocation factor values decreased with the increasing PFAS chain length, or the increase of the PFASs hydrophobicity (log K_{OW}) (Felizeter et al., 2012; Krippner et al., 2014; Mei et al., 2021; Tuan et al., 2020; Wen et al., 2014). Hydroponic experiments also showed that long-chain perfluoroalkyl acids (PFAAs) were retained in crop roots, and short-chain PFAAs can transport upwards to leaves, stems, and fruits (Felizeter et al., 2014). PFASs with smaller molecular size and lower hydrophobicity demonstrated higher translocation tendency in plants (Liu et al., 2017). However, some studies showed that wheat, tomato or pea root-soil concentration factors were not apparently correlated with PFAA carbon chain length (Blaine et al., 2014; Wen et al., 2014). This could be attributed to the fact that the varying sorption for PFAAs by soils determines the bioavailability to plant root uptake; strongly sorbed PFAAs by soils manifested a lower accumulation in plant roots (Mei et al., 2021). Moreover, the bioaccumulation of PFAS via plant could also alter their fate and transport in various environments (Adu et al., 2023). Therefore, the bioaccumulation and distributions of legacy PFASs and their emerging alternatives among different plant organs nearby landfills warrant further study, in particular those studies involving field data.

Cabbage is a widely cultivated and essential leafy vegetable, and the concentration of PFASs in its edible portion is strongly associated with the health risks to PFASs (Xu et al., 2021b; Zhang et al., 2020). To further investigate the distributions of PFASs and their translocation among different plant fractions due to the exposure to a landfill site, cabbage samples were collected from the investigation field. Additionally, we conducted a hydroponic experiment to further elucidate the root uptake of PFASs and their translocation within cabbage. The objectives of this study were to (1) investigate and explore the impact of landfill on the concentration and distribution patterns of PFASs in cabbage root, stem, and leaf, (2) quantify the bioaccumulation of legacy PFASs and their emerging alternatives, and (3) provide additional insights to the translocation mechanism of legacy PFASs and their emerging alternatives among different plant fractions. The results could provide additional information to evaluate the risk of PFASs associated with vegetable exposure to landfill sites and potential impact to human health.

2. Materials and method

2.1. Sampling design and collection

Cabbage (*Brassica oleracea* L. var. *capitata* L.) samples were collected from the rosette stage to the heading stage in May 2018 at 14 locations within 5 km of a landfill site, which is located in a valley surrounded by mountains on the three sides, in Hangzhou city, China (Fig. 1). The cabbage roots, stems, and leaves were separated, wrapped in aluminum foil and stored in clean paper bags. Meanwhile, the associated surface soil (0–20 cm) around the roots of each plant in each site was collected with a small stainless-steel spatula, and mixed into one composite sample. Details of samples collection are described in the Supporting Information (SI).

2.2. Plant exposure experiments

To further investigate the bioaccumulation and translocation mechanisms of PFASs among different cabbage fractions, a greenhouse hydroponic experiment was conducted to evaluate PFAS uptake and transport in cabbage. This experiment allowed excluding the impacts from the sorption by soils and uptake of airborne PFASs effects (Felizeter et al., 2012; Li et al., 2018). The cabbage plants were grown hydroponically in a greenhouse located at Nanjing, China. The exposure experiments were performed from May 29 to August 19, 2021, and samples were collected at the heading stage on August 19th, 2021, the last day of the exposure experiment. Plants were pre-grown in soils until the development of 4–6 leaves, then the soil adhering to seedlings roots was washed off using distilled water and seedlings were transferred to hydroponic glass pots. The pots were covered by a floating board, and wrapped around with light impermeable thermal insulation materials to prevent potential algal growth. Each floating board was drilled with five holes for the accommodation of cabbage plants, and each plant was held within the hole with a PP (Polypropylene) sponge so that only the plant roots were exposed to the solution.

The plants were first grown for 5 days in 8 L of half-strength Hoagland's nutrient solution to adapt to the hydroponic system (Felizeter et al., 2014). After 5 days, the pots were replaced with 8 L of full-strength Hoagland's nutrient solution. The exposure experiment included four different exposure concentrations of the PFAS mixture and one control without PFASs. The PFAS mixture consisted of 10 representative compounds, and the detection ratios of PFASs in the field samples, their chemical structures, and toxicity were considered in the selection of 10 PFASs. Specifically, the 10 PFASs include PFOA and PFOS, the two legacy compounds that have been studied most extensively. Due to the restrictions on PFOA and PFOS, there has been a shift from traditional PFASs to emerging alternatives. Therefore, the emerging alternatives of PFOA, HFPO-DA and HFPO-TA, and those of PFOS, F-53B and 6:2FTS were selected. In addition, the short-chain analogues of PFOA and PFOS, including PFBA, PFPeA, and PFBS, have also been used as their alternatives, hence, were included in the 10 selected PFASs. Finally, for the representative of long-chain PFAS compounds, PFDA (C10) was detected in most cabbage field samples, with much higher detection ratio than those of with C11 or C12. Therefore, PFDA was chosen to as the model compound to evaluate the bioaccumulation of long-chain PFAS chemicals.

Previous studies have conducted initial PFAAs hydroponic treatments at concentrations of 10–1000 ng/L (Felizeter et al., 2014), but some studies have shown much higher concentrations of PFASs in the field (Liu et al., 2016). Thus, the cabbage plants were grown in hydroponic solution with equal concentration for each of the 10 PFASs, nominally being 100, 1000, 5000, and 10,000 ng/L (T1–T4, respectively). Blank samples were incubated in the greenhouse in consistent with other pot plants. Every 5 days, 300 mL of Hoagland's nutrient solution was added to the pots, and each pot was randomly placed in the greenhouse and the positions were periodically adjusted during the experiment.

2.3. Standards and reagents

A total of 17 PFASs, including 9 perfluoroalkyl carboxylic acids (PFCAs), 3 perfluoroalkyl sulfonic acids (PFSAs), and 5 emerging alternatives were identified and quantified. The chemical formulas and Kow of the 17 PFASs are provided in Table S3. Among the 17 PFASs, 10 PFASs, including 4 representative PFCAs, 2 representative PFSAs, and 4 emerging alternatives, were selected in the hydroponic experiment. Native and isotope-labeled PFASs were purchased with >98 % purity from Wellington Laboratories (Guelph, Ontario, Canada). Details of the standards and reagents used are described in the SI.

2.4. Extraction and analyses

The PFAS extraction methods for the cabbage, soil, and nutrient solution samples extraction were described in previous studies (Felizeter et al., 2012; Xu et al., 2021b). The PFASs concentrations were determined using an AB Sciex 5500 TripleQuad LC-MS/MS at negative electrospray ionization mode. Detailed information of extraction and instrumental analysis are provided in the SI.

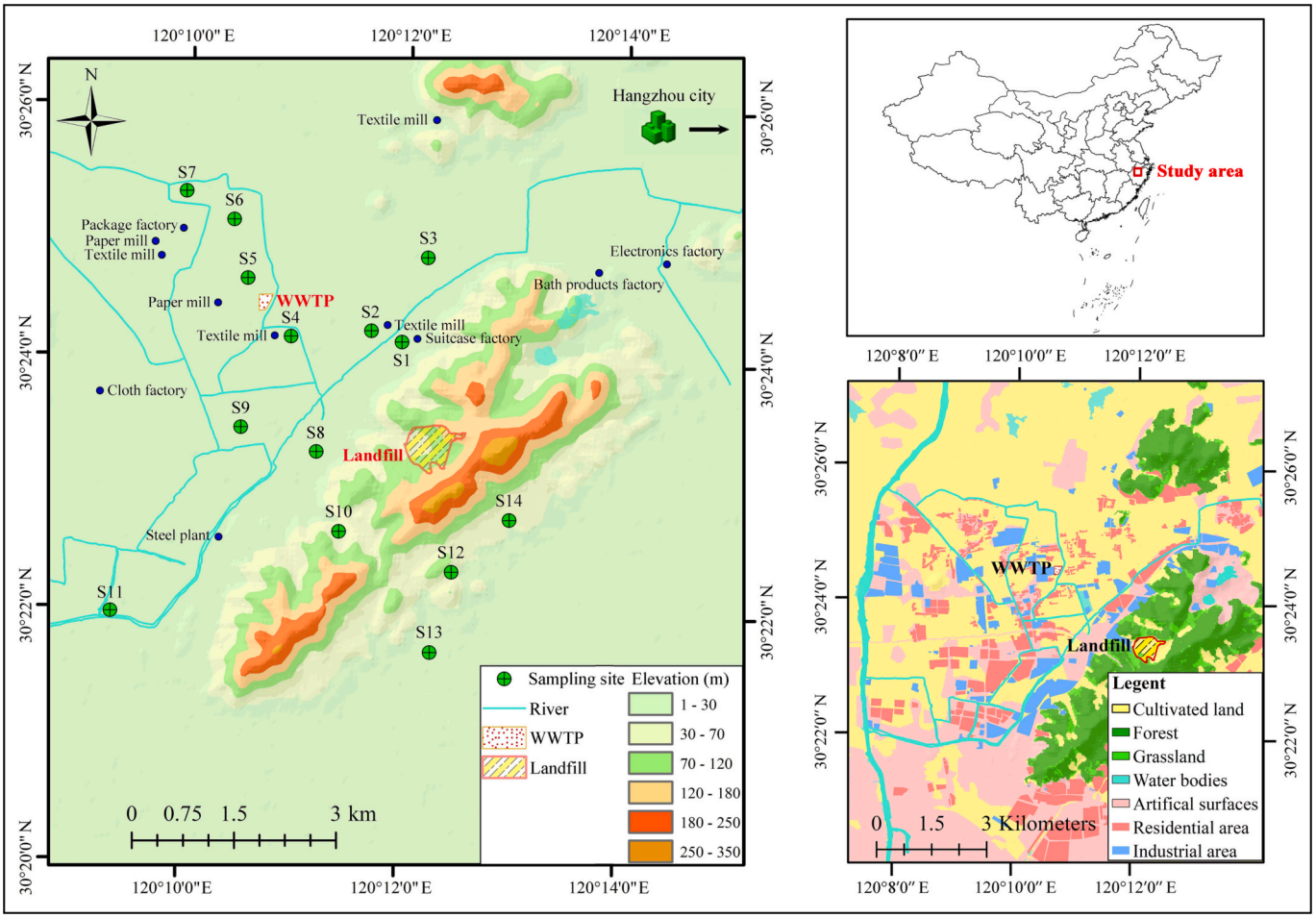

Fig. 1. Sampling locations for cabbage plants around a landfill located in the northwest of the City of Hangzhou, Zhejiang Province, China.

2.5. Quality assurance and quality control (QA/QC)

To avoid the cross-contamination, plant and soil samples were stored in triple-sealed PP bags, and nutrient solution samples were stored in PP bottles. Each batch of samples included field, transport, procedural and solvent blanks to determine if external contamination occurred during the sampling, transportation, extraction and analytical stages. The internal standard calibration curve was based on 10 calibration points (0.1, 0.2, 0.5, 1, 2, 5, 10, 20, 50 and 100 ng/mL) of 1 mL native standards, and spiked with the internal standard of 5 ng in 1 mL, resulting in a final concentration of 5 ng/mL, which was prepared for quantification of the individual PFAS with coefficients ($R^2 > 0.99$) for each target analyte (Table S2). The limit of detection (LOD) and limit of quantification (LOQ) were calculated with a signal-to-noise (S/N) ratio of 3 and 10, respectively, and Table S2 provided the LOD and LOQ for each PFAS. To determine the recovery for each target PFAS, each matrix was spiked with 1 mL of the native standard solution (20 ng/mL). The matrix spike recoveries (MSRs) ranged from 62.9 ± 2.79 % to 99.4 ± 1.74 % for soil, 62.2 ± 3.51 % to 112 ± 12.9 % for plants. More detailed information of QA/QC is provided in the SI.

2.6. Bioconcentration factors (BCF) and translocation factors (TF) calculations

The uptake and bioaccumulation potential of PFASs from the environment by cabbage was evaluated using BCFs (Gredelj et al., 2020b; Torralba-Sanchez et al., 2017). The BCFs of the field samples were expressed on a dry weight (DW) basis as the ratio of the concentration for each PFAS in cabbage roots to the concentration in soil. The BCFs of hydroponic exposure experiment samples were defined as the ratio of the concentration of each PFAS in cabbage fractions to the concentration in the nutrient solution:

$$\mathrm{RCF} = \frac{\text{PFAS concentration in plant root (ng/g DW)}}{\text{PFAS concentration in soil (ng/g DW) or nutrient solution (ng/mL)}} \quad (1)$$

where RCF represents the root concentration factor (Felizeter et al., 2012).

Following the uptake by roots, PFASs can transport from roots to aboveground tissues such as stems, and leaves via xylem or phloem, which can be expressed by translocation factors (TF) (Xu et al., 2022). The TFs of roots-to-stems (expressed as RSTF) are calculated as the mean concentrations in stems divided by the mean concentration in roots, and TFs of stems-to-leaves (expressed as SLTF) are calculated as the mean concentrations in leaves divided by the mean concentration in stems (Felizeter et al., 2012).

2.7. Statistical analysis

Statistical analyses were performed using SPSS Statistics 25.0, OriginPro 9.0, and Excel 2018. To analyze the correlation of BCFs and the number of carbon atoms of perfluoroalkyl chain, BCFs were $\log_{10}$-transformed (Gredelj et al., 2020a). Independent samples *t*-tests were used to determine the differences in PFAS concentrations in soil, nutrient solution, and plant organs, as well as BCFs of individual PFASs in different plant organs. ANOVA was used to determine the significance of differences in BCFs of PFASs in different plant tissues. The Shapiro-Wilk test was used to assess the data normality.

3. Results and discussion

3.1. Occurrence and bioaccumulation of PFASs in cabbage organs collected from the field

3.1.1. PFASs distributions among different organs

To facilitate the discussion on the occurrence and bioaccumulation of PFASs in different cabbage organs, the composition profiles of PFASs in soil are described briefly here, with the details published in Xu et al. (2021b) and Table S3. Among the 17 PFASs, fifteen were detected at concentrations above the LODs, with the exception of HFPO-TA and ADONA. The sum concentrations of PFASs (Σ_{15}PFASs) in the soil samples were within the range of 6.07–63.91 ng/g. The dominant PFAS in soil was PFBA at the concentration of 9.34 ± 6.58 ng/g, followed by HFPO-DA, F-53B, 6:2 FTS, PFPeA, and PFOS, with concentrations of 5.83 ± 10.19, 4.73 ± 8.82, 3.08 ± 6.19, 2.45 ± 2.19 and 2.26 ± 5.31 ng/g, respectively. The detected concentration of HFPO-DA, F-53B, and 6:2 FTS were surprisingly high in the soil, much higher than their legacy PFOA and PFOS, being ranked the second, third, and fourth in this study, respectively. The average concentrations of PFHxS, PFBS, PFHpA, PFDoDA, PFNA, PFUnDA, PFDA, PFHxA, and PFOA in soils were relatively low, at concentrations of <1 ng/g.

The Σ_{15}PFASs concentration in cabbage roots, stems, and leaves were in the ranges of 24.8–365, 49.2–204, 11.9–115 ng/g (Fig. 2a–c), respectively, much higher than the concentration range measured in soil samples (6.07–63.91 ng/g) reported by Xu et al. (2021b). The concentration trend generally followed the order of roots > stems > leaves. The highest Σ_{15}PFASs concentrations was detected at S4, located next to the wastewater treatment plant, suggesting that local point sources might also contributed to the accumulation of PFASs in cabbage, consistent with previous conclusions (Xu et al., 2021b). In the roots, PFBA (45.9 ng/g) was the dominant compound with a contribution of 35.3 % to the Σ_{15}PFASs, followed by 6:2 FTS (33.5 ng/g, 25.7 %), PFPeA (31.0 ng/g, 23.8 %), HFPO-DA (5.51 ng/g, 4.23 %), PFHxA (5.25 ng/g, 4.03 %), and F-53B (3.56 ng/g, 2.74 %) (Fig. 2a and d). The trend in the cabbage stem samples followed the order of PFBA (50.6 ng/g, 46.9 %), PFPeA (37.5 ng/g, 34.6 %), HFPO-DA (5.97 ng/g, 5.5 %), PFHxA (5.63 ng/g, 5.2 %), 6:2 FTS (3.57 ng/g, 3.3 %), and PFHpA (3.01 ng/g, 2.8 %) (Fig. 2b and d). For leafy samples, the top three dominant PFASs were short-chain PFCAs, including PFBA (22.0 ng/g, 58.82 %), PFPeA (9.77 ng/g, 26.09 %), and PFHxA (1.46 ng/g, 3.89 %), followed by F-53B (1.43 ng/g, 3.82 %), HFPO-DA (1.24 ng/g, 3.32 %), and PFHpA (0.46 ng/g, 1.22 %) (Fig. 2c and d).

Much higher average relative abundances of the four short-chain PFCAs (C4–C7; PFBA, PFPeA, PFHxA, and PFHpA) were observed in stems (89.3 %) and leaves (90.0 %) compared to roots (65.7 %). In addition, higher average relative abundances of long-chain PFCAs (C8–C12; PFOA, PFNA, PFDA, PFUdA, and PFDoA) were observed in roots (1.0 %) compared to those in stems (0.7 %) or leaves (0.7 %), consistent with observations in previous studies (Zhang et al., 2020). These observations suggested that long-chain PFCAs (C8–C12) mostly accumulated in roots, whereas short-chain PFCAs were more likely to transfer to stems and leaves (Gredelj et al., 2020a).

The emerging alternatives HFPO-DA, F-53B, and 6:2 FTS were detected in all of the cabbage root, stem, and leaf samples, with their concentration generally exceeding those of the legacy PFOA and PFOS. The HFPO-TA was detected in 64.2 % of roots samples with an average concentration of 0.26 ng/g. Concentration of F-53B and 6:2 FTS in stem samples was 0.84 ng/g and 3.57 ng/g, respectively, lower than those detected in roots. The lower concentration and contribution of 6:2FTS and F-53B (two alternatives of PFOS) detected in stems compared to root samples suggest that stem could be a duct for the translocation of PFASs. Surprisingly, the ranking of F-53B in the leaf samples was higher than that in the stem samples, indicating the presence of a second source of F-53B, e.g., wet/dry depositions of particulate matters (Liu et al., 2016).

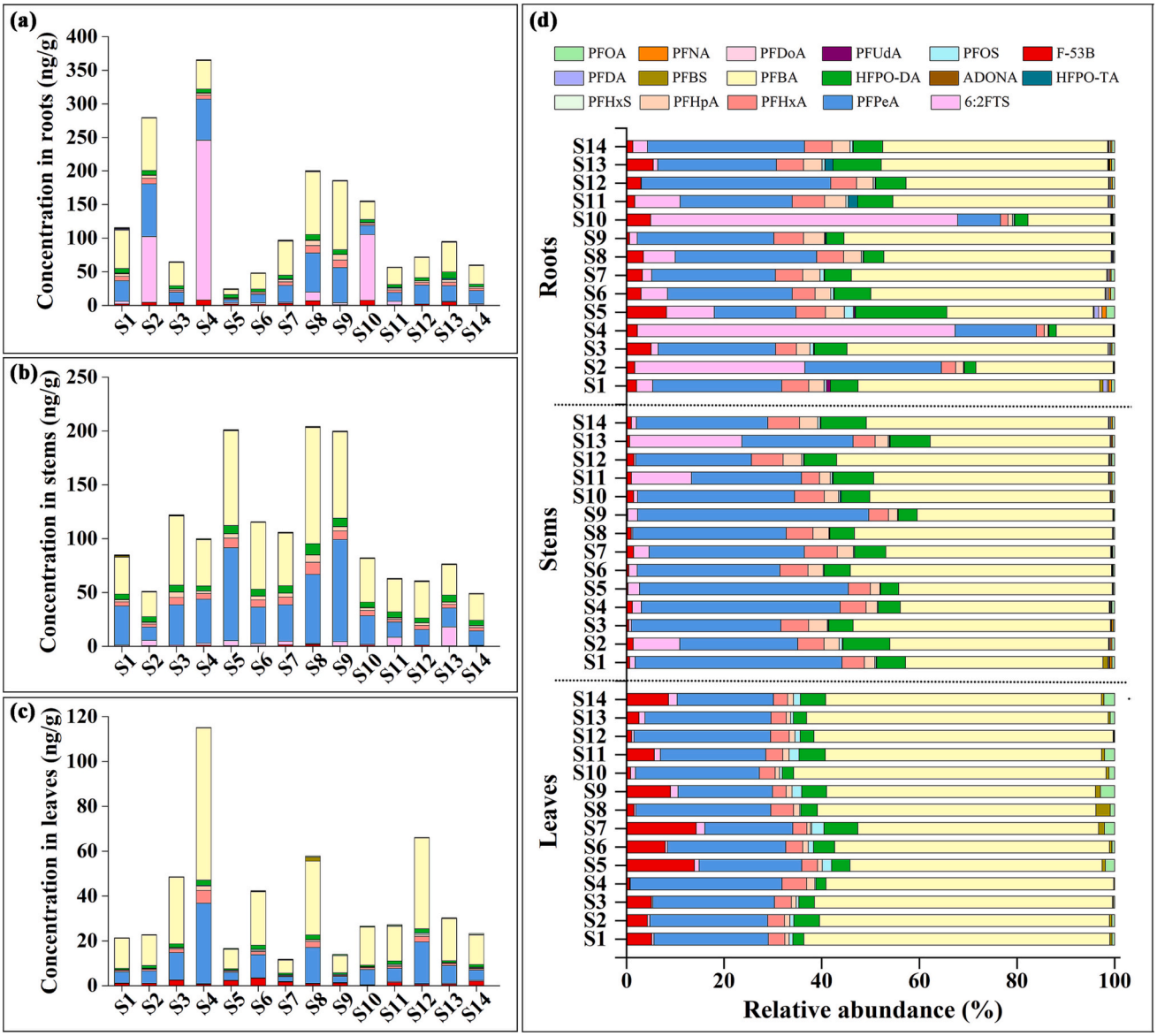

Fig. 2. PFASs concentration profile in cabbage (a) roots, (b) stems, (c) leaves, and (d) the relative abundance of individual PFAS in different cabbage fractions collected in the field.

3.1.2. PFASs uptake and bioaccumulation by cabbage

The PFASs uptake by different cabbage fractions and the potential bioaccumulation were assessed using their RCF, RSTF and SLTF (Fig. 3). In general, RCF, RSTF, and SLTF were apparently higher for short-chain PFCAs (C4–C7) than those for long-chain PFCAs (C8–C11), indicating that short-chain PFCAs were readily taken up from soil, and transferred more freely among different cabbage fractions (Fig. 3). Surprisingly, the RCF and RSTF of PFDoA (C12) were higher than those of long-chain PFCAs (C8–C11). Different bioconcentration patterns were observed for PFSAs. The RCFs for PFBS and PFHxS were lower than those of PFOS, F-53B and 6:2FTS, which could be attributed to higher sorption of PFOS, F-53B, and 6:2FTS by proteins in roots with a larger specific surface area (Miller et al., 2016). On the other hand, the RSTFs of PFBS and PFHxS can be readily transferred to stems, making them less accumulative in roots. Therefore, the higher accumulation of long-chain PFASs in roots with higher hydrophobicity and Kow values could be attributed to the higher number of lateral or fibrous in cabbage roots (Felizeter et al., 2014; Liu et al., 2023; Zhou et al., 2020). The RSTFs and SLTFs were different for PFOS, F-53B and 6:2 FTS, suggesting that chemical structure also plays a role in their translocation. And much higher SLTF (as compared to RCTF) was observed for PFBS, implying an additional source contributed to the PFBS in leaves. It was speculated that PFBS in the atmosphere can accumulate in the above-ground parts of plants via dry/wet depositions (Wen et al., 2014; Yao et al., 2022).

The RCF values decreased with the increasing C-F chain length. Blaine et al. (2014) reported that the RCFs values for four soil-cultured plants (radish, celery, tomato, and sugar snap pea) were slightly dependent on PFAA chain length. This could be attributed to the fact that PFAS with strong sorption by soils could demonstrate less bioavailability for plant root uptake (Mei et al., 2021). PFASs with a small molecular size and low hydrophobicity may be preferentially translocated (Mei et al., 2021), and several studies showed that TF values always decrease with increasing PFAS chain length (Gredelj et al., 2020b; T.T. Wang et al., 2020; W. Wang et al., 2020), which is consistent with our study. However, it was observed that the transfer capacity of PFBS was higher than that of PFBA and PFPeA in the stem-to-leaf and higher than that of in root-to-stem process (Fig. 3). Thus, despite having the same chain length of carbon atoms, they may have diverse uptake and transfer patterns. Several studies have shown that the translocation and partitioning behavior of a chemical in a plant is highly varied and

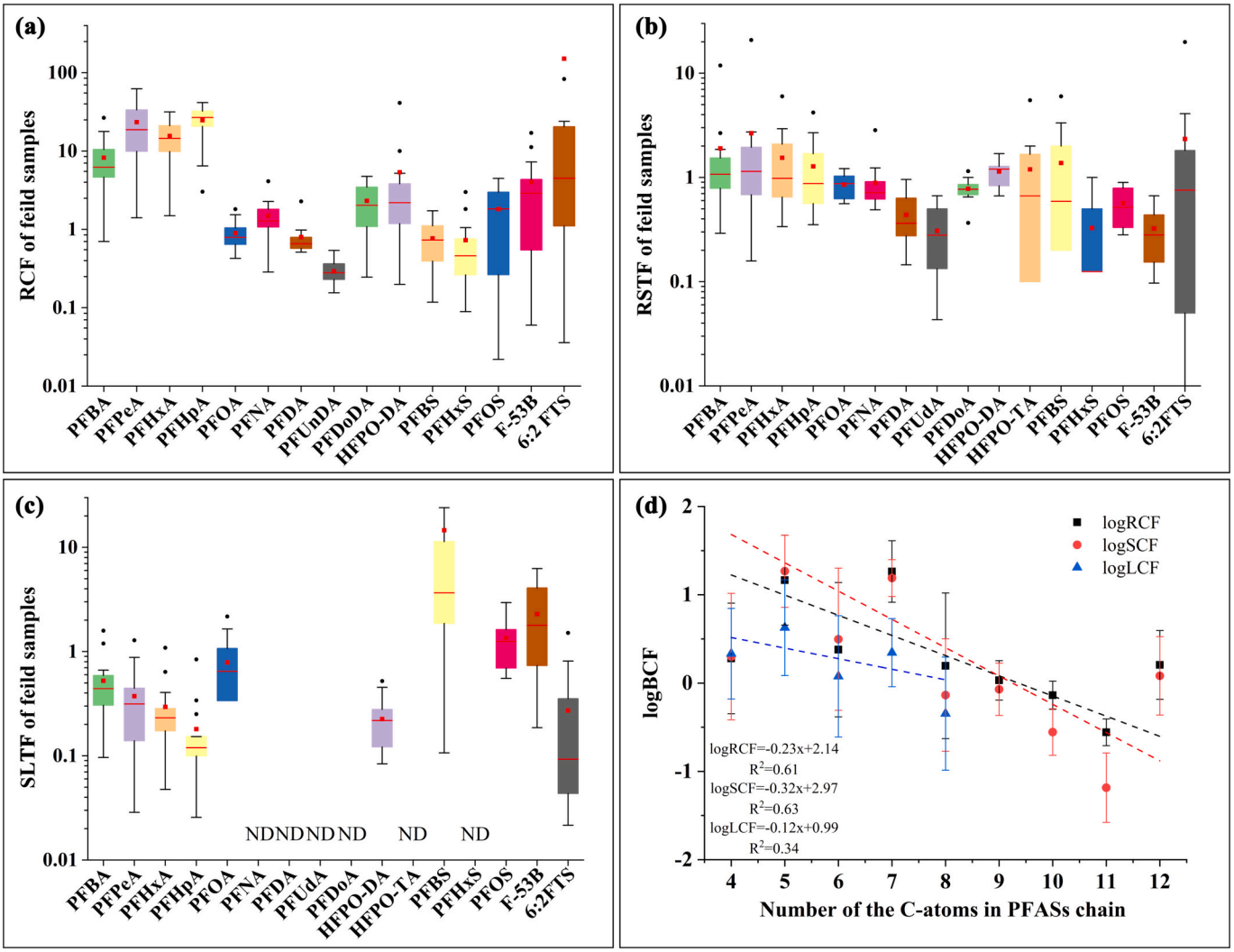

Fig. 3. PFASs uptake and bioaccumulation in different cabbage fractions (a) root concentration factors, (b) translocation factors of roots-to-stems, (c) translocation factors of stems-to-leaves of PFASs in cabbage in the field samples, and (d) correlations between logRCF, logRSTF, logSLTF, and the number of the carbon atoms of PFASs' perfluoroalkyl chain. Note: box and whisker plots show the concentration factor values of individual PFASs. Boxes represent the 25th to 75th percentiles; red lines in boxes represent medians; red filled squares shown inside or outside of boxes represent arithmetic means; distances between the whiskers represent nonoutlier ranges, and the closed black circles indicate outlier values; ND indicates that the compound was not detected in samples. The PFASs listed in the figure in the order of PFCAs, PFSAs, and their emerging alternatives.

complex (Blaine et al., 2014). PFOA and PFOS were observed may have distinct uptake mechanisms in maize, with entry by anion channels and entry by either aquaporins or anion channels (Wen et al., 2013). In addition, root proteins and lipids can interact with ionized organic compounds via hydrophobic interactions and electrostatic interactions, showing the importance of the relationship between root protein content and transport efficiency (Wen et al., 2016). Also, polar chemicals can enter the transpiration stream and migrate throughout the plant, while nonpolar chemicals are mostly confined to the surface of root membranes due to lipid partitioning (Blaine et al., 2014). Therefore, the transfer of PFASs was not only determined by the chain length, but also influenced by their functional groups, molecular structure, along with the physicochemical properties of the soil.

The RCF and TFs of PFASs can be influenced by their sorption to soil, which is often mainly affected by organic carbon, pH, and clay content, in addition to cation exchange capacity, anion exchange capacity, index cations, and ionic strength (Li et al., 2018; Milinovic et al., 2015). In addition, PFASs present in the ambient atmosphere of landfills and fluorochemical manufacturing parks have been found in leaves and barks, resulting in accumulation of PFASs in the above-ground parts of plants (Jin et al., 2018; Tian et al., 2018). Recent study indicated that foliar uptake can overweigh root uptake (Yao et al., 2022). These above factors have made the exploration on the uptake and translocation mechanisms within plants more challenging, even though field samples provide a more realistic understanding of plant uptake and distribution. Therefore, the following discussion shift the focus on the results from hydroponic experiment, to shed lights on the influence of chemical structure, such as the carbon chain-length, functional groups and the replacement of F atom by Cl.

3.2. *Bioaccumulation of PFASs under hydroponic condition*

3.2.1. *Root uptake of PFASs under hydroponic condition*

The concentrations of PFASs measured in cabbage roots, stems, and leaves harvested from pots with different initial PFASs concentrations are shown in Fig. 4a. The Σ_{10}PFASs concentrations were 400, 1390, 4030, and 6620 ng/g in roots, 40, 200, 800, and 1050 ng/g in stems, and 80, 390, 1570, and 2520 ng/g in leaves, for T1–T4, respectively (Fig. 4a). The Σ_{10}PFASs concentration in roots was the highest, as expected. The PFAS concentration followed the order of PFDA > F-53B >

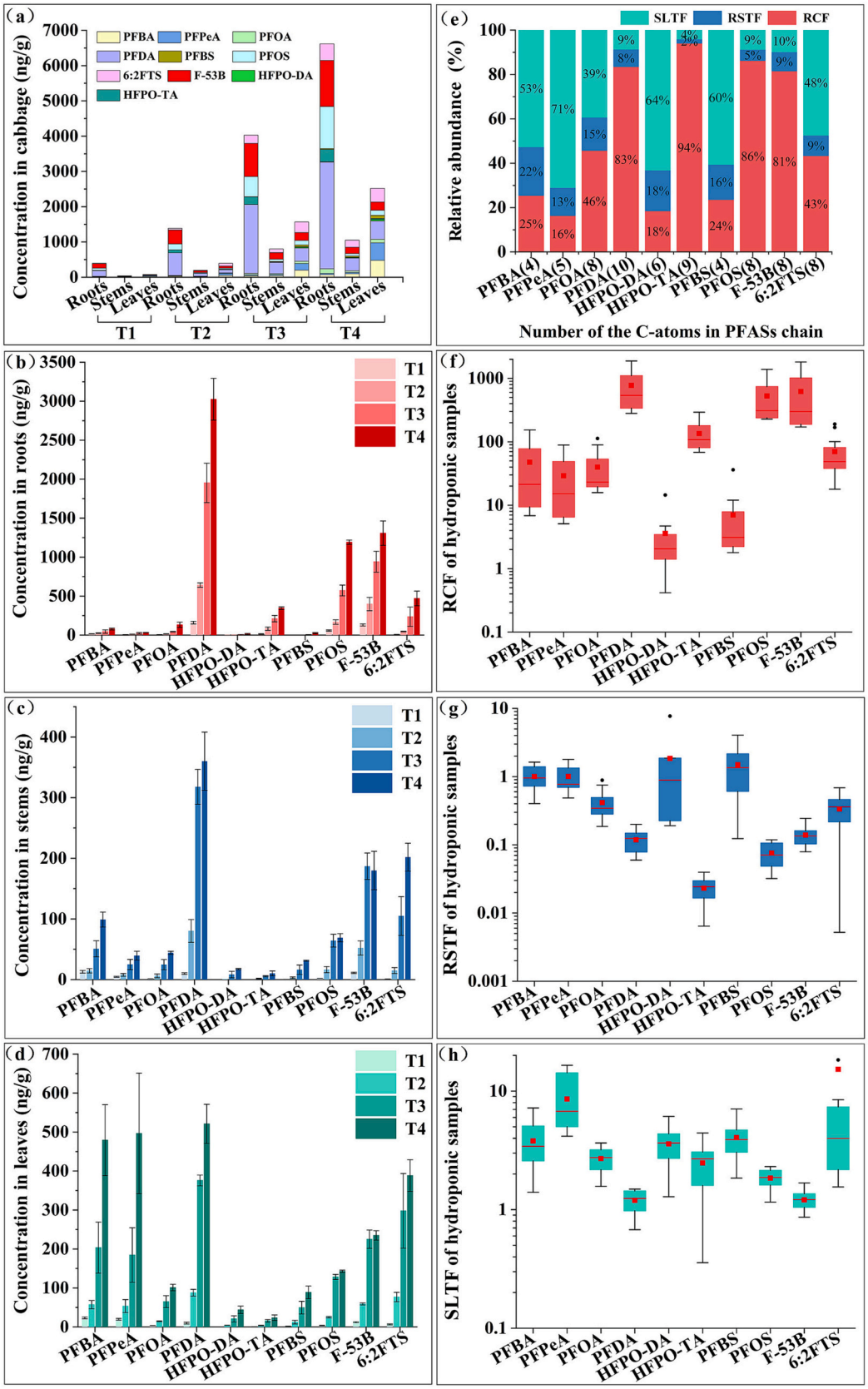

Fig. 4. The relative abundance of individual PFASs (a) and concentrations of PFASs in cabbage roots (b), stems (c), and leaves (d) in the hydroponic conditions. Different numbers of the carbon atoms of PFASs' perfluoroalkyl chain of the BCFs' relative abundance (e), bioconcentration factors of roots (f), translocation factors of roots-to-stems (g), and translocation factors of stems-to-leaves (h) in all of the treatments. Error bars represent the standard error estimates (n = 3). The numbers in parentheses in (e) represents the number of carbon atoms in the individual PFASs. The PFASs listed in the figure in the order of PFCAs, PFSAs, and their emerging alternatives.

PFOS > 6:2FTS > HFPO-TA in roots (Fig. 4b). Based on a meta-analysis of published data, Mei et al. (2021) concluded that the RCF of PFASs was significantly positively correlated with hydrophobicity (log K_{OW}) under hydroponic conditions. Therefore, it was reasonable to expect that the long-chain PFASs, including PFOA, PFDA, HFPO-TA, PFOS and F-53B, were the main components due to their higher root uptake potentials.

3.2.2. Translocation of PFASs to stems and leaves under hydroponic condition

The Σ_{10}PFASs concentrations were much higher in leaves than stems, indicating that PFASs can be transferred to the leaves with the transpiration stream via xylem (W. Wang et al., 2020). Several studies further confirmed that transpiration was one of the main drivers for PFASs uptake by plants, and PFASs in soil could be transported from the roots to the aboveground organs through transpiration (Felizeter et al., 2014). The PFAS concentrations in stems followed the order of PFDA > F-53B > 6:2FTS > PFBA > PFOS (Fig. 4c); and PFDA >6:2FTS > PFBA > PFPeA > F-53B > PFOS in leaves (Fig. 4d). Interestingly, the PFDA concentrations were the highest in roots, stems, and leaves among all four treatments, with the majority of PFDA stored in roots. In addition, it is worth noting that the abundance of PFDA in leaves was slightly higher than that in stems, indicating that PFDA can be translocated from stems to leaves, in particular considering its long-chain (10-carbon) (Fig. 4e).

The concentrations of short-chain PFBA, PFPeA, and PFBS followed the trend of leaves > stems > roots, indicating that they were readily being transferred from roots to stems and then to leaves, and most of them were stored in leaves (Fig. 4). It was reported that the small-sized chemicals and their low affinity to plant roots contributed the most to being readily transported to the upper portions of the vegetables (Chuang et al., 2019). More interestingly, the profiles of PFBA and PFPeA suggested that PFPeA was more easily transferred to leaves, even though there is an extra carbon in the molecular structure of PFPeA. In addition, the translocation factors of PFPeA were the highest among all PFASs in leaves, and significantly higher than its RSTF and RCF values, suggesting their high mobility among different organs. Furthermore, the RSTFs were the lowest for each PFAS compared to the RCF and SLTF values observed (Fig. 4e). The concentrations in stems were expected to be largely dependent on the amount of PFAA delivered by the transpiration stream from the roots, and the balance between PFAA retention in stem tissues and further transfer to twigs and leaves with transpiration stream (Felizeter et al., 2014). This suggests that cabbage stems were primarily responsible for the transfer of these chemicals to the leaves, rather than their accumulation.

Long-chain PFOA, PFDA and PFOS had higher RCF values and lower RSTF and SLTF values compared to other long-chain PFASs, indicating their lower mobility within the cabbage plant. The BCFs and TFs are often affected by the carbon chain length and functional groups of PFASs (Gredelj et al., 2020c). The results of the hydroponic experiment suggested that the carbon chain length played a more important role in determining the BCFs than the functional groups, consistent with the conclusion of Gredelj et al. (2020a). This was also consistent with previous hydroponic studies in cabbage, where only a small proportion of the long-chain PFCAs were transferred from roots to stems (Felizeter et al., 2014).

Both HFPO-DA and HFPO-TA, the alternatives for PFOA, were less bioaccumulative than PFOA, with the exception of HFPO-TA in roots, which was higher than that of PFOA (Fig. 4). However, HFPO-DA were more readily translocated to the stems and leaves, primarily due to its shorter carbon chain, whereas HFPO-TA, like PFOA, was more readily stored in the roots, likely due to its longer carbon chain. Consistent with previous studies (Lin et al., 2020; Pan et al., 2021), F-53B, an alternative of PFOS, was more accumulative than PFOS. In contrast, 6:2FTS was less bioaccumulative in roots and stems than PFOS, but more bioaccumulative in leaves. The profiles of 6:2FTS indicated that it was more readily transferred among different organs than PFOS, as further shown by its higher relative abundance in leaves and higher RSTF and SLTF values, suggesting that the replacement of fluorine atoms with hydrogen atoms in PFASs can make them more mobile. In addition, a study showed that exposure to PFOS and its alternatives have the neurotoxicity potential and that PFOS alternatives even exhibited comparable potency to PFOS, suggesting that PFOS alternatives may have the same toxicological profile as PFOS (Zhang et al., 2016). Therefore, the much higher concentrations of 6:2 FTS in cabbage leaves (the edible part) suggested that the potential health risk via the intakes of 6:2 FTS can be higher than that of PFOS.

3.3. Comparison of PFAS bioaccumulation in cabbage collected from field and hydroponics investigations

The uptake of PFASs via the foliar exposure pathway has been reported under field conditions (Xu et al., 2021b; Yao et al., 2022). It has also been shown that dissolved chemicals can be transported through the phloem or xylem in hydroponically-cultured tomato and zucchini plants, and the translocation routes can be compound/plant species specific (Felizeter et al., 2014; Mei et al., 2021; Miller et al., 2016), especially for the short-chain PFASs, the uptake of which by roots from soil moisture were easier and transferred to the above-ground parts. In contrast with field conditions, the PFAS can only be absorbed from the solution via the roots, then translocated to stems and leaves under hydroponic conditions. The distribution patterns of PFASs in field and hydroponic samples were quite different in general (Fig. 5). The abundance of the majority of PFASs in leaves under field conditions were generally lower than those in hydroponic experiments, primarily due to the availability of PFASs, which is influenced by the hydrophobicity of the chemical and the physicochemical properties of the soil (Li et al., 2022; Mei et al., 2021). Due to the PFOS sorption to soils in the rhizosphere, PFASs were less available in soil pore water for plants uptake, thus contributing to the lower transfer efficiency to the stems and leaves in field samples. Correspondingly, the abundance of the majority of PFASs in roots tended to be higher or similar to those observed under hydroponic condition.

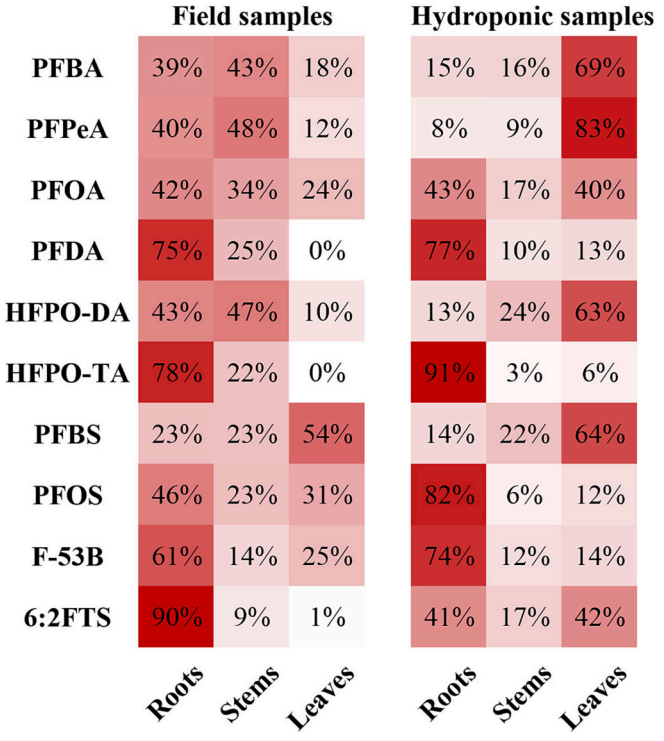

Fig. 5. Comparison of the percentage of PFASs in different organs in field and hydroponic samples.

However, there are exceptions to the above general observations, such as PFOS and F-53B, which were detected with higher abundance in leaves (and lower abundance in roots) for samples collected in the field, indicating there is a contribution from the leaf uptake from the air in the field. This is consistent with the previous findings (Xu et al., 2021b). Furthermore, no detections of PFDA and HFPO-TA were observed in leaves collected in the field, suggesting that no contributions of these two chemicals from the air, and they were more likely to accumulate in roots, less likely to be translocated to the leaves due to their longer chain length. In addition, 6:2 FTS was detected in the leaves with an abundance of 1 %, significantly less than that in the hydroponic experiment, suggesting that it probably follow the same trend of PFDA and HFPO-DA. Finally, it is worth noting that while PFBA and PFBS shared a similar distribution trend within the different organs of cabbage under hydroponic conditions, their distributions in the field were quite different. The abundance of PFBS in the leaves was much higher than that of PFBA under field conditions, suggesting a contribution of PFBS from air uptake, as discussed in the source identification (Xu et al., 2021b).

4. Conclusions

Studies on the accumulation of PFASs in plants, especially edible plant parts, are important to the security of food consumption. The results of this study revealed that different PFAS accumulation patterns were observed between the field and hydroponic conditions, suggesting that the field conditions can be more complex due to the heterogeneities and contributions of other sources (e.g., dry/wet depositions from the air), which is very important for the evaluation of PFASs toxicity. Furthermore, the emerging alternatives, such as HFPO-DA, F-53B, and 6:2 FTS, were more accumulative in cabbage roots than PFOA and PFOS under both field and hydroponic conditions. Yet the profiles of 6:2FTS indicated that it was easier being transferred among different organs than PFOS, and the much higher SLCF of 6:2 FTS in cabbage leaves (the edible part) poses a higher potential health risk via intake of 6:2 FTS than that of PFOS. For the short-chain analogues, such as PFBA, PFPeA, PFBS, are more mobile than the legacy PFOA and PFOS, resulting in the much higher concentrations in the edible part, posing a higher potential health risk. Therefore, it is recommended that the choice of alternatives for legacy PFOA and PFOS be carefully considered. Moreover, the analyses of plant physical data in future studies be carried out, which can shed lights on the mechanisms of PFAS translocation. It was unexpected that PFDA was the dominant compound in roots, stems, and leaves under hydroponic condition, with the majority of PFDA stored in roots. The relatively high bioaccumulation of PFDA under hydroponic condition warrants further investigation. Finally, it is recommended the contributions of dry/wet depositions be further studied with PFASs in air samples being analyzed to verify the observations in this study. In summary, the findings of this study provide valuable insight into the transfer of PFASs among cabbage organs, and lay a foundation for the evaluation of PFAS toxicity via food intake.

CRediT authorship contribution statement

Jilu Che: Conceptualization, Investigation, Visualization, Data curation, Writing -original draft, Writing - Review. **Chang Xu**: Conceptualization, Investigation, Visualization, Data curation, Writing - Review. **Xin Song**: Supervision, Conceptualization, Writing - Review & Editing, Project administration, Funding acquisition. **Xiaoyan Ding**: Data curation, Methodology, Writing - Review. **Mukhtiar Ali**: Data curation, Writing - Review. **Hong Chen**: Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgement

This work was supported by the National Natural Science Foundation of China (32061133001 and 41771354).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167578.

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