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Review

Translocation, bioaccumulation, and distribution of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in plants

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SUMMARY

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are persistent in the environment and have been detected in a variety of plants such as vegetables, cereals, and fruits. Increasing evidence shows that plants are at a risk of being adversely affected by PFASs. This review concludes that PFASs are predominantly absorbed by roots from sources in the soil; besides, the review also discusses several factors such as soil properties and the species of PFASs and plants. In addition, following uptake by root, long-chain PFASs (C $\geq$ 7 for PFCA and C $\geq$ 6 for PFSA) were preferentially retained within the root, whereas the short-chain PFASs were distributed across tissues above the ground — according to the studies. The bioaccumulation potential of PFASs within various plant structures are further expressed by calculating bioaccumulation factor (BAF) across various plant species. The results show that PFASs have a wide range of BAF values within root tissue, followed by straw, and then grain. Furthermore, owing to its high water solubility than other PFASs, PFOA is the predominant compound accumulated in both the soil itself and within the plant tissues. Among different plant groups, the potential BAF values rank from highest to lowest as follows: leaf vegetables > root vegetables > flower vegetables > shoot vegetables. Several PFAS groups such as PFOA, PFBA, and PFOS, may have an increased public health risk based on the daily intake rate (ID). Finally, future research is suggested on the possible PFASs degradation occurring in plant tissues and the explanations at genetic-level for the metabolite changes that occur under PFASs stress.

INTRODUCTION

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) have been extensively used in consumer products and during industrial processes (Xu et al., 2020a; Giesy and Kannan, 2002). Owing to the high stability of carbon–fluorine (C—F) bonds in PFASs (Xu et al., 2020b), these compounds are very stable and frequently detected throughout numerous environmental media, i.e., atmosphere, soil, water, groundwater, and sediments (Xu et al., 2021; Ahmed et al., 2020a; Yu et al., 2020). Unfortunately, with continual usage and long half-lives, PFASs have been detected in aquatic and terrestrial plants and animals, as well as in humans, making them an important focus of study, particularly as a common environmental toxicant worldwide (Xu et al., 2017). For example, it was recently reported that 16 PFASs were measured in the livers of neonatal Australian pinnipeds (*Arctocephalus pusillus doriferus*) at concentrations ranging from 0.5–2119 ng g^{-1} wet weight (wet w.t.) (Taylor et al. 2021). Perfluorooctanoate acid (PFOA), perfluorooctane sulfonate (PFOS), the predominantly detected perfluoro octanoic acid (PFCA), and perfluorosulfonates acid (PFSA) were determined to be ranging between 1.3–2.0 and 2.7–5.9 ng L^{-1}, respectively, in the cord sera of 942 newborns from a cohort in Wuhan, China, between 2013 and 2014 (Liu et al., 2021). Other PFASs have also been detected in the environment, such as perfluorobutanoic acid (PFBA), Perfluorobutane sulfonic acid (PFBS), and perfluorododecanoicacid (PFDoA). PFASs, especially PFOA or PFOS, have been proved to induce reactive oxygen species (ROS) production and oxidative stress, causing a series of toxic responses such as neurotoxicity, immunotoxicity, developmental and reproductive toxicity, and endocrine disruption in organisms (Ahmed et al., 2020b; Li et al., 2016; Zeng et al., 2019). In 2017, the International Agency for Research on Cancer (IARC) classified PFOA as a possible human carcinogen based in part on limited epidemiologic evidence

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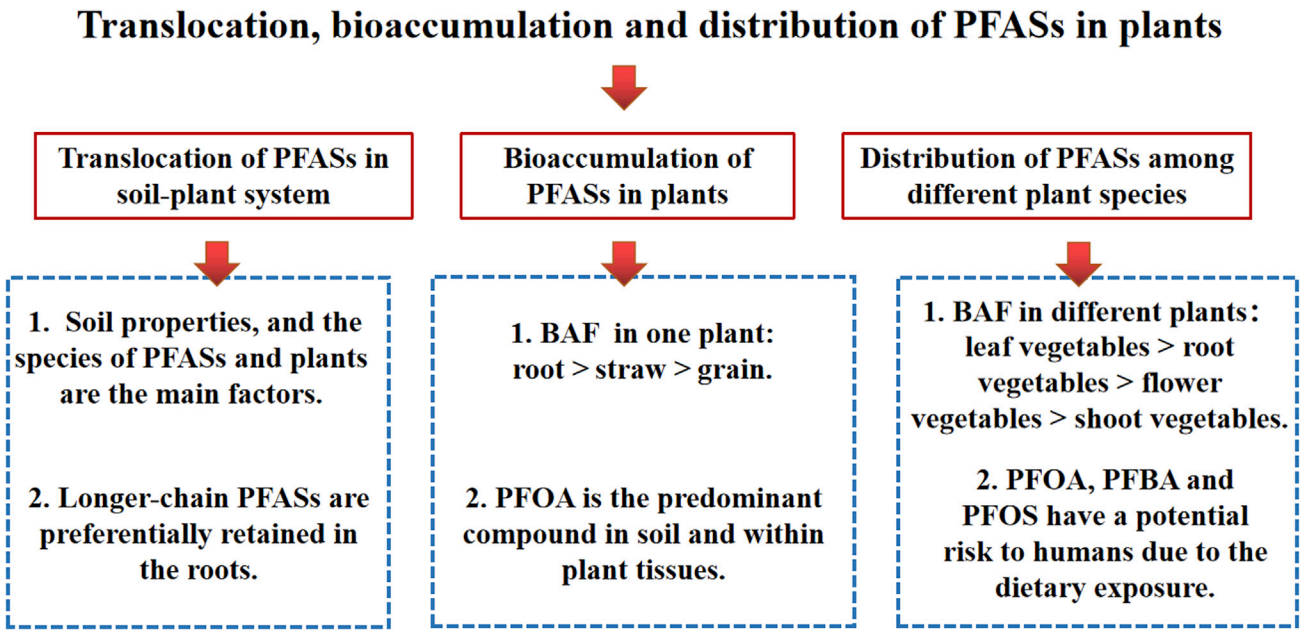

Figure 1. The bullet points of the review in each section

related to cancers of the kidney and testis in heavily exposed subjects. Flynn et al. (2019) investigated the acute and chronic effects of PFAS mixtures on wildlife of larval American bullfrogs (Rana catesbeiana), claiming that the LC_{50} of PFOS and PFOA were 144 mg/L and 1004 mg/L, respectively, based on the 96 h acute toxicity tests. As such, characterizing the risk of PFASs posed to public health is of increasing importance.

Recent studies indicated that the presence of PFASs, specifically PFCA, were detected in a variety of plants, including vegetables, fruits, and cereals, in Belgium, the Czech Republic, Italy, and Norway (D'Hollander et al., 2015; Herzke et al., 2013). A large number of studies have determined the bioaccumulative potential PFASs in plants via soil or irrigation water, with levels of PFASs in plants thought to be significantly underestimated because of their ubiquitous occurrence in environmental media (Ghisi et al., 2019). Therefore, plants are considered as the possible contributors to the uptake of PFASs in humans, either directly as a part of the human diet or indirectly through livestock (Klenow et al., 2013; Kowalczyk et al., 2013).

Soil itself represents an important reservoir for a number of pollutants including PFASs (Ahmed et al., 2020b). Because of the adsorptive capacity of soil for several PFASs, it is possible for PFASs to stay bound to soil and then be transported and accumulate into plant tissues, thereby posing a potential health risk to humans following uptake. So far, multiple studies have been conducted to better understand the toxic potential of PFASs contamination to organisms in the environment, though less focus has been placed on understanding the role of PFASs interactions within soil-plant systems. Dalahmeh et al. (2018) determined that PFASs concentrations were within a range of 160 pg g^{-1} d. w. in maize cobs and 380 pg g^{-1} d. w. in sugarcane stems, and the PFASs in soil nearby were reported to be 1700–7900 pg g^{-1} dry weight (d. w.). Li et al. (2020a) found the disrupted metabolic profiles regulating mineral elements and organic compounds in lettuce and the impaired defense properties, following PFOA and PFOS exposure.

The main aim of this review is to summarize the work conducted on PFASs within soil-plant systems by characterizing the main route of PFASs from soil to plants and the translocation/movement to plant tissues. In addition, we discuss the bioaccumulation of PFASs in whole plants and possibility of distribution, the difference of PFASs accumulation in different plants, and the potential risks to human health. The bullet points of the review have been exhibited in Figure 1 for the better understanding.

METHODOLOGY

Based on an online database search (Mainly in Web of Science) of peer-reviewed articles, nearly 27 journal articles that reported PFAS concentrations in plants and the surrounding soil were identified and reviewed

for the calculation in this article. The concentration ranges, median, and other statistical values were listed in figures and tables. During data analysis, the Methods detection limit (<MDL) was assumed to be zero.

TRANSLOCATION OF PFASS IN SOIL-PLANT SYSTEM

Root uptake of PFASs from soil

The root uptake from soil has been considered as a main source of PFASs to plants, which is primarily a diffusive process (Wang et al., 2020). Commonly, PFASs desorbed in interstitial water could be absorbed by root epidermis and then penetrate through the epidermis into vascular root tissue via two routes i.e., the symplastic route (between cells along cell walls) and the apoplastic route (through cells via plasmodesmata) (Zhao et al., 2014). The process regarding PFASs uptake and accumulation has been discussed in detail previously (Mei et al., 2021). Wen et al. (2013) presented that the nonlinear absorption of PFOS and PFOA fit the Michaelis-Menten model well, indicating that the uptake of PFASs by roots was likely a carrier-mediated process (Yang et al., 2010). In addition, Wang et al. (2020) investigated the uptake mechanism of PFOA and PFOS by wetland plant *Alisma orientale* (*A. orientale*), determining that PFASs are likely to be transported through water and anion channels, as the pore diameter matches the C-F bond size. Accordingly, the transportation rate of this process is considered to be decided by factors including soil property and the species of PFASs and plants.

The soil property plays a critical role in the transportation of PFASs. The soil organic matter (SOM) is an important sorbent for PFASs, with the root uptake of PFASs beinginversely proportional to their sorption to SOM (Mei et al., 2021), whereas the sorption capacity of PFASs to SOM was mostly correlated with the soil organic carbon (SOC). With increased amounts of SOC, more PFASs can bind to soil particles than being dissolved in the soil solution, which would lead toreduced uptake of PFASs by plants (Blaine et al., 2014b; Wen et al., 2014). Notably, montmorillonite clay included in soil effectively binds to a variety of environmental chemicals, and the nutrient-amended clays could decrease the bioavailability from soil and translocation to plants (Hearon et al., 2022). In addition, other soil properties such as soil temperature, soil salinity, pH, soil moisture content, and cation exchange capacity may also affect PFASs absorption by plants (Mei et al., 2021). Zhao et al. (2013) found that wheat plants growing in a nutrient solution with a pH of 6-8 had the highest accumulation of PFOS, with a low accumulation at a pH between 4 and 10. Conversely, Krippner et al. (2014) did not observe a pH-dependent absorption of PFOS in maize plants growing in nutrient solution, but found that PFDA had a higher accumulation at a pH 5 than at pH 7. Zhao et al. (2016) showed that the root concentration of each PFCA increased with increase in temperature, and that with a temperature increase from 20°C to 30°C, the root absorption of long-chain PFCA was faster than that of short-chain PFCA in wheat.

In addition, the species of PFASs seem to play an essential role during the root uptake process, which could be valued by the root concentration factor (RCF, the ratio of the FPAS concentration in root to that in hydroponic solution or bulk soil). However in the latest study, Mei et al. (2021) concluded that the RCF values have a sure relationship with neither the initial PFAS concentration nor the PFAS chain length in soil. Several studies have proved that the RCF values are significantly related to the PFAS chain length because of the greater hydrophobicity of longer chain compounds; however, some studies have the contradictory conclusion that RCF is independent of the PFAS chain length. This uncertainty might be attributed to the competition among PFASs of different chain lengths for sorption sites between soil particles and the root.

With regard to plants themselves, different species have different root uptake effects of PFASs. Root protein and lipid content have been considered as the main influencing factors as they can interact with ionized PFASs in interstitial water via hydrophobic interactions and electrostatic interactions (Mei et al., 2021). In the laboratory studies, seven soil-cultured plants such as alfalfa (*Medicago sativa L. cv. Chaoren*), lettuce (*Lactuca sativa L.*), maize (*Zea mays L. cv. Nongda 108*), mung bean (*Vigna radiata L. Wilczek*), radish (*Raphnus sativus L. cv. Dahongpao*), ryegrass (*Lolium multiflorum Lam.*), and soybean (*Glycine max L. Merrill*) were studied to determine the role of protein and lipid content in the accumulation potential of PFOA and PFOS by Wen et al. (2016). They found that the RCF values were significantly positively correlated with root protein content and negatively correlated with root lipid content, possibly because lipids might compete with PFOA and PFOS for root protein adsorption sites, whereas the specific proteins could mediate the transport of organic compounds during the root uptake process. In addition, root exudates of plants are found to be associated with the PFASs uptake by root. Especially, oxalic acid in root exudates plays a key role in activating PFOA uptake in lettuce with more than 80% attribution proved by Xiang et al.

(2020). The more oxalic acid in root exudates leads to the more PFOA bioaccumulated in the plant. Besides, the plant height, evaporative potential, and other morphological and physiological variations among different plant species likely affect the accumulative potential of PFASs from soil, which should be considered in future studies.

Relating natural field-based scenarios to laboratory-based simulations can introduce complex interactions that can be difficult to replicate and may influence the true accumulative potential of PFASs. Li et al. (2018) recently reviewed the role of soil and sediment properties in determining PFASs sorption and argued that it cannot be explained by a single soil or sediment property to discuss the behavior of these substances in the environment. Moreover, the analyses of influencing factors of PFASs adsorption are not always clear or consistent, making it difficult for cross-lab comparisons and relating field-based data to laboratory findings.

Transfer from root to the other organs

Following sorption by roots, PFASs can transport from the root to aboveground tissues such as stems, leaves, shoots, flowers, and fruits via xylem or phloem, which can be expressed by translocation factor (TF). As previously described, TF values always decrease with the increasing PFASs chain length, and branched PFOA and PFOS isomers generally have the higher TF values than those of linear isomers because of the higher hydrophilicity of branched isomers (Felizeter et al., 2012; Gredelj et al., 2020). Therefore, theoretically, a small molecular size of PFASs with a short-chain and branched isomers would be preferentially translocated and accumulated in each tissue of the plant. For example, Zhang et al. (2020) investigated the translocation of PFASs from roots to shoots in *T. angustifolia* and found the TF values of PFASs to be greater than one for short-chain, except for longer-chained compounds such as PFOS, PFOA, and PFHxS, although the longer-chain PFASs are preferentially retained in the roots. For example, Zhang et al. (2020) found that the highest individual PFASs concentration in the roots was observed for PFOS (68.9 ng g^{-1}), PFOA (18.5 ng g^{-1}), and PFHxS (13.4 ng g^{-1}), which was higher than the short-chain PFBA (1.5 ng g^{-1}), PFPeA (4.4 ng g^{-1}), and PFHxA (6.9 ng g^{-1}) compounds.

The metabolic processes also occur during transformation within the plant. PFASs would go through three phases of metabolism after root uptake including transformation, conjugation, and sequestration (Jiao et al., 2020). During the phase of conjugation, the metabolites of PFASs after oxidation, reduction, or hydrolysis are conjugated with phytochemicals. For example, Zhang et al. (2016) identified two glutathione conjugates during the metabolic process of 8:2 FTOH in soybean tissues.

BIOACCUMULATION OF PFASS IN PLANTS

PFASs in different plant organs

PFASs can be transported from the root to other organs and accumulated (Miller et al., 2016) as shown in Table 1. The first systematic study describing the bioaccumulative potential and interorganizational distribution of PFASs in plants was conducted by Stahl et al. (2009). They found that PFOA and PFOS can be transferred from the soil into plant by testing five commonly cultivated plants (spring wheat, oats, potatoes, maize, and perennial ryegrass) and specifically described the concentrations of PFOA/PFOS accumulated in different plant organs (Stahl et al., 2009). Subsequently, more studies have found differences in the enrichment of PFASs in plant tissues as shown in Table 1. Wen et al. (2013) indicated that PFOA and PFCA were mainly retained in the root of crops, whereas Krippner et al. (2015) and Stahl et al. (2009) found that straw had a greater rate of accumulation than kernels. However, because of various environmental factors, different PFASs confirmations, and plant types/properties, the bioaccumulation potential and mechanistic understanding of PFASs movement within plant tissues remains diverse and relatively inconsistent (Ghisi et al., 2019).

Maize serves as an important model organism for basic and applied research on chemical migration because of its ability to produce flowers, fruits, stems, roots, and leaves at the same time and is easy to grow (Strable and Scanlon, 2009); in addition, itis also one of the most studied plants regarding the bioaccumulation of PFASs (Ghisi et al., 2019). To characterize the bioaccumulative potential of PFASs in maize, we assessed the BAF of PFASs with the dried weight basis from different organs and used this value to calculate the accumulation ability in these plant organs as reported by previous studies (Stahl et al., 2009; Wen et al., 2013; Krippner et al., 2015; Navarro et al., 2017; Lan et al., 2020; Song et al., 2021). The BAF values were calculated by using the following formula:

Table 1. Initial soil concentrations (mg kg^{-1} soil), concentrations in plants (μg kg^{-1} d.w.) and BAF of typical PFSA (PFBA, PFBS, PFOA, and PFOS) in different plants and their different tissue

Plant species		Plant parts	Compounds	Initial soil concentrations (mg/kg soil)	Concentrations in plants (μg/kg d.w.)	Bioaccumulation factor (BAF)	References
Cereals	Oat	Straw	PFOA	0.25, 1	220, 690	0.88,0.69	Stahl et al. (2009)
			PFOS	0.25, 1	56, 150	0.224, 0.15	
		Grains	PFOA	1	54	0.054	
			PFOS	1	17	0.017	
	Wheat	Straw	PFBA	0.014	22.2	1.64	Stahl et al. (2009)
			PFBS	0.031	21.8	0.64	Wen et al. (2014)
			PFOA	0.026, 0.25,1	22.1, 800, 1900	0.85, 3.2, 1.90	
			PFOS	0.041, 1	11, 470	0.27, 0.47	
		Grains	PFBA	0.013	6.4	0.48	Stahl et al. (2009)
			PFBS	0.0312, 0.01	<MDL, <MDL	–	Wen et al. (2014)
			PFOA	0.026, 0.25, 1, 83.16	2.9, 24, 9, 6.68	0.11, 0.096, 0.009, 0.08	Liu et al. (2019)
			PFOS	0.041, 1, 0.14	2.53, <MDL, 0.18	0.062, -, 1.29	
		Husks	PFBA	0.014, 4.76	5.77, 1768.13	0.43, 371.46	Wen et al. (2014)
			PFBS	0.01, 0.031	0.4, <MDL	40, -	Liu et al. (2019)
			PFOA	0.026, 83.16	4.19, 244.47	0.16, 2.94	
			PFOS	0.041, 0.14	2.2, 2	0.054, 14.26	
	Soybean	Leaf	PFBA	–	2378.31	–	Liu et al. (2019)
			PFBS	–	<MDL	–	
			PFOA	–	3966.62	–	
			PFOS	–	2.35	–	
		Grain	PFBA	–	1078.02	–	
			PFBS	–	<MDL	–	
			PFOA	–	11.06	–	
			PFOS	–	0.12	–	
Vegeta-bles	Carrot	Carrots (peeled)	PFOA	0.68, 0.68, 0.53, 0.49	333, 328, 148, 144	0.49, 0.49, 0.28, 0.3	Lechner and Knapp. (2011);
			PFOS	0.01, 0.46, 0.45, 0.34	5.3, 196, 240, 162	0.53, 0.43, 0.55, 0.49	Bizkarguenaga et al. (2016)
		Root	PFBA	7.19	2552.74	355.04	
			PFBS	0.03	1.1	36.67	Liu et al. (2019)
			PFOA	91.26	1468.08	16.09	
			PFOS	<MDL	1.31	–	
		Leaf blade	PFBA	7.19	279.75	38.91	
			PFBS	0.03	0.09	3	
			PFOA	91.26	51.64	0.57	
			PFOS	<MDL	0.79	–	
	Welsh onion	Pseudostem	PFBA	8.47	40.5	4.78	Liu et al. (2019)
			PFBS	<MDL	<MDL	–	
			PFOA	119.37	16.97	0.14	
			PFOS	0.06	0.09	1.5	
		Leaf blade	PFBA	8.47	270.39	31.92	
			PFBS	<MDL	0.07	–	

(*Continued on next page*)

Table 1. *Continued*

Plant species	Plant parts	Compounds	Initial soil concentrations (mg/kg soil)	Concentrations in plants (μg/kg d.w.)	Bioaccumulation factor (BAF)	References
		PFOA	119.37	360.58	3.02	
		PFOS	0.06	0.1	1.67	
Celery	Root	PFBA	3.88	517.84	133.46	Liu et al. (2019)
		PFBS	<MDL	0.07	–	
		PFOA	81.81	218.15	2.67	
		PFOS	0.06	0.11	1.83	
	Leaf petiole	PFBA	3.88	433.2	111.65	
		PFBS	<MDL	0.05	–	
		PFOA	81.81	75.44	0.92	
		PFOS	0.06	0.07	1.17	
	Leaf blade	PFBA	3.88	1049.61	270.52	
		PFBS	<MDL	<MDL	–	
		PFOA	81.81	1119.41	13.68	
		PFOS	0.06	1.62	27	
Radish	Root	PFBA	5.55, 0.0047, 0.0009	84.13, 13.67, <MDL	15.16, 2.92, -	Liu et al. (2019)
		PFBS	<MDL, 0.049, 0.00021	0.06, 61.89, 23.88	-, 1.27, 114	
		PFOA	68.9, 0.07852, 0.01491	95.34, 66.89, 8.11	1.38, 0.85, 0.54	
		PFOS	0.07, 0.050, 0.32	0.06, 34.86, 21.03	0.86, 0.7,0.066	
	Shoot	PFBA	5.55	1167.52	210.36	
		PFBS	<MDL	<MDL	–	
		PFOA	68.9	1879.76	27.28	
		PFOS	0.07	1.85	26.43	

"MDL" means the method detection limit; "-" means not detected.

$$BAF = \frac{\text{PFASs concentration in plant (ng g}^{-1}\text{ d. w.)}}{\text{PFASs concentration in soil (ng g}^{-1}\text{ d. w.)}} \quad \text{(Equation 1)}$$

BAF in roots varied from 23.94 to 75.52, with the root exhibiting the highest BAF in maize tissue for both PFOS and PFOA after 1 mg L^{-1} treatment (Wen et al., 2013). Similarly, a preferential accumulation in the root was found in other PFASs pollutants such as PFBS in maize (Navarro et al., 2017), as well as in other plants such as wheat (Lin et al., 2020) and radish (Blaine et al., 2014b), which suggests a strong enrichment of PFASs in root tissue relative to other plant tissues. Aside from root tissue, the BAFs of leaves (from 0.8 to 38.3) and shoots (from 5.76 to 29) were much higher than the husk (from 0.54 to 1), straw (from 0.126 to 5.16), kernel (from 0.002 to 3.29), and ear (from 0.003 to 0.581) (Figure 2).

The accumulative preference of PFASs in root tissue is mainly because of the absorption of nonionized organic compounds from soil or nutrient solution through roots (Wen et al., 2013). Navarro et al. (2017) studied the mass distribution of 11 PFASs in spinach, tomato, and corn tissues by their chain length classification and found that long-chain PFASs (75%) (C7–C10) (54–96%) preferentially remained in roots and the short-chain PFASs (C4–C6) tended to be translocated to aboveground tissues (leaf: 31–56% and fruit: 32–48%). This predominant accumulation phenomenon was also consistent with other studies conducted in the typical plant models tomato (*Solanum lycopersicum* var. Moneymaker), zucchini (*Cucurbita pepo* var. Black Beauty), cabbage (*Brassica oleracea convar. capitata* var. alba) (Felizeter et al., 2014), lettuce (*Lactuca sativa*) (Felizeter et al., 2012), maize (*Zea mays*) (Krippner et al., 2014), and wheat (*Triticum aestivum L.*) (Wen

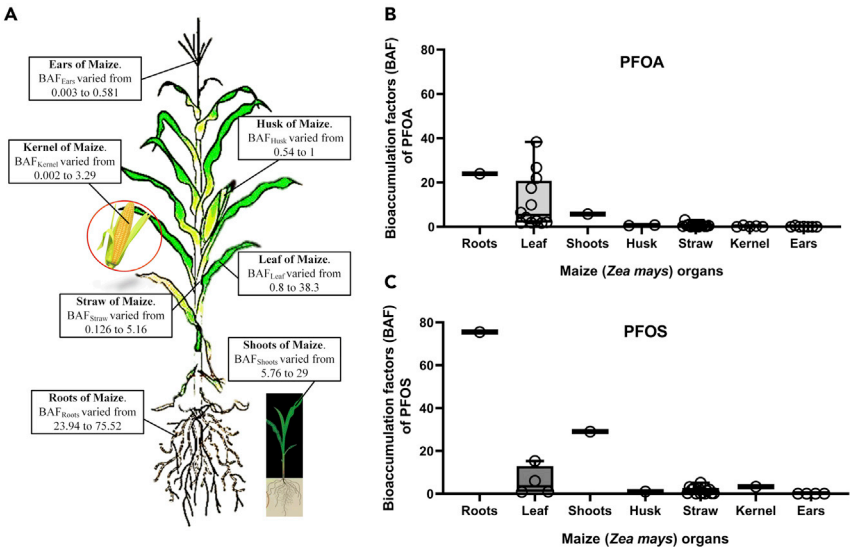

Figure 2. Bioaccumulation factor (BAF) for PFOA and PFOS in maize different organs

(A) BAF values of PFOA and PFOS in maize organs diagram.

(B) BAF values of PFOA in maize different organs.

(C) BAF values of PFOS in maize different organs with the error bars in boxplot. Data from Stahl et al. (2009) for straw and ears, from Krippner et al. (2015) for straw and kernels, from Lan et al. (2020) for leaf, straw, and kernel, from Wen et al. (2013) for root and shoots, and from Navarro et al. (2017) and Song et al. (2021) for root and leaf.

et al., 2014). Notably, emerging PFASs such as F-53B, 6:2Cl-PFAES, and 8:2Cl-PFAES were also found to be strongly sequestrated in root tissues (BAF ranged from 139.8 to 226.7) and hard to transport further to the shoots (translocation factor is 0.024 for 6:2Cl-PFAES and 0.005 for 8:2Cl-PFAES) in wheat (Lin et al., 2020).

Relatively limited research has been conducted in other species besides maize for understanding the enrichment of PFASs in different tissues (oat, wheat, soybean, carrot, welsh onion, celery, radish, tomato, lettuce, and beet plants) (Stahl et al., 2009; Lechner and Knapp, 2011; Wen et al., 2014; Bizkarguenaga et al., 2016; Liu et al., 2019). Because most of the studies that compare the enrichment of different tissues in plants include PFBA, PFBS, PFOA, and PFOS, these four most common PFASs were focused on in this review. Consistent with PFOA and PFOS in the maize tissues, BAFs of these four most common PFASs in other plants largely had the following order of a higher accumulation to less accumulation rate: straw > grain. Stahl et al. (2009) reported that the BAFs of these PFASs in oats ranged from 0.15 to 0.88 in straw, but only ranged from 0.017 to 0.054 in grain. The same trend occurred in wheat, with the BAF of these PFASs ranging from 0.27 to 3.2 in straw but only ranging from 0.009 to 1.286 in grain (Stahl et al., 2009; Wen et al., 2014; Liu et al., 2019). Moreover, other PFASs seem to follow this pattern regarding the distribution within the plant, based on the nine C4–C14 PFCA and three PFSA studied, with results confirming the order of accumulation rate straw > grain (Krippner et al., 2015; Stahl et al., 2009, 2013). Therefore, it can be comprehensively considered that the general order for the enrichment of PFASs in plants is root > straw > grain (Ghisi et al., 2019). It is worth noting that the leaves of plants also have a prominent enrichment effect for PFASs (Table 1). Liu et al. (2019) found that soybean grains and leaves planted on the same land had a PFOA concentration in soybean grains equating to 11.06 μg kg^{-1} d. w., whereas the concentration in leaves was as high as 3966.62 μg kg^{-1} d. w.

Distribution patterns of PFASs in different tissues

There was a relatively variable distribution pattern in PFASs in each matrix of the soil-plant system. PFASs in soil would dissolve in the void water, be taken up by root tissue, and are then distributed to other tissues. As previously discussed, during the root uptake process, long-chain PFASs such as PFOA and PFOS showed higher root uptake potentials. During translocation, long-chain compounds preferred to stay in the root. Thus, it is reasonable to suggest that the long-chain PFOA and PFOS would be the main composition, whereas short-chain PFASs would be dominated in aboveground plant tissues. Kim et al. (2019) investigated the uptake of PFASs from soil by rice and analyzed the relative distribution patterns of PFASs in

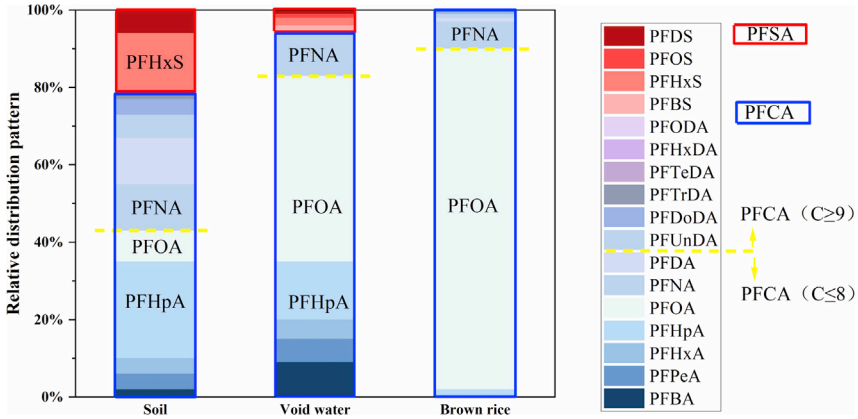

Figure 3. Distribution patterns of PFASs (%) in soil, void water, and brown rice
Reproduced with permission from Kim et al. (2019).

each matrix (Figure 3). They found that the composition of PFCA with ≤8 carbons and >8 carbons were similar (44.4% and 40.4%, respectively) in soil (Kim et al., 2019), whereas in the void water, PFCA with ≤8 carbons was the dominant form (83.8%). In brown rice, PFOA was the most predominant form (86.7%), relative to other PFCAs. This carbon-chain-based phenomenon, because of overall size selection, was also observed in carrots, onions, celery, and radish as shown in Table 1. PFOA was the predominant species in both soil and void water; therefore, the dominant state could have been distributed during the translocation. In addition, as PFOA has been reported to have relatively high water solubility compared with other PFCAs, PFOA could be transported into rice in void water and translocated to other tissues more readily.

PFASs-induced metabolomic profile changes in plants

Increased PFAS levels in plant tissues induce metabolomic changes *in vivo*, such as changes in purine metabolism, tricarboxylic acid cycle (TCA cycle), glyoxylate and dicarboxylate metabolism, pyruvate metabolism, nitrogen metabolism, and linoleic acid (fatty acid) metabolism in lettuce roots following PFOA and PFOS exposure (Li et al., 2020b). As a result, 23 antioxidants present in the lettuce roots were significantly altered after exposure to PFASs, reducing levels of arbutin and cinnamic acid and increasing levels of caffeic, cycloheterophyllin, equol, homovanillic acid, and hydroxytyrosol (Figure 4). In addition, 16 lipids were dysregulated in the root because of PFOA and PFOS-induced cellular stress including, decreasing isopentenyl pyrophosphate (IPP) and phytol. Moreover, 15 amino acids (nitrogen metabolism), 13 fatty acids (fatty acid metabolism), and carbohydrates, such as Succinic acid, uridine diphosphate glucose (UDGP), and sugars in roots were affected because of PFASs treatment. Decreased phytol levels induced by exposure to PFOA and PFOS in root tissue may lower the capacity of antioxidants in membranes and likely leads to lipid peroxidation in the plant. Alterations in these metabolites-impaired proper physiological activities in the roots, significantly impairs plant growth.

In addition to altered metabolite levels in lettuce roots, Li et al. (2020a) noted changes in metabolites under PFOA and PFOS exposure in lettuce leaves. Similar to dysregulations seen in roots, altered levels of amino acids, peptides, fatty acids, lipids, purines, and purine nucleosides were altered in leaves of the plant. In addition, several mineral elements and organic compounds in leaves were significantly altered, with Na, Mg, Cu, Fe, Ca, and Mo levels reduced by 1.8%–47.8%, whereas Zn was increased 7.4%–24.2%. Therefore, PFASs accumulation in the tissues could disrupt the metaplastic pathways *in vivo* and affect the normal growth of the plant.

DISTRIBUTION OF PFASS AMONG DIFFERENT PLANT SPECIES

Vegetables and cereals

Plants's uptake of PFASs from contaminated soil has been identified as an important pathway for PFASs to enter terrestrial food webs (Lechner and Knapp, 2011; Blaine et al., 2013; Wen et al., 2014; Krippner et al., 2015; Liu et al., 2019). It has been shown that the accumulative potential of PFASs in different plants varies according to the plant species, kinds of PFASs, seasons, and locations (Wen et al., 2014; Yu et al., 2018; Liu et al., 2019; Zhang et al., 2020). As shown in Table 2, the BAF values of main PFASs such as PFBA, PFBS,

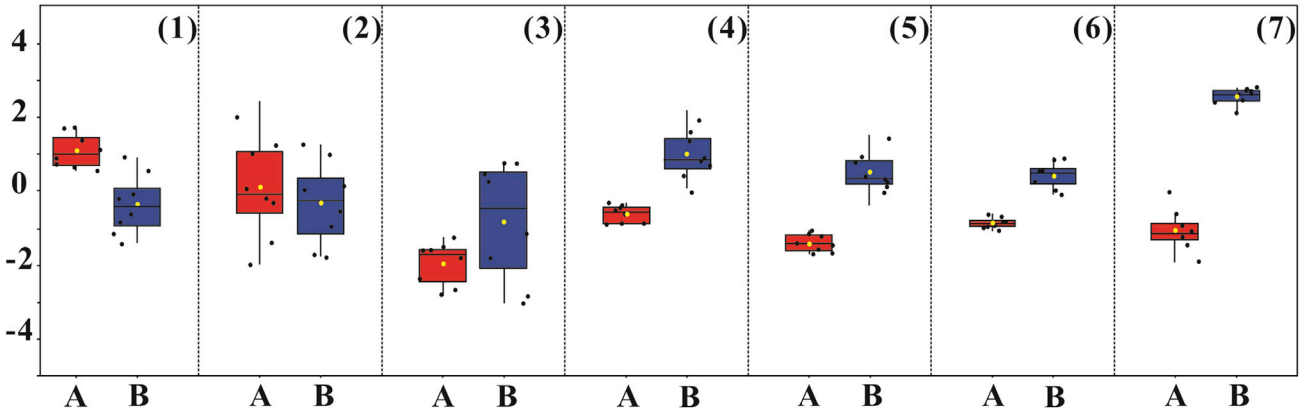

Figure 4. Box-whisker plots with error bars for the relative abundance of antioxidants

(1. Arbutin acid, 2. Cinnamic, 3. caffeic acid, 4. Cycloheterophyllin, 5. Equol, 6. Homovanillic acid, and 7. hydroxytyrosol) in lettuce root induced by (A and B) PFOA and PFOS exposure (A = control; B = 1000 ng/L of both PFOA and PFOS)

Reproduced with permission from Li et al. (2020b).

PFOA, and PFOS varied in one plant under similar conditions. For example, BAF of PFBA ranged from 20.63 to 191.45, whereas PFOA and PFOS ranged from 4.74 to 8.14 and about 5.67, respectively, in cabbage as investigated by Liu et al. (2019). Commonly, PFBA had a higher range of BAF than PFBS, PFOA, or PFOS as shown in Table 2. In addition, the soil properties also lead to the various BAF values of PFASs in plants; Higgins and Luthy (2006) pointed out SOC as a dominant parameter affecting PFASs sorption and thus causing the variation of PFASs accumulation in the plant. Besides, the BAF of PFBA in cabbage (20.63–191.45) was higher range than that in chives (20.63–44.04), cauliflowers (8.65–42.01), and welsh onions (3.55–31.92) (Liu et al., 2019). Similarly, the BAF of PFOS in chives (about 109) was much higher than that in the other three plants (about 5.67 in cabbages, about 5 in cauliflowers, and 1.5–4.67 in welsh onions). To sum up, all these factors contributed to PFASs accumulation in plants, and therefore, all these conditions should be considered when valuing the PFASs occurrence in plants.

The BAF of vegetables was greater than that of cereals, which was consistent with the findings of Liu et al. (2019), possibly because of higher protein and lipid contents of cereals versus vegetables (Figure 5A), particularly because of the high affinity of PFASs to proteins and lipids (Wen et al., 2016). In addition, the amount of water use and transpiration during growth may also play an important role in the variability in uptake and bioaccumulation rates, as vegetables need a greater amount of water than cereals during growth (Blaine et al., 2014a and 2014b). 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 organs aboveground through transpiration (Blaine et al., 2013).

Notably, some plants such as tea leaves and herbs are consumed as beverages or medicine worldwide, which are essential to human health directly. Zheng et al. (2014) analyzed PFOS and PFOA in 43 representative tea products and found that PFOA ranged from n.d. to 0.25 ng g^{-1} d.w. with the mean value of 0.04 ng g^{-1} d.w. detected in 33 samples, whereas PFOS ranged from n.d. to 0.083 ng g^{-1} d.w. detected only in 6 samples. In addition, Scheurer and Nodler (2021) showed that Trifluoroacetate (TFA), an ultrashort-chain perfluoroalkyl substance, was between 0.39 μg L^{-1} and 13 μg L^{-1} in tea/herbal infusion after the maximum brewing time, proving the accumulation of PFAS in tea/herbal beverages.

BAF variation in specific species

Clear differences in BAF values were found among different plant species. The BAF of PFASs in cabbage were highest (46.13 in average), followed by celery (39.89 in average) and lettuce (39.49 in average), which are all representative of green leafy vegetables; all these had greater BAF than that of fruit vegetables (pumpkins and peppers) and root vegetables (carrots and radish) (Figure 5B). In addition, the BAF of root vegetables (carrots and radishes) were much higher than that of flower vegetables (cauliflower) and fruit vegetables (tomatoes and peas), possibly because of the lack of casparian strip of carrots and radishes, which could help prevent chemicals from entering the organs aboveground via the apoplastic pathway

Table 2. BAF values of PFASs (i.e., PFOA, PFOS, PFBA, and PFBS) in different types of plants

Plant species		Compound	BAF	Reference
Cereals	Maize	PFBA	0.13–318.37	Blaine et al. (2013)
		PFBS	0.005–5.00	Krippner et al. (2015)
		PFOA	0.002–26.71	Liu et al. (2019)
		PFOS	0.104–15.29	Navarro et al. (2017)
	Wheat	PFBA	0.035–371.46	Stahl et al. (2009)
		PFBS	0.64–51	Liu et al. (2019)
		PFOA	0.084–6.45	Stahl et al. (2009)
		PFOS	0.004–139.8	Wen et al. (2014)
	Oat	PFOA	0.048–0.88	Stahl et al. (2009)
		PFOS	0.004–0.224	
Vegetables	Carrot	PFBA	7.56–355.04	Bizkarguenaga et al. (2016)
		PFBS	3–36.67	Lechner and Knapp (2011)
		PFOA	0.28–16.09	Liu et al. (2019)
		PFOS	0.43–0.55	
	Celery	PFBA	49.49–270.52	Bizkarguenaga et al. (2016)
		PFBS	2.21–21.4	Liu et al. (2019)
		PFOA	0.13–13.68	
		PFOS	0.05–27	
	Cucumber	PFOA	0.79–0.85	Lechner and Knapp (2011)
		PFOS	0.067	
	Lettuce	PFBA	13.00–488.67	Blaine et al. (2013)
		PFBS	2.02–14.5	Bizkarguenaga et al. (2016)
		PFOA	1.85–11.82	Liu et al. (2019)
		PFOS	0.10–1.67	
	Potato	PFOA	0.045–0.065	Lechner and Knapp (2011)
		PFOS	0.01	
	Radish	PFBA	2.92–210.36	Blaine et al. (2013)
		PFBS	1.27–114.00	Liu et al. (2019)
		PFOA	0.54–27.28	
		PFOS	0.066–26.43	
	Spinach	PFOA	12.47	Navarro et al. (2017)
		PFOS	4.50–4.63	
	Cabbage	PFBA	20.63–191.45	Liu et al. (2019)
		PFOA	4.74–8.14	
		PFOS	5.67	
	Chive	PFBA	20.63–44.04	Liu et al. (2019)
		PFOA	5.94–9.32	
		PFOS	109	
	Cauliflower	PFBA	8.65–42.01	Liu et al. (2019)
		PFOA	0.76–0.98	
		PFOS	5.00	
	Welsh onion	PFBA	3.55–31.92	Liu et al. (2019)
		PFOA	0.14–4.61	
		PFOS	1.5–4.67	
	Tomato	PFBA	12.20–35.43	Blaine et al. (2013)
		PFBS	0.42	Navarro et al. (2017)
		PFOA	0.035–0.11	
		PFOS	0.0022	
	Pea	PFBA	32.07	Blaine et al. (2013)
		PFBS	0.33	

(*Continued on next page*)

Table 2. *Continued*

Plant species	Compound	BAF	Reference
	PFOA	0.03	
	PFOS	0.03	
Pepper	PFBA	16.67–87.96	Liu et al. (2019)
	PFOA	0.292–0.848	
	PFOS	62.00	
Pumpkin	PFBA	104.10	Liu et al. (2019)
	PFOA	0.084	
	PFOS	3.00	

(Blaine et al., 2014a; Bizkarguenaga et al., 2016; Wen et al., 2016; Liu et al., 2019). Furthermore, the different compositions and surface areas of their root systems might also be an important reason for accumulative differences (Miller et al., 2016). Moreover, proteins could interact with PFASs and slow down the transport of PFASs in plants. The higher BCF observed in wheat compared to maize might be related to their higher protein content, as previously discussed (Wen et al., 2016). Therefore, it can be suggested that the BAF in vegetables is higher than that in cereals. The BAF levels of vegetables, from greatest to least, could best be represented as leaf vegetables > root vegetables > flower vegetables > shoot vegetables.

Human exposure to PFASs could be through direct or indirect ingestion of PFASs-containing food, inhaling PFASs-containing air, drinking PFASs-containing water, and using PFASs-containing household products, which provide health risks for potential consumers. The BAF of PFASs in readily consumable plant tissues showed large variations across species. According toLiu's study, the highest concentration of PFOA in vegetables exceeded 1800 ng g^{-1}, which is nearly 2–5 orders of magnitude higher than other vegetables purchased in markets (Jian et al., 2017; Sungur, 2018; Liu et al., 2019). Figure 5C exhibits the BAF of PFASs in edible parts of the vegetables and cereals. There were comparatively higher BAF values in celery leaf and stem, cabbage leaf, lettuce leaf, chives leaf, pumpkin fruit, and in pepper fruit relative to other edible plants. This suggests that these plants have a greater capacity for the uptake of PFASs from the soil and can store them in tissues that are commonly consumed by humans, potentially increasing health risks because of higher levels of PFASs.

RISK ASSESSMENT OF PFASS IN PLANTS

As previously determined, risk was evaluated by the daily intake rate (DI) of humans compared with the tolerable dietary intake (TDI). The formula used for DI was as follows (ATSDR):

$$DI = \frac{C_V \times C_I + C_S \times BCF}{BW} \quad \text{(Equation 2)}$$

where C_v is the concentration of PFASs in plants (ng g^{-1}). C_I is the minimum intake of plants (g day^{-1}) in either 160 g children or 180 g adult. C_s is the concentration of PFASs in soil (ng g^{-1}). BCF is the bioconcentration factor. BW is the body weight (b. w.) for children 35 kg and adults 70 kg. The TDI of PFOA and PFOS recommended by the European Food Safety (EFSA) were adopted from early studies, which were 1500 and 150 ng kg^{-1} b. w. per day, respectively. BFBA was similar to PFOA. Based on previous studies, the estimated daily intake of PFASs via tomato, cucumber, lettuce, or carrot consumption by children and adults were provided in Figure 6. In Figures 6A and 6B, the average daily vegetable consumption was 145 g and 207 g per day by children and adult, respectively, based on the Liaoning Bureau of Statistics (2018). As a result, the DI of PFBA was higher than that of PFOA and PFBS through ingestion of either tomatoes or cucumbers (Bao et al., 2020). Nevertheless, the half-life of PFOA could reach up to an average of 3.4 years (1.5–9.1 years), with the long-term ingestion of PFOA via vegetable consumption could have a high risk to people's health (Olsen et al., 2007). PFBS had a lower half-life because shorter-chain lengths than PFOA. However, Chen et al. (2019) noted that exposure to PFBS might be positively related to childhood adiposity, particularly for girls aged 5 years. Therefore, even though the DI values were all lower than that of TDI (18 ng kg^{-1} bw^{-1} day^{-1} for PFOA, 3600 ng kg^{-1} bw^{-1} day^{-1} for PFBA, and 1600 ng kg^{-1} bw^{-1} day^{-1} for PFBS) (Minnesota Department of Health, 2017; 2018a; 2018b), these compounds are of potential concern to human when consumed because of relatively long half-lives, and further supported (Bao et al., 2020). The average daily vegetable consumption was 160 g and 180 g per day by children and adult, respectively (Lal et al., 2020). (Figure 6C). The DI values of PFOS via lettuce and carrot consumption were both higher than that of the

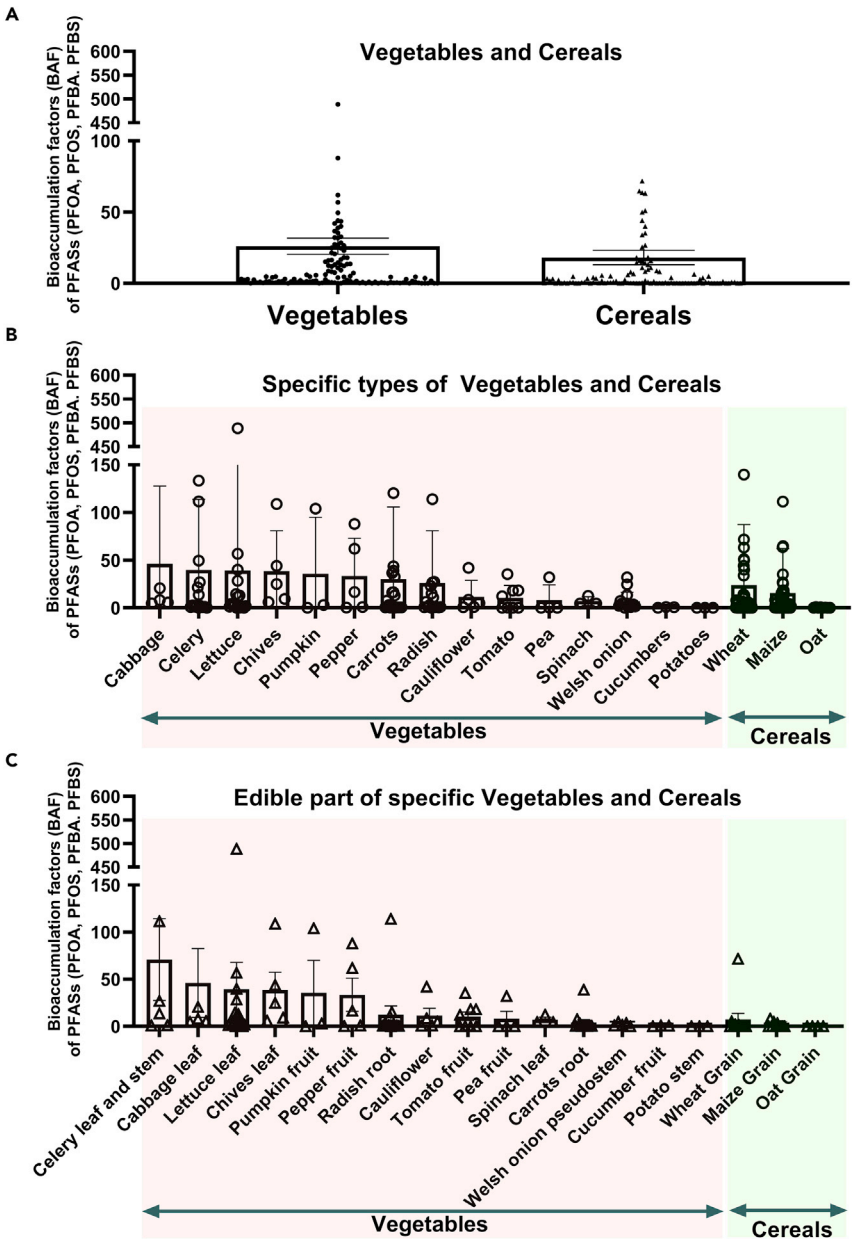

Figure 5. BAF values of PFASs in different vegetables and cereals with the error bars in boxplot

(A) BAF values of PFASs in vegetables and cereals.

(B) BAF values of PFASs in specific types of vegetables and cereals.

(C) BAF values of PFASs in edible parts of specific vegetables and cereals. Data from Stahl et al. (2009), Blaine et al. (2013), Krippner et al. (2015), Liu et al. (2019), Navarro et al. (2017), and Wen et al. (2014) for cereals, from Bizkarguenaga et al. (2016), Lechner and Knapp (2011), Liu et al. (2019), Blain et al. (2013), and Navarro et al. (2017) for vegetables.

TDI for children aged 2–6 years (1.1 ng g^{-1} bw^{-1} day^{-1}). Thus, PFOS pose potential risks for human health. Therefore, food safety risks of PFASs were reflected in these aspects, especially regarding the consumption of the edible components of plants where PFASs are stored and accumulated at greater levels.

Conclusions

In summary, PFASs can be taken up by plant root from the soil and accumulated throughout the plant through several internal and external processes; these processes are influenced by factors such as soil property and the

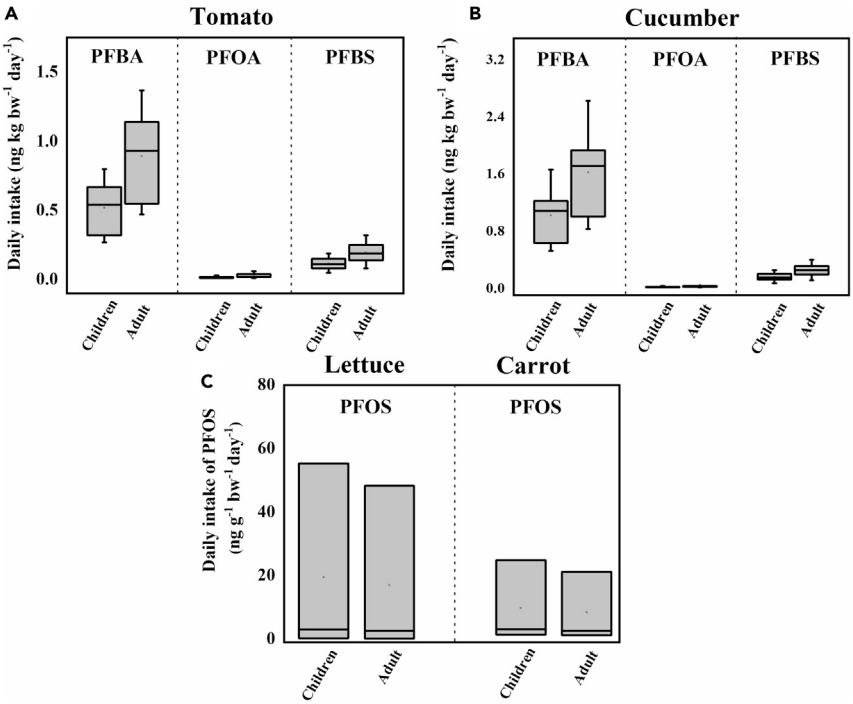

Figure 6. The boxplot with error bars for the estimated daily intake of PFBA, PFOA, and PFBS by children and adult via tomato and cucumber consumption (ng kg^{-1} bw^{-1} day^{-1})

(A and B) and PFOS via lettuce and carrot consumption (ng g^{-1} bw^{-1} day^{-1}) (C) .

The average body weight (b.w.) of children and adults are assumed to be 40 kg and 60kg, respectively in A and B and the b.w. are 35 kg and 70 kg in C. The data were abstracted from Bao et al. (2020) and Lal et al. (2020).

species of PFASs and plants. In addition, other environmental factors such as soil temperature, soil salinity, pH, soil moisture content, and cation exchange can also affect the uptake of PFASs through the root. Furthermore, the transport/distribution of PFASs from the root to aboveground tissues of the plant, is largely dependent on PFASs characteristics themselves, as longer-chain PFASs prefer to accumulate in root tissue, with shorter-chain compounds more easily transported to other tissues. PFASs accumulated in different organs and the enrichment of PFASs expressed by BAF in plants is root > straw > grain. PFOA, because of its high water solubility, constituted the main component of PFASs within the soil and plant. The accumulation of PFASs can further induce deleterious effects to the plant itself, such as altering the metabolomic profile in plant roots and leaves following exposure. Among different plants, the BAF values of vegetables was mostly higher than that of cereals. Specifically, BAF of vegetables, from greatest to least, was leaf vegetables > root vegetables > flower vegetables > shoot vegetables. In addition, DI values showed that the risk of PFASs in plants, particularly PFASs such as PFOA, PFBA, and PFOS, have a potential risk to humans throughdietary exposure.

Limitations of the study

Our review focused on translocation, bioaccumulation, and distribution of PFASs in plants. Some suggestions are provided as follows for future research:

(i) Exploring whether PFASs can be metabolized in plant tissue and if parent compounds or metabolites would pose a greater health risk. For example, if it would be possible for long-chain PFASs to have a defluorination reaction and turn into short-chain components, and if that would influence the potential distribution of short-chain PFASs in various plant tissues.

(ii) More comprehensive investigations should be conducted to identify the distribution patterns of PFASs in different tissues such as stem, leaves, shoots, and fruit which were not mentioned in this review.

(iii) The investigation on the translation of PFAS from soil to the special plants such as tea or herbs should be focused.

(iv) Vegetable consumption was not the main route of PFASs intake. Terrestrial and aquatic meat-based food sources also comprise a large dietary intake of food for humans. Thus, PFASs accumulation in the raw materials of these foods should be paid more close attention to for characterizing potential PFASs risks.

(v) Studies at genetic-level should be conducted to explain how some plant metabolites are altered and respond to PFASs-induced stress and variations among plant species.

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AUTHOR CONTRIBUTION

B. X. wrote the manuscript. J. D., C. Z., and H. C. collected the previous literature and summarized the contents. R. L. and Z. W. helped in acquiring all the data. J. T. M. helped to revise the manuscript. W. Q. and J. L. Z. conceived and designed the study. All the authors wrote and approved the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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