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Distribution, behaviour, bioavailability and remediation of poly- and per-fluoroalkyl substances (PFAS) in solid biowastes and biowaste-treated soil

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ABSTRACT

Aqueous film-forming foam, used in firefighting, and biowastes, including biosolids, animal and poultry manures, and composts, provide a major source of poly- and perfluoroalkyl substances (PFAS) input to soil. Large amounts of biowastes are added to soil as a source of nutrients and carbon. They also are added as soil amendments to improve soil health and crop productivity. Plant uptake of PFAS through soil application of biowastes is a pathway for animal and human exposure to PFAS. The complexity of PFAS mixtures, and their chemical and thermal stability, make remediation of PFAS in both solid and aqueous matrices challenging. Remediation of PFAS in biowastes, as well as soils treated with these biowastes, can be achieved through preventing and decreasing the concentration of PFAS in biowaste sources (i.e., prevention through source control), mobilization of PFAS in contaminated soil and subsequent removal through leaching (i.e., soil washing) and plant uptake (i.e., phytoremediation), sorption of PFAS, thereby decreasing their mobility and bioavailability (i.e., immobilization), and complete removal through thermal and chemical oxidation (i.e., destruction). In this review, the distribution, bioavailability, and remediation of PFAS in soil receiving solid biowastes, which include biosolids, composts, and manure, are presented.

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1. Introduction

Poly- and perfluoroalkyl substances (PFAS) are a group of synthetic compounds which do not occur naturally in the environment but are introduced through human activities (Buck et al., 2011). Because these substances are resistant to heat, water, and oil exposure, they are used extensively in a wide range of applications including in fire-fighting foam, non-stick cookware, fast-food wrappers, water-repellent fabrics (e.g., carpets and clothing), medical equipment, and plastic and leather products (Kannan et al., 2004). Aqueous film-forming foams (AFFF) used in firefighting, landfill leachate, sewage effluent, and solid biowastes, including biosolids (formerly called treated sewage sludge) and composts, are the major sources of PFAS in soil and surface- and groundwaters (Bolan et al., 2021, Sinclair and Kannan, 2006). Large volumes of biowastes (e.g., crop residues, biosolids, composts, and animal manures) are produced and applied to soil to improve soil health and productivity in different countries (Wijesekara et al., 2017). For instance, Australia is one of the leading countries that has demonstrated the beneficial use of biosolids for land application. About 75% of the total biosolids produced in Australia are used in agriculture for soil improvement and crop productivity, while the rest is utilised for land application in mining or industrial settings (Wijesekara et al., 2016). This practice also leads to PFAS input to soils, thereby reaching the food chain (Zhu and Kannan, 2019) (Fig. 1). Some of these biowastes are considered as diffuse sources of PFAS in soil and groundwater and the second significant sources of PFAS after AFFF (Eggen et al., 2010, Sepulvado et al., 2011, Weber et al., 2011). In 2017, the Australia New Zealand Biosolids Partnership (ANZBP) published a report on 'Assessment of Emergent Contaminants in Biosolids,' which indicated the presence of a range of PFAS including perfluorooctanesulfonic acid (or perfluorooctanesulfonate) (PFOS) and perfluorooctanoic acid (PFOA) in biosolids produced in Australia (Hopewell and Darvodelsky, 2017). Similarly, the presence of PFAS in biosolids and composts have been reported in many other countries including the USA (Choi et al., 2019), and countries in Europe (Goldenman et al., 2019).

Due to their extremely recalcitrant nature, the concentrations of PFAS chemicals are expected to increase in biowaste-applied soils with frequent applications, similar to heavy-metal accumulation from applications of biosolids (Wijesekara et al., 2017), or cadmium build-up due to phosphatic-fertiliser applications (Bolan et al., 2013). Thus, PFAS derived from biowastes is likely to enter the food chain through plant uptake, thereby leading to potential risks to human and ecological health (Choi et al., 2019). The fact that PFAS have been detected in influents, effluents, and sludges from a number of wastewater treatment plants (WWTPs) worldwide suggests that WWTPs and sludges are key links for widespread diffuse contamination of PFAS in the environment (Chen et al., 2012, Coggan et al., 2019, Higgins et al., 2005, Lin et al., 2010, Sinclair and Kannan, 2006). PFOA and PFOS concentrations ranging from < 0.5 to 1057.1 ng L^{-1} and < 0.06 to 461.7 ng L^{-1}, respectively, have been reported in influents and effluents of several WWTPs across the world (Guo et al., 2010, Lin et al., 2010). Similarly, mean concentrations of 0.021 mg kg^{-1} PFOS and 0.03 mg kg^{-1} PFOA were recorded in biosolid samples collected as part of Australia New Zealand Biosolids Partnership report on 'Assessment of Emergent Contaminants in Biosolids' (Hopewell and Darvodelsky, 2017). PFAS from PFAS-contaminated and biosolid-amended soils can enter earthworm and plant bodies posing a potential risk to the terrestrial food chain (Das et al., 2015, Wen et al., 2015, Zhao et al., 2016). It could be hypothesised that biogeochemical behaviour and fate of PFAS derived from biowastes in soil and groundwater would be different from those derived from AFFF. For example, Sepulvado et al. (2011) demonstrated that soil-derived organic-carbon normalized sorption coefficients were not accurate predictors of the release of long-chain PFAS inputs from biosolid application.

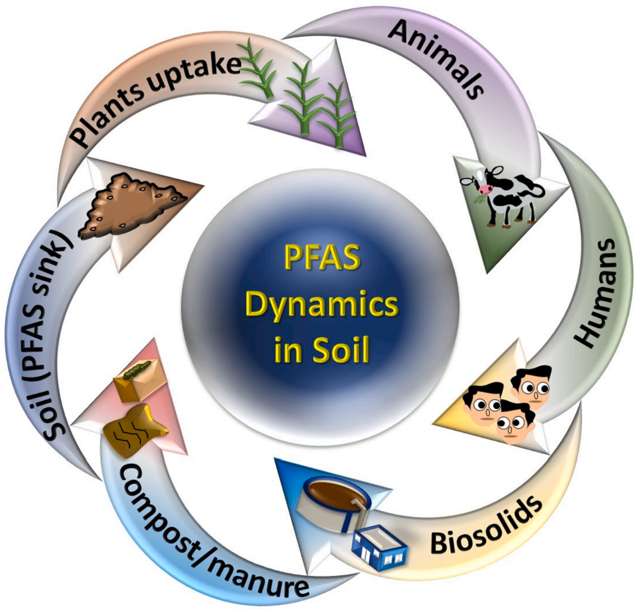


Fig. 1. PFAS dynamics in the biowaste-soil–plant-animal continuum.

Ingestion of PFAS is considered as the major human exposure pathway, which includes the potable use of contaminated water, consuming contaminated land-based food and seafood, and food packaged in PFAS-containing materials (Poothong et al., 2020). Ingestion of PFAS that are commonly present in a contaminated food or drink matrix can aggravate human health issues, including causing cancer and damaging reproductive and developmental systems (Liu et al., 2017, Sunderland et al., 2019).

PFAS are a large and complex manufactured chemical mixture in the environment, having high mobility in soil and water, and they are chemically, biologically, and thermally stable. Therefore, remediation of PFAS in both solid (i.e., soil and wastes) and aqueous (i.e., groundwater and storm water) media is challenging (Ross et al., 2018). For solid media such as soil and wastes, PFAS can be removed through abiotic and biotic degradation (bioremediation) or immobilized using adsorbents. Nevertheless, using a single bioremediation approach for PFAS under in situ conditions is challenging, and may not be sucessful because the process is very slow (Shahsavari et al., 2021). Abiotic degradation, including chemical and thermal degradation processes to remove PFAS from solid media and biowastes prior to soil application, has potential. But such processes are costly and energy intensive (Ross et al., 2018).

There have been a number of reviews on PFAS contamination resulting mainly from AFFF (Backe et al., 2013, Seow, 2013, Xiao, 2017). A few previous reviews highlighted PFAS contamination in water resources (Banzhaf et al., 2017, Chohan et al., 2020, Newell et al., 2020). However, no comprehensive review on PFAS derived from biowaste application to soil has been reported. The current review, therefore, focuses on the distribution, behaviour, and remediation of PFAS in solid biowastes, including biosolids, composts and manures, and in soils receiving these biowastes. After giving a brief account of the biowastes commonly used as soil amendments and the PFAS accumulation pattern in them, we discuss the contaminants' physico-biochemical behaviours in the biowaste matrices and biowaste-amended soils and give a critical appraisal of remediation approaches of the contaminants. This is the first major review article dedicated solely to the PFAS issue in agriculturally important biowastes that are gaining increasing popularity in recent days for sustainable land management practices.

2. Sources and soil application of biowastes

A wide range of commercially available organic amendments (biowastes), including animal manures, yard waste composts, crop residues, and biosolids, are used for improving soil quality (Quilty and Cattle, 2011). In view of the ever-growing energy demands and environmental impacts of inorganic fertilizers, extensive application of biowastes as a nutrient source will become more popular in future agricultural production (Park et al., 2011). The advantages and limitations of major biowastes, such as animal manures, composts, plant residues, and biosolids, are discussed in this section.

2.1. *Forms and values of biowastes*

With the growing human consumption of livestock and poultry products, a tremendous amount of animal manures (SI Table 1), including urine and flushing materials from confined poultry and animal industries, requires environmentally benign disposal (Shen et al., 2017). The enrichment of trace elements (e.g., Cu and Zn) derived from animal diets endows great nutritional potential of these organic wastes for promoting the plant growth (Bolan et al., 2010). Land disposal of animal manures, which is related to weather conditions, must comply with the legally prescribed maximum content of organic-N in the manures to prevent aquatic eutrophication due to infiltration and surface runoff (Aga et al., 2005). Typically, before land application, animal manures should be processed via composting, anaerobic digestion, granulation, and alum treatment (Wallace et al., 2018).

Composting is a controlled process to stabilize organic wastes (e.g., crop residues, organic residues, and animal manures) for recycling and disposal via aerobic biological decomposition (Chia et al., 2020). Compost improves the stability of soil aggregates (Annabi et al., 2011), lowers the soil bulk density (Somerville et al., 2018), reduces the chance of soil erosion, increases the soil porosity and water holding capacity (Hargreaves et al., 2008), and provides soil nutrients (e.g., C and N) (Benitez et al., 2003, Eghball, 2002). Household biowastes (e.g., yard waste) may contain slowly decomposable materials (e.g., plastic debris), while sewage sludge and animal manures can be contaminated by various chemical compounds. Thus, the quality of the final composting products should be rigorously monitored and controlled before field applications.

Sustainable management of plant residues (e.g., stalks, stems, leaves, and seed pods), which contain a significant amount of plant nutrients (Kumar and Goh, 1999), is vital in environmental protection practices. When applied as soil amendments, plant residues can provide important nutrients for subsequent crop uptake (Liu et al., 2010), thus enhancing agricultural productivity. Among the plant residues commonly used as amendments of agricultural soil are rice straw and corn straw, and they potentially can be used for humus production. Straw application to farmland has been reported to increase water holding ability (Ampofo, 2018), bulk density (Anik et al., 2017), aggregation (Yang et al., 2018), pesticide retention (Joshi et al., 2019), and microbial biomass and activity (Rong et al., 2018) of the amended soil, as well as the water use efficiency of plants (Yin et al., 2015).

With the ever-increasing number of high-volume WWTPs, a tremendous amount of biosolids are produced globally (SI Table 1) (Letcher et al., 2020). Typically, the organic matter and organic carbon account for 40–70% and 20–50% of biosolids, respectively (Torri et al., 2014). Biosolids contain a high level of plant nutrients such as N (~3.2%), P (~2.3%), and K (~0.3%). Biosolids can increase the soil organic matter (Sharma et al., 2017), macro- and micro- nutrients (e.g., N, P, K, Zn, Fe, Mn, and Cu), air and water permeability (Samaras et al., 2010), aggregate stability (Roca-Pérez et al., 2009), porosity (Rostagno and Sosebee, 2001), water holding capacity (Veeresh et al., 2003), cation exchange capacity, and microbial activity (Lloret et al., 2016) of agricultural soils, which subsequently enhance the soil fertility and crop yield (Walter et al., 2000) and inhibit soil erosion (Zerzghi et al., 2010).

2.2. *Limitations of soil-applied biowastes*

In addition to the above biowastes, food waste composts (Beiyuan et al., 2018), papermill and pulp sludges (Wang et al., 2019), and anaerobic digestates (Chen et al., 2017) have been used as soil amendments. Nevertheless, high concentrations of potentially toxic elements (e.g., Zn, Cu, Ni, Cd, Co, As, and Pb) (Zhi et al., 2020), contaminants of emerging concern (e.g., polychlorinated biphenyls (PCBs), disinfectants, detergents, pharmaceuticals and personal care products, and steroid hormones) (Bourdat-Deschamps et al., 2017, Verlicchi and Zambello, 2015), and pathogenic microorganisms (Jiang et al., 2020) in the biowastes have limited their land application potential because of plausible contaminant issues. If not managed properly, biowastes might also cause nutrient imbalances in the amended soil, surface water contamination with excess nutrients, and increased greenhouse gas emission. Several biowastes, therefore, require appropriate pre-processing or pre-treatment (e.g., alkaline stabilization of biosolids and anaerobic digestion) before they can be applied to farmland soils.

Recently, there has been increasing concern over the presence of PFAS, which are environmentally persistent, bio-accumulative, and toxic to animals and humans, in commercially available biowastes owing to their ubiquitous usage in myriads of industrial, agricultural, and household products. PFAS have been recently identified in organic composts (Choi et al., 2019), biosolids (Coggan et al., 2019, Lakshminarasimman et al., 2020, Lazcano et al., 2020, Ulrich et al., 2016), and plants (Wang et al., 2020) (SI Table 1). For example, high levels of PFASs were detected in vegetables (87 mg kg^{-1}), wheat grains (480 mg kg^{-1}), and maize grains (59 mg kg^{-1}) grown near the fluorochemical industrial parks (Wang et al., 2020). A recent survey of the nine Canadian sludge treatment systems reported that PFDA (<53 ng g^{-1}) was frequently detected in over 85% of the biosolids samples (Lakshminarasimman et al., 2020). The above biowastes are considered as organic materials for composting process and it is likely that the composting products contain PFAS. Researchers have shown that PFAS were found in different commercial compost products. Choi et al. (2019) reported the total PFOA + PFOS range in nine US municipal organic solid waste composts was 0.54 – 11.5 μg kg^{-1} detected in a backyard compost sample. Similarly, Lazcano et al. (2020) investigated the occurrence of 17 perfluoroalkyl acids (PFAAs) in six organic composts (manure, mushroom, peat, untreated wood, food waste, and yard waste). PFAAs were detected in all the composts, and higher PFAA concentration was found in food and yard waste (18.5 μg kg^{-1}) than in the other four (0.1–1.1 μg kg^{-1}) composts (Lazcano et al., 2020). In most cases, the conventional pre-treatments of biowastes have been found inefficient to remove PFAS, because of the extremely persistent nature of the contaminants (Choi et al., 2019). Finding an effective pre-treatment method to immobilize PFAS, and simultaneously tackle common issues associated with biowastes, is needed in order to promote sustainable use of biowastes for soil application.

3. Distribution and accumulation of PFAS in biowastes and biowaste-treated soil

3.1. *Wastewater effluents and sludge (biosolids)*

In the past, investigations on PFAS risk assessment and remediation mostly focused on concentrated point source contamination. However, diffused PFAS contamination via landfills, wastewater treatment facilities, and biosolids should not be overlooked (Table 1). It is important to note that while treated wastewater is considered as a point source of pollutant input, biosolid application can be considered as a diffuse or non-point source of pollutant input. For example, household wastewater containing PFAS (e.g., from non-stick coatings on cookware) in low concentration can reach WWTPs and tend to accumulate in biosolids (Masoner et al., 2020). The concentrations of PFAS within biosolids are dependent on the nature of the treatment processes from where PFAS are released (Fig. 1). Treatment of sludge is necessary to produce commercial biosolid-based products to meet the EPA Part 503 Biosolids regulations. Among four commercially available types of biosolids (heat treated, composted, blended, and thermally hydrolysed), only the

Table 1
Selected references on PFAS derived from biowastes input to soil.

Origin	Source	Country	PFAS content	References
Wastewater treatment plants (WWTP)	Biosolids	Australia	Persistent, bioaccumulative and toxic substances (PBTs) in biosolids Decabromodiphenyl ether (BDE-209) (<0.4–2300 ng g^{-1}) Perfluoroctanesulfonate (PFOS) (<LOD 380 ng g^{-1}) A national estimate of masses of PBTs accumulated in Australian biosolids reached 167 kg year^{-1} (BDE-209)	(Gallen et al., 2016)
WWTP	Sewage sludge and treated biosolids	Canada	PBDE concentrations in primary sludge: 230–82,000 ng g^{-1} Sludge: 530–8800 ng g^{-1} Treated biosolids: 420–6000 ng g^{-1}	(Kim et al., 2019)
WWTP	Biosolids	USA	PFOS: 403 ± 127 ng g^{-1} dw PFOA: 34 ± 22 ng g^{-1} dw Perfluorodecanoate (PFDA): 26 ± 20 ng g^{-1} dw The mean load of ΣPFASs in U.S. biosolids was estimated at 2749–3450 kg year^{-1} (1375–2070 kg is applied on agricultural land and 467–587 kg landfills)	(Venkatesan and Halden, 2013)
WWTP	Biosolids	Canada	Total 22 PFAS: 4.93 – 92.6 ng g^{-1} dw	(Letcher et al., 2020)
WWTP	Influent Effluent Sewage sludge	Guangzhou, China	Total PFAS: 19.6 – 232 ng L^{-1} in influents 15.5 – 234 ng L^{-1} in effluents 31.5 – 49.1 ng g^{-1} dw in sludge	(Pan et al., 2016)
WWTP	Limed biosolids	Mid-Atlantic region of US	Perfluorononanoic acid (PFNA): 25.1 ng g^{-1} dw PFOA: 23.5 ng g^{-1} dw PFOS: 22.5 ng g^{-1} dw	(Armstrong et al., 2016)
WWTP	Sewage sludge	Shanghai, China	Total PFAs (PPFAs): 126–809 ng g^{-1} dw PFOA: 23.2–298 ng g^{-1} dw	(Yan et al., 2012)
WWTP	Sludge	Nigeria	Perfluoroalkyl carboxylates: 10–597 pg g^{-1} Perfluoroalkyl sulfonates: 14–540 pg g^{-1}	(Sindiku et al., 2013)
WWTP (municipal, livestock and industrial)	Effluent and sewage sludge	Korea	PFOS sludge: 3.3–54.1 ng g^{-1} PFOA: wastewater – 2.3–615 ng L^{-1} influent: 3.4 ng L^{-1} effluent: 591 ng L^{-1} No PFCs were detected in livestock wastewater	(Guo et al., 2010)
Sewage treatment plants	Influent, effluent and sludge	Spain	Sludge samples – L-PFOS: 1.79 µg g^{-1} PFBA: 1.88 µg g^{-1} dw	(Campo et al., 2014)
WWTP	Influent and effluent	17 different provinces in China	Influent samples: Total PFAS was highest in Shanghai (12,000 ng L^{-1}) and lowest in Kunming (220 ng L^{-1}) Effluent samples: Total PFAS was highest in Qingdao (9,100 ng L^{-1}) and lowest in Kunming (250 ng L^{-1})	(Wang et al., 2020)
WWTP	Influent and effluent	Taiwan	PFOS (293 ng L^{-1}) and PFHxA (406 ng L^{-1}) were the highest values detected in influent and effluent samples Up to 10,000 ng L^{-1} PFAS were found in industrial WWTP	(Lin et al., 2010)
WWTP	Influents, effluents and sludge	China	Sludge: PFOS: 0.5 to 19.8 ng g^{-1} PFOA: 0.5 to 158.0 ng g^{-1}	(Chen et al., 2012)
WWTP	Influent, effluents and sludge	Thailand	Total PFAS: 674 ng L^{-1} in influent 1143 ng L^{-1} in effluent 1404 ng g^{-1} in sludge	(Kunacheva et al., 2011)
WWTP	Sludge	Hong-Kong	Total concentrations of perfluoroalkylsulfonyl-based chemicals in sludge: <100 ng g^{-1}	(Ma and Shih, 2010)
WWTP	Biosolids	USA	PFOS: 80–219 ng g^{-1}	(Sepulvado et al., 2011)
WWTP	Sludge	Greece	PFOS: 6.7 ng g^{-1} dw	(Arvaniti et al., 2012)
WWTP	Influents and effluents	Tianjin, China	Perfluorooctanoic acid (PFOA) Influents: 20–170 ng L^{-1} Effluents: 30–145 ng L^{-1} Sludge samples PFOS: 42–169 g kg^{-1} PFOA: 12–68 g kg^{-1} The mass flow: 26, 47, and 3.5 kg year^{-1} for perfluorohexanoic acid, PFOA, and PFOS	(Sun et al., 2012)
WWTP	Sludge	Japan and Thailand	Japan- PFCs: 124.95 g day^{-1} (PFASs: 49.81 g day^{-1}; PFCAs: 75.14 g day^{-1}) Thailand: PFCs: 55.04 g day^{-1} (PFASs: 12 g day^{-1}; PFCAs: 43.04 g day^{-1})	(Shivakoti et al., 2010)
WWTP	Influent, effluent, and sludge	Korea	PFOA and PFOS were dominant in influent and effluent samples (accounted for 66% and 49% of the total 11 PFAAs Up to 91 ng PFAAs g^{-1} was found in sludge samples	(Kim et al., 2016)

(continued on next page)

Table 1 (*continued*)

Origin	Source	Country	PFAS content	References
WWTP	Sludge	Czech Republic	High PFAS contamination in sludge samples (5.6 – 963.2 ng g^{-1})	(Semerád et al., 2020)
WWTP	Sludge	China	Total PFAS concentration ranged from 4.95 to 980 ng g^{-1} in sludge samples	(Na et al., 2020)
Crude organic kitchen waste and green waste	Compost	Switzerland	Compost Perfluorinated sulfonates, PFS: 1.0–23.6 μg kg^{-1} dw Perfluorinated carboxylates, PFCA: 1.3–9.9 μg kg^{-1} dw Digestate Perfluorinated sulfonates, PFS: 2.0–8.6 μg kg^{-1} dw perfluorinated carboxylates, PFCA: 2.4–6.6 μg kg^{-1} dw	(Brändli et al., 2008)
Compost and digestate	Compost	Switzerland	Median PFAS concentration (Sum of 6:2 fluorotelomer sulfonate (6:2 FTS), saturated/unsaturated fluorotelomer carboxylates (FT(U) CA), perfluorinated sulfonates (PFS), perfluorinated carboxylates (PFCA), fluorooctane sulfonamides (FOSA), fluorooctane sulfonamidoethanols (FOSE)) was 6.3 μg kg^{-1} dw, ranging from 3.4 to 35 μg kg^{-1} dw	(Brändli et al., 2007)
Soils amended with biosolids	Sewage sludge	Beijing, China	$\sum_9$PFCA ranged from 18 to 113 ng g^{-1} dw $\sum_3$PFSA ranged from 23.4 to 107 ng g^{-1} dw$\sum$PFAA ranged from 41.4 to 220 ng g^{-1} dw	(Wen et al., 2014)
Sludges generated from WWTP	Sludges	USA	Highest concentrations were PFDA ($\leq$990 ng g^{-1}), PFDDA ($\leq$530 ng g^{-1}), PFOA ($\leq$320 ng g^{-1}), and PFOS ($\leq$410 ng g^{-1})	(Washington et al., 2010)
Municipal biosolids	Biosolids	Shandong, China	PFOS 154.4 ng g^{-1} and PFOA 416.8 ng g^{-1}.	(Wen et al., 2014)
Compost	Five composts	Turkey	Mean PFOA and PFOS concentrations were between 26.1 and 102.0 ng g^{-1} and 0.211–0.649 ng g^{-1}, respectively.	(Sungur et al., 2020)
Biosolids generated from WWTP	Biosolids	Canada	Biosolid-amended soil exhibited increased concentrations of PFCA (0.1–19 ng g^{-1} dw)	(Lee et al., 2014)
Biosolids	16 biosolids	Spain	Biosolid amendment increased concentrations 1.5–14-fold for PFAS	(Navarro et al., 2016)
Biosolids		Australia	Annual load of PFOA in agricultural soils estimated 2.2 kg Annual load of PFOS in agricultural soils estimated 13 kg	(Gallen et al., 2016)
Biosolids/ soil mixture exposed to ambient outdoor conditions		USA	PFOA: 24.1 ng g^{-1} dw PFUnDA: 18.4 ng g^{-1} dw PFDA: 17.4 ng g^{-1} dw	(Venkatesan and Halden, 2014)
Sewage sludge		Shanghai, China	Predicted PFOA in agricultural land was 1.08 ng g^{-1} dw Predicted PFOA in agricultural land was 7.53 ng g^{-1} dw	(Yan et al., 2012)
Biosolids		China	PFOS ranged from 1.44 to 43.2 ng g^{-1} PFOA ranged from 1.21 to 28.5 ng g^{-1}	(Wen et al., 2015)
Biosolid	Sludge	USA	Perfluorooctanoic acid (PFOA) was a major homologue ($\sim$10-200 ng g^{-1} dw), followed by perfluorodecanoic acid ($\sim$3-170 ng g^{-1})	(Yoo et al., 2011)
Biosolid from a secondary wastewater treatment plant	Biosolids	Canada	Perfluorooctane sulfonamide in biosolid-augmented agricultural soil 41.87 to 622.46 ng g^{-1} dw	(Chu and Letcher, 2017)
Municipal biosolids	Biosolids	USA	PFOS (243 ng g^{-1}, dw) and PFDS (113 ng g^{-1}) and concentrations of PFOA (14.8 ng g^{-1}), PFHxS (3.03 ng g^{-1}) and PFUdA (5.32 ng g^{-1}).	(Rich et al., 2015)
Contaminated paper sludge	Biosolids	Germany	diPAPs and PFCAs are suggested to be the major contaminants in all four samples. In sample 1, diSAmPAP and its TPs (PFOS and EtFOSAA) are the dominating contaminants followed by diPAPs and its TPs.	(Bugsel and Zwiener, 2020)
Dewatered municipal biosolids	Biosolids	Canada	Perfluoroalkyl acids (PFAAs; PFHxS, PFOS, PFDS, PFOSA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFTA) were monitored in soil cores (0–0.3 m)	(Gottschall et al., 2017)
Biosolids-amended soils	Biosolids	Shandong, China	In two biosolids-amended soils, the sum of PFAA concentrations followed$\sum$PFOA (22.5 and 37.1 ng g^{-1}) > $\sum$PFOS (6.29 and 13.5 ng g^{-1}) > $\sum$PFHxS (0.048 and 0.085 ng g^{-1}).	(Zhang et al., 2018)

biosolids blended with materials containing no PFAS reduced perfluoroalkyl acids (PFAA) concentration in the treated product due to a dilution effect (Lazcano et al., 2019). Heating and composting treatments increased PFAA concentration in the products owing to the breakdown of PFAA precursors during the treatment processes. In addition, part of the PFAA precursors remained in biosolids after the treatments which can become a source of PFAA after application of biosolid-based products. The authors suggested that common post-treatments were ineffective in reducing PFAS contents in biosolid-based products. Therefore, controlling sources potentially contributing PFAS loads into wastewater treatment plants could be a better alternative (Lazcano et al., 2019).

PFAS can reach the sewage system in WWTPs through a number of industrial sources, including PFAS, fluoropolymer and AFFF manufacturing facilities (Prevedouros et al., 2006). The PFAS issue arises in sewage sludge because conventional WWTP systems are not efficient in the removal of the PFAS recalcitrant compounds during the treatment (Coggan et al., 2019). PFAS have been found in influents, effluents and biosolids from WWTPs across the globe (Chen et al., 2012). The WWTPs act as transporters for releasing the contaminants into the environment as effluents and biosolids (Becker et al., 2008) (Table 1). PFAS could be generated in WWTPs from very stable PFAA, which are formed from PFAS precursors, through a hydrophobic interaction between PFAA and hydrophobic moieties of organic solids present in the system (Zhang et al., 2013). Water treatment processes could increase the concentration of perfluorocarboxylic acids (PFCA) and perfluorosulfonic acids (PFSA) production by degrading the PFAS precursors and fluoride compounds, making the chemicals soluble, and highly persistent in the environment (Wang et al., 2011).

A number of PFAS, including PFOA and PFOS, were measured in Australian WWTP solids in recent years with concentrations ranging from 2.0 to 130 ng g^{-1} (Coggan et al., 2019) (Table 1). Higgins et al. (2005) observed total PFAS concentrations ranging from 55 to 3370 ng g^{-1} in domestic sludge in the USA. Similarly, Sun et al. (2011) reported total PFAS concentrations in digested domestic sewage sludge in Switzerland ranging from 28 to 637 ng g^{-1}, which was dominated by

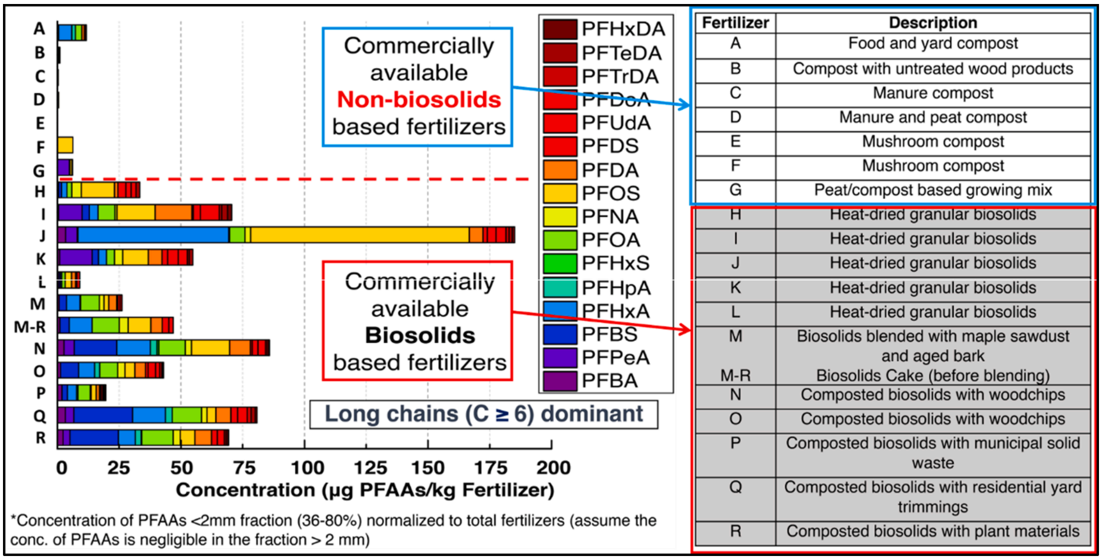
PFOS ranging in concentrations from 15 to 600 ng g^{-1}. Kallenborn (2004) observed that the domestic sewage sludges in Nordic countries had relatively low PFAS concentrations (0.6 to 15.2 ng g^{-1}). For example, PFOA and PFOS concentrations in the domestic sludge from Sweden ranged from 0.6 to 23.9 ng g^{-1} and 1.6 to 54.8 ng g^{-1}, respectively (Haglund and Olofsson, 2009). It has been estimated that the annual mean load of total PFAS in biosolids across the USA is 2749–3450 kg, of which around 1375–2070 kg PFAS reach agricultural land through soil application of biosolids (Venkatesan and Halden, 2013). Sepulvado et al. (2011) noticed that PFAS accumulated to 120 cm soil depth with concentration as high as 483 ng g^{-1} in field soils receiving continuous biosolid applications. Washington et al. (2010) investigated soils treated with biosolid applications in proximity to a WWTP treating sewage effluent from PFAS industries (Table 1). The PFOS and PFOA concentrations in the biosolid treated soil were as high as 408 ng g^{-1} and 312 ng g^{-1}, respectively. Sludge from the WWTP contained PFOA concentrations up to 1875 ng g^{-1} (Fig. 2). The application of biosolids from the WWTP was discontinued from 2007, and since then the concentration of PFAS in the sludge-applied surface soils decreased remarkably (Washington et al., 2010). The decrease was attributed to leaching loss to deeper soil over time. PFAS could be taken up by plants and farm animals from biosolid-applied agricultural soils suggesting a potential pathway to enter human bodies though dietary exposure (Domingo and Nadal, 2019, Navarro et al., 2017, Wen et al., 2016).

In addition to the direct sources of PFAS into WWTPs, a number of PFAS precursors could indirectly contribute to the final concentration of PFAS in the effluent and biosolids. The Organisation for Economic Cooperation and Development (OECD) listed 875 chemicals with the potential to produce PFAA through incomplete degradation of former chemicals. For PFOS, the precursors include derivatives and polymers of perfluoroalkyl sulfonyl or sulfonamide compounds. The PFOA precursors could have high diversity, including derivatives and polymers of perfluoroalkyl alcohols, amines, carboxylic acids, esters, ethers, and iodides (OECD, 2007). WWTPs could show a 9 to 352% increase in PFOA concentration in effluents compared to influents (Schultz et al., 2006). However, PFOS concentrations in sewage effluents is generally less than that of sewage influents owing to the retention of PFOS in the sludge (Yu et al., 2009). Becker et al. (2008) reported a 20-fold increase in PFOA concentrations from sewage influents compared to those in sewage effluents. In addition, 10% PFOA and 50% PFOS from the sewage influents were retained in the sludge solids (Becker et al., 2008).

Coggan et al. (2019) analysed 21 PFAS-containing liquid and solid samples to investigate PFAS release from 19 Australian WWTPs. The average concentration of PFAS in liquid and solid samples was 110 ng L^{-1} and 34 ng g^{-1}, respectively. The concentration of PFCA was higher in the discharged effluents than influents, and their sorption on the solid matrix within WWTPs increased with increasing chain length of fluoroalkyl from 0.05 to 1.22 log units. PFCA, such as perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), and perfluorodecanoic acid (PFDA), were increased in the discharged effluent compared to the influent. A PFSA of 6:2 fluorotelomer sulfonate (FTS), which was an intermediate degradant from C$_6$ based precursors, was found as the key chemical that enhanced the release of PFAS from Australian WWTPs (Coggan et al., 2019).

Gallen et al. (2018) reported the emission of PFAS through effluents, influents, and biosolids of 14 different WWTPs in Australia (Table 1). They identified nine PFAS and revealed that the total PFAS concentrations varied from 0.98 to 440 ng L^{-1} (influents), 21–560 ng L^{-1} (effluents), and 5.2–150 ng g^{-1} (biosolids). The total PFAS concentration was found 9.8 times higher in effluents than influents. The authors also estimated the Australian national annual load of PFOA and PFOS to be 65 and 26 kg in effluents, respectively, and correspondingly 2 and 8 kg in biosolids. Combining the effluents and biosolids, the estimated total annual national load of PFOA and PFOS through WWTPs, therefore, was 67 and 34 kg, respectively. In this connection, Nguyen et al. (2019) compared the amount of PFAS released from two large WWTPs (A and B) of Australia. A total 11 PFAS were detected, and the overall concentration ranged between 57 and 94 ng L^{-1} at WWTP A, and 31 and 42 ng L^{-1} at WWTP B. A higher level of 6:2 FTS (1.8–11 fold higher) than that of PFOA or PFOS at WWTP A indicated a replacement of PFAA by a fluorotelomer-based PFAS in this plant (Nguyen et al., 2019). The temporal trend of per capita mass load for PFOS was 67 µg day^{-1} inhabitant^{-1}, which was higher during October 2017 than other periods, suggesting the need for long term sampling and monitoring.

The content of PFAS and other pollutants depends on the sources of the biowastes. In Germany, the release of PFAA from households and industries raised a serious concern about applying biosolids to agricultural fields. Around 23.7% of total sewage sludge (1.8 billion tonnes on a dry weight basis) produced from municipal sewage sludge treatment plants in Germany was used as fertilizer (Roskosch and Heidecke, 2018). Owing to considerable quantities of PFAS released into the wastewater treatment plants, the application of sewage sludge on land can cause soil and groundwater contamination. The German directive then set the limit of applying toxic substances, including PFOA and PFOS, through



*Concentration of PFAAs <2mm fraction (36-80%) normalized to total fertilizers (assume the conc. of PFAAs is negligible in the fraction > 2 mm)

Fertilizer	Description
A	Food and yard compost
B	Compost with untreated wood products
C	Manure compost
D	Manure and peat compost
E	Mushroom compost
F	Mushroom compost
G	Peat/compost based growing mix
H	Heat-dried granular biosolids
I	Heat-dried granular biosolids
J	Heat-dried granular biosolids
K	Heat-dried granular biosolids
L	Heat-dried granular biosolids
M	Biosolids blended with maple sawdust and aged bark
M-R	Biosolids Cake (before blending)
N	Composted biosolids with woodchips
O	Composted biosolids with woodchips
P	Composted biosolids with municipal solid waste
Q	Composted biosolids with residential yard trimmings
R	Composted biosolids with plant materials

Fig. 2. Biosolid-based compost as a source of PFAS (Lazcano et al., 2019).

sewage sludge to 100 μg kg^{-1} dry weight (Stahl et al., 2018). The authors analysed a total of 201 sewage sludge and 45 biowaste samples and found concentrations of PFOS and perfluorohexane sulfonic acid (PFHxS) in sewage sludge and compost of 698 and 29 μg kg^{-1} dry weight, respectively. Sewage sludge contained more short-chain PFAS (85.9%) and fewer long-chained compounds (14.1%), whereas composts followed an opposite trend of containing 53.2% long-chained and 46.8% short-chain compounds. The higher level of short-chain PFAS in sewage sludge could be attributed to the preferential partition of short-chain PFAS in sludge during wastewater treatment process, whereas the slightly higher level of long-chain PFAS in composts could be attributed to the leaching loss of short-chain PFAS during the composing process. Thus, PFAS from land-applied sewage sludge was taken up easily by plants because the amendment contained abundant short-chain compounds that were difficult to remove because of their high mobility and increased solubility (Ghisi et al., 2019, Navarro et al., 2017). An estimated 15.3 kg of PFAA per year was accumulated in German agricultural lands through compost and sewage sludge applications (Stahl et al., 2018). Reports on PFAS in sewage sludge in developing nations like Nigeria revealed the overall concentrations of perfluoroalkyl carboxylate (or PFCA) and perfluoroalkyl sulfonate (or PFSA) ranged from 10 to 597, and 14 to 540 pg g^{-1}, respectively, in sludge discharged from industrial, domestic, and hospital WWTPs (Sindiku et al., 2013). The concentration of PFOS in hospital sewage sludge was low (539.6 pg g^{-1}) in Nigeria, and it possibly originated from medical equipment. A continuous monitoring for PFAS release from industrial and societal activities is needed not only in biowastes in developed countries but also in developing nations.

3.2. *Composts*

Municipal solid waste composts are nutrient-rich soil amendments and reduce the load of waste released into the environment. But they might carry toxic substances, including PFAS, when applied to the land (Allred et al., 2015, Choi et al., 2019) (SI Table 1). PFAS have been used in many compostable food-packaging products, because of their non-sticky and waterproof properties (Schaider et al., 2017). The PFAS found in food contact materials include PFSA, PFCA, fluorotelomer alcohols (FTOH), polyfluoroalkyl phosphate esters (PAP/di-PAP), FTS, and polyfluorinated ethers (PFE) (Choi et al., 2019). The content and variety of PFAS in food-contact materials depend on the type of packaging materials (e.g., greasy food) and food-production sources (Liu et al., 2013). In this context, the concentration and leachability of 17 PFAA chemicals in nine municipal solid waste composts (MSW) and one backyard compost were evaluated by (Choi et al., 2019) (Fig. 3). The PFAA concentration ranged from 28.7 to 75.9 μg kg^{-1} in MSW composts (with food packaging wastes), and from 2.38 to 7.60 μg kg^{-1} in backyard composts (without food packaging wastes). PFOA and PFOS were detected in all composts, and the majority of contaminants belonged to short-chain PFAA ($<C_6$) (>64%) and short-chain PFCA ($<C_7$) (>68%) groups. Besides, 6:2 FTS and 6:2 dipolyfluoroalkyl phosphate ester were identified in the three compost samples (Choi et al., 2019).

Similarly, Brändli et al. (2006) evaluated composts and digestates from 39 commercial composting and digestion plants in Switzerland in order to measure the loads of PFAS in these materials. The composts and digestates were being applied to agricultural soils as recycled fertilizers to improve the physicochemical conditions and health of soils. The total PFAS load in the compost and digestate samples was 6.3 μg kg^{-1} dry weight, and this value was compared with levels in sludge and sediment samples that had concentrations varying from 3.4 to 35.2 μg kg^{-1}. The type and source of raw materials, output materials, maturity levels, and seasonal variation of sample collection primarily influenced the type and content of PFAS in the samples (Brändli et al., 2006). A uniform distribution of PFAS, especially 6:2 FTS, perfluorobutane sulfonate, perfluorocarboxylic acid, and perfluorooctane sulphonamide, was observed in the digestates and composts from Switzerland, irrespective of the difference in sources and applications (Brändli et al., 2006). The 6:2 FTS, as the safer alternative to PFOS, was detected in 50% of the compost and digestate samples (n = 9 out of 18) from Switzerland, and the maximum level observed was 1.2 μg kg^{-1} dry weight. The concentration range of perfluorinated sulfonates (PFS) (except perfluorobutane sulfonate (PFBS)) was 1–24 μg kg^{-1} dry weight. The perfluorinated hexa- to dodecanoates were also detected in composts, and they had a concentration of 2.8 μg kg^{-1} dry weight (Brändli et al., 2007).

Sungur et al. (2020) evaluated five biological waste composts applied to the soil as fertilizer to determine the distribution of PFAS in wheat (*Triticum aestivum* L.) and corn (*Zea mays* L.) crops. The results revealed that the mean PFOA and PFOS concentrations in all five composts varied between 26.1 and 102 ng g^{-1} and 0.21–0.65 ng g^{-1}, respectively. The grain accumulation of PFAS was found the least in the case of both plants, while stalk was the main sink for PFAS accumulation. Thus, the stalk if used as fodder could incorporate PFAS in the food

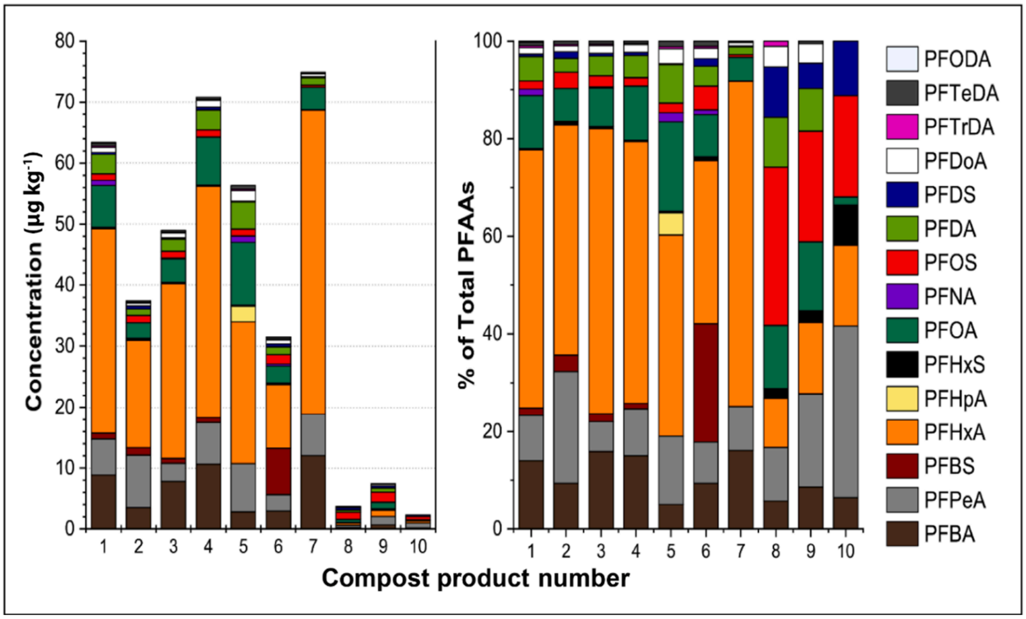


Fig. 3. Concentrations (μg kg^{-1}) of various PFAS compounds and their relation contribution (%) in various compost products (Choi et al., 2019).

chain. In contaminated vegetated soils, root vegetables could be vulnerable for increased PFAS uptake through the chemicals' direct contact with edible plant parts, and this warrants future studies under diverse soil types. Recently, a comprehensive review by Wang et al. (2020) reported the uptake of PFAS by field crops and vegetables. Many factors, including environmental conditions, the physicochemical nature of the PFAS, and plant-physiological responses, could influence the rate and extent of PFAS accumulation and distribution in plants.

3.3. Manures

As discussed earlier, several soil organic amendments can introduce PFAS into soils. For example, the application of sludge and biowastes to soils increases the loads of PFAS in the plant-growth media, which increases the possibility of PFAS transport and accumulation in plant parts including stalks, leaves, and grains (Sungur et al., 2020). In addition, food composts generated from households might carry a significant amount of PFAS to soils (Choi et al., 2019). When these plant parts and food materials (grains, stalks, straws, packaged food) are consumed by pets, farm animals, and humans, a substantial amount of PFAS could accumulate in animal and human excreta, which subsequently are used as manures and again the PFAS are transferred to soils (Domingo and Nadal, 2019). Monitoring of PFAS in animal excreta and manures is, thus, important to understand the exposure and toxicity of PFAS to humans and animals. Until now, though only few studies have covered this aspect of research, there is enough evidence about the accumulation of PFAS in pets and livestock.

In the USA, Ma et al. (2020) measured 15 PFAS in cat and dog faeces from the area around Albany, New York. Except for a few samples, almost all the PFAS were detected in the faeces samples. The PFAS concentration varied from 21.6 to 474 ng g^{-1} dry weight for dog faeces, which was higher than cat faeces, which had concentrations of 18.0–165 ng g^{-1} dry weight. Long-chained PFCA were dominant in all the pet faeces; some PFAS precursors were found at low concentrations. Cui et al. (2010) studied the excretion of PFOA and PFOS from rats during consecutive exposures to PFAS in diets. After 24 h, the release of PFOA through faeces and urine was 24.7–29.6% of the oral dose (5 and 20 mg kg^{-1} body weight day^{-1}), whereas the PFOS release was just 2.6–2.8% of the same oral dose. The accumulation of PFOA was smaller than PFOS in the rat body, but both compounds showed considerable release into the environment via rat excreta.

Farm animals also can potentially be exposed to PFAS through dietary pathways or grazing on contaminated land (Death et al., 2021). Biosolids are known sources of PFAS contamination to soils, and they are often utilized for growing pasture and fodder crops, allowing the contaminants to enter the animal body through their feeds. The dominant intake route of PFAA into dairy-cow bodies could be through the consumption of silage grown in PFAS contaminated soils (Vestergren et al., 2013). By providing a single 8 mg kg^{-1} body weight PFOS dose orally to beef cattle, Lupton et al. (2014) identified the major excretion route of the contaminants, which was through faeces (11% of the dose), and a substantial PFAS concentration persisted and accumulated in the cattle tissues. Therefore, PFAS load and release from manures, especially originating from farm animal excreta, warrant future research to eliminate PFAS bioaccumulation in crops, humans, and animals.

4. Behaviour and bioavailability of PFAS in biowastes and biowaste-treated soil

4.1. Behaviour of PFAS in biowastes and biowaste-treated soil

Behaviour of per- and poly-fluorinated alkyl substances (or per- and poly-fluoroalkyl substances) in the environment is governed by the length of the alkyl chain and functional groups (Bräunig et al., 2019). Furthermore, it affects the availability of PFAS in soils, wastes, or surface waters along with physicochemical properties of PFAS and total organic content (TOC) (Higgins and Luthy, 2006). It is difficult to identify the specific soil properties that are responsible for the sorption of PFAS to soils because of the complexity of the sorption process (Li et al., 2018). However, a major role has been played by hydrophobic interactions between PFAS and soil organic carbon, electrostatic interactions between soil organics, minerals, and functional groups of PFAS, and divalent cation binding to ligands (Bräunig et al., 2019, Jeon et al., 2011). Li et al. (2018) reported factors, such as clay content, organic carbon content, and pH of the soil, also affect the sorption of a few PFAS.

Though soil organic matter plays a key role in the sorption of PFAS (Zhi and Liu, 2018), other factors such as salinity and soil texture are also important in the sorption process (Askeland et al., 2020, Jeon et al., 2011). Dalahmeh et al. (2018) hypothesized that ionic strength from high mineral content in the soil affects sorption. High cation content in soil decreases the sorption of PFAS (Wang and Shih, 2011). For example, a soil with sugarcane had a high cation content (130 g kg^{-1}) and it exhibited the lowest concentration of PFAS in soil (Wang and Shih, 2011).

4.2. Bioavailability of PFAS in biowastes and biowaste-treated soil

Release of PFAS from biowastes, including biosolids and composts, may induce high bioavailability and bioaccumulation of PFAS in soil organisms and plants (Bizkarguenaga et al., 2016; Wen et al., 2015). The literature confirms the bioavailability of PFAS in soils (Table 2), and the adverse effects on living beings by accumulation of PFAS (Ghisi et al., 2019; Pérez et al., 2013). Therefore, bioaccumulation of PFAS can be found in plants and animals, and their influence in food chain is shown in Table 3.

Higher sorption capacities to soils and sediments have been exhibited by long-chain PFAS [$C_nF_{2n+1}COOH$ ($n \geq 7$), and $C_nF_{2n+1}SO_3H$ ($n \geq 6$)] rather than short-chain PFAS, which determines the bioavailability. Studies have shown that high organic carbon content and alkyl chain length increase the sorption of PFAS, whereas high pH reduces sorption (Higgins and Luthy, 2006), thereby impacting bioavailability of PFAS. A few studies have investigated the bioavailability of PFAS in biowaste-applied soils, while most studies have focused on bioavailability of PFAS in soils (Table 2).

Bräunig et al. (2019) studied the bioavailability of PFAS in three soil types collected from two different airport sites and explored the bioaccumulation in earthworms, phytoavailability in grass, and bioavailability in leachate for a range of PFAS types. PFOS had the highest concentration in soils, leachate, wheat plants, and earthworms for both airport firefighting training grounds, as well as waste soils (Table 2). A similar study was conducted by Hale et al. (2017) at an airport firefighting training ground in Norway. They reported a reduction of leaching of the dominant PFAS compound, PFOS by 94–99.9% for activated carbon, 29–34% for compost, and 28–40% for the activated carbon, compost and montmorillonite amended soil samples, respectively (Hale et al., 2017). Field soil samples collected from areas near potential PFAS contamination sources including industrial complexes and WWTPs have found to be exhibited a direct contamination of PFAS in minute concentrations (Kim et al., 2019, Liu et al., 2017, Zhu and Kannan, 2019).

Despite direct contamination, another possible pathway of PFAS contamination is via soil amendments such as biosolids. Wen et al. (2015) studied PFAS contamination through biosolids in seven soil samples taken from the same site and their effects on earthworms. In alkaline soils, high concentrations of PFOS and PFOA were detected in biosolids-amended agricultural fields in Changping, China, which had different soil organic matter contents (1.38 to 3.9%). A correlation was obtained between the bioavailability of PFOS and PFOA and the organic matter content, which also increased with the application of biosolids (Wen et al., 2015). Furthermore, PFOS was identified as the most abundant PFAS in biosolid and biosolid-amended soil samples (Sepulvado et al., 2011). The authors detected trace levels of PFAS in soil cores

Table 2
Selected references on the bioavailability of PFAS in soil.

Media	Location	Media description	PFAS type and Bioavailability (ng g^{-1})						References
			PFBA	PFOA	PFNA	PFDA	PFBS	PFOS	
Soil	Airport firefighting training grounds	In soil leachate	0.6	2.6	0.6	0.4	2	550	(Bräunig et al., 2019)
		In wheat plant	766	16	0.8	<0.25	550	1070	
		In earthworms	18	45	12	17	202	65,100	
	Waste soil	In soil leachate	370	0.6	<0.9	<0.6	0.5	90	
		In wheat plant	296	8	<0.34	<0.25	179	406	
		In earthworms	2.3	9.4	2	4.3	48	18,000	
	Soil in military aviation centre	Grass	11	0.6	<0.3	<0.2	7	32	(Bräunig et al., 2017)
		Egg yolk	0.25	0.53	0.12	0.24	0.07	70	
		Cow serum	0.55	0.24	4	9.7	<0.2	509	
	Airport firefighting training facility	In soil leachate	–	–	–	–	–	1.2–212	(Hale et al., 2017)
	Vicinity of fluoropolymer industry	In soil leachate	–	130	2.7	4.3	–	–	(Zhu and Kannan, 2019)
		Earthworm	–	270	13	26	–	–	
		Grass roots	–	47	1.4	2.5	–	–	
		Grass leaves		66	0.06	0.12	–	–	
		Tree leaves	–	410	2.7	3.2	–	–	
	Rice field	Void water	–	1–1760	–	–	–	ND-17.5	(Kim et al., 2019)
		Rice grain	–	ND-1.73	–	–	–	ND	
Biosolids application	Biosolids-amended soil	In earthworms		1.21–28.5				1.44–43.2	(Wen et al., 2015)
	Municipal biosolids	Biosolids	–	8–68	–	–	–	80–219	(Sepulvado et al., 2011)
	Wastewater treatment plant sludge applied soil	Sludge	–	320	–	990	–	–	(Washington et al., 2010)
	Biosolids-amended agricultural areas	In soil leachate	–	<0.05–1.573	–	–	–	<0.05–0.741	(Choi et al., 2017)
		Agricultural water	–	0.001–0.007	–	–	–	0.001–0.22	
	Compost amended soil	Carrot root peel	–	73–291	–	–	–	60–188	(Bizkarguenaga et al., 2016)
		Carrot root core	–	33–154	–	–	–	64–256	
		Carrot leaves	–	412–1468	–	–	–	320–777	
	Municipal biosolids- applied soil	Biosolid	48.6	78.5	–	93.5	–	49.7	(Blaine et al., 2013)
		Lettuce	25.5	20	4	3.5	3	102	
		Tomato	<0.07	<0.14	<2.8	<2.8	–	<0.14	
	Industrially impacted Biosolids-amended soil	Biosolid	–	15	6	10	–	319.5	
		Lettuce	266	197	57	48	205	83	
		Tomato	56	9	<2.8	<2.8	19.4	<0.14	
	Biosolids-amended agricultural field	Wheat roots	36.5	45	63	15	60	55	(Wen et al., 2014)

(at a depth of 120 cm) indicating vertical movement of PFAS, especially short-chain PFAS, in the soil profile over time (Sepulvado et al., 2011).

In different areas, such as the United States and Asia, soils from biosolids-applied agricultural fields have been investigated for the bioavailability of PFC (Washington et al., 2010, Wen et al., 2014). A high concentration of bioavailable PFAS (~5 μg g^{-1}) was identified in biosolids-applied fields in Alabama in the USA (Washington et al., 2010). Over time, the concentration of PFAS decreased in the top layers of soil, due to leaching into deep soils. However, the estimated half-life of PFAS ranged from 1 to 3 years based on chain length, and, therefore, PFAS remain in the surface and deep soils for long periods of time (Washington et al., 2010).

Blaine et al. (2013) conducted PFAA bioaccumulation studies in biosolids-treated industrial soils involving both greenhouse and field-scale plant growth experiments. Lettuce (*Lactuca sativa* L.) and tomato [*Lycopersicon lycopersicum* (L.) H. Karst.] were chosen for the study. The uptake and accumulation of perfluorobutanoic acid (PFBA) and PFPeA were high in lettuce and relatively less in tomato (Table 3). The authors found that bioaccumulation factors of lettuce for PFAA decreased with increasing number of CF_2 group of the compounds (approx. 0.3 log units per CF_2 group) (Blaine et al., 2013). In another greenhouse and field study, Lee et al. (2018) detected significant concentrations of PFAS such as PFCA in WWTP biosolids, print paper sludge, and compost. This study also reported that the uptake of PFCA by plants was more favourable for the short-chain congeners, such as PFBA, PFPeA, and PFHxA, and the transformation of PFCA compounds was confirmed by the presence of its metabolites. In a wheat crop (*Triticum aestivum* L.) grown in a soil treated with biosolids at various levels, nine PFCA and three PFSA were measured in roots, straw, husk, and grain. Total concentrations of PFAA in soil and different parts of the wheat plants increased with increasing application levels of biosolids (Wen et al., 2014). Similarly, Navarro et al. (2017) found PFAA in anaerobically digested, thermal-dried sludge and municipal solid waste compost. Tomato plants grown in soils treated with anaerobically digested, thermal-dried sludge accumulated high concentrations of PFBA and PFPeA (Navarro et al., 2017).

To date, only limited studies have been published concerning the bioavailability of PFAS in biowaste-treated soils. Nevertheless, there is evidence about bioaccumulation and phytoavailability of PFAS from biosolids, as well as the bioavailability of PFAS in soils (Lee et al., 2014, Wen et al., 2014). Given PFAS can accumulate at each step of the food chain, there is a possibility that biowaste-treated soils pose adverse risks on living beings (Ghisi et al., 2019, Giesy and Kannan, 2001). Bioaccumulation of PFAS can lead to environmental toxicity, as shown in Table 4. PFOA and PFOS have been reported as phytotoxic compounds and also germination and seedling growth suppressants in wheat plant systems (Lin et al., 2020, Zhou et al., 2016). Further, PFAS compounds have adversely affected microorganism such as green algae inhibiting their growth (Boudreau et al., 2003, Niu et al., 2019). Moreover, in vivo and in vitro studies have confirmed immunosuppressive, reproductive, neurological and endocrine disruptive toxicity of PFAS in animals (Grandjean and Clapp, 2015). Direct ingestion of PFAS through food, water and hand-to-mouth transmittance are potential pathways of PFAS exposure in human beings. Nevertheless, research on toxicological effects of PFAS on human beings are limited. Scientists suspect for potential relationship between PFAS and health defects such as immune suppression, cancer and obesity of children (Cui et al., 2020).

5. Remediation of PFAS in biowastes and biowaste-treated soil

Remediation of PFAS in biowaste-treated soil can be achieved

Table 3
Selected references on biowaste-derived PFAS reaching the food chain.

Origin	Sources	Country	PFAS content	References
Industrially impacted biosolids-amended soil, municipal biosolids-amended soil, and control soil	Greenhouse-grown radish (*Raphanus sativus*), celery (*Apium graveolens* var. dulce), tomato (*Lycopersicon lycopersicum*), and sugar snap pea (*Pisum sativum* var. *macrocarpon*)	USA	Industrially impacted biosolids were highest for Radish root – PFOA: 67 ng g^{-1} Celery shoot - perfluorobutanoate PFBA: 232 ng g^{-1} Pea fruit – PFBA: 150 ng g^{-1} Edible compartments of crops grown in municipal biosolids-amended soil and control soil - PFAA $<$ 25 ng g^{-1}	(Blaine et al., 2014)
Feld soil amended with a single application of biosolids Grown in spiked soil (~50 mg PFOS kg^{-1} soil (T1), ~5 mg Deca-BDE kg^{-1} soil (T2) and a mixture of both, ~50 mg PFOS and ~ 5 mg Deca-BDE kg^{-1} soil (T3)	Spinach (*Spinacia oleracea*) and tomato (*Solanum lycopersicum* L.) Corn (*Zea mays*)	Spain	PFAS - spinach Control: 1.72 ng g^{-1} dw T1: 5.33 ng g^{-1} dw T2: 0.99 ng g^{-1} dw PFASs in tomato T1: 61.3 ± 8.04 ng g^{-1} dw T2: 3.47 ± 1.55 ng g^{-1} dw Corn (T1) -PFOS Root: 254 ± 72.3 (µg g^{-1} dw) Leaf: 23.1 ± 6.13 (µg g^{-1} dw) Corn (T3) -PFOS Root: 133 ± 52.6 (µg g^{-1} dw) Leaf: 24.8 ± 7.37 (µg g^{-1} dw) Earthworms - PFOS 79.50 ± 7.35 µg g^{-1} dw	(Navarro et al., 2017)
Co-contaminated soil.	wheat and rapeseed	China	PFASs - wheat and rapeseed Roots: 332–1411 ng g^{-1} Shoots: 39.6–821 ng g^{-1}	(Zhao et al., 2017)
Compost amended soils fortified with perfluorooctanoic acid (PFOA), perfluorosulfonate acid (PFOS) and perfluorosulfonamide (FOSA)	Carrot and lettuce (greenhouse)	Spain	Highest carrot bioconcentration factors (BCFs) Leaves PFOA: 0.6 and PFOS: 3.4 lower values were in the core (0.05–0.6) and peel (0.05–1.9)	(Bizkarguenaga et al., 2016)
Grown in nutrient solution	Grass (*Bromus diandrus*)	Spain	Plants absorbed (per g) PFBA: 31 µg PFBS: 14 µg, PFDA: 12 µg PFHxS, PFOA, PFOS: 18 µg	(Garcia-Valcarcel et al., 2014)
Industrially impacted biosolids-amended soil, a municipal biosolids-amended soil, and control soil (greenhouse)	Lettuce (*Lactuca sativa*) and tomato (*Lycopersicon lycopersicum*)	U S A	Industrially contaminated biosolids Lettuce perfluorobutanoic acid (PFBA): 266 ng g^{-1} perfluoropentanoic acid (PFPeA): 236 ng g^{-1} Tomato PFBA: 56 ng g^{-1} PFPeA: 211 ng g^{-1}	(Blaine et al., 2013)
Soil mixed with contaminated sewage sludge	potato, carrot, and cucumber	Germany	peeled edible parts µg kg^{-1} (dw) Potato (Tub 1) PFOA: 2.9 ± 0.3 Potato (Tub 2) PFOA: 7.7 ± 0.9 PFOS: 0.7 ± 0.1 Carrot (T1) PFOA: 31.3 ± 2.5 PFOS: 0.5 ± 0.03 Carrot (T2) PFOA: 30.8 ± 1.8 PFOS: 18.4 ± 2.5 Cucumbers (T1) PFOA: 11.3 ± 0.4 Cucumbers (T2) PFOA: 23.8 ± 1.0 PFOS: 1.3 ± 0.2	(Lechner and Knapp, 2011)
Field experiments- soils amended with biosolids (dw, t/ha/y) Control- 0 Plot 4–36.0	Wheat (*Triticum aestivum* L.)	China	ΣPFAAs (ng g^{-1} dw) Control Grains and straw: 0 Plot 4 Grains: 35.6 and Straw: 178	(Wen et al., 2014)
Variant 1: control PFOA/PFOS 0 mg kg^{-1} soil Variant 6: 50 mg PFOA/PFOS kg^{-1} soil (Mitscherlich Pots)	Spring wheat, oats, potatoes, maize, and perennial ryegrass	Germany	Maize ear V1 (µg kg^{-1} dw) PFOA and PFOS: 0 V6 (µg kg^{-1} dw) PFOA: 440 and PFOS: 288 Oat grain V1 (µg kg^{-1} dw) PFOA and PFOS: 2 V6 (µg kg^{-1} dw) PFOA: 1480 and PFOS: 124	(Holly Lee et al., 2014)

(*continued on next page*)

Table 3 (*continued*)

Origin	Sources	Country	PFAS content	References
			Potato tuber V1 (μg kg^{-1} dw) PFOA and PFOS: 0 V6 (μg kg^{-1} dw) PFOA: 52 and PFOS: 34 Wheat grain V6 (μg kg^{-1} dw) PFOA: 1110 and PFOS: 34	
Reclaimed water augmented with varying concentrations (0.2–40 μg L^{-1}) of PFAAs (greenhouse)	Lettuce and strawberry	USA	Highest concentration applied (40 μg L^{-1}), Strawberry root – PFHxA: 5450 ng g^{-1} shoot – PFBA: 3900 ng g^{-1} fruit – PFPeA: 11,500 ng g^{-1} Lettuce 2% OC (organic carbon) soil PFBA: 15 μg g^{-1} PFNA: 47 ng g^{-1} 6% OC (organic carbon) soil PFBA: 5 μg g^{-1} PFNA: 21 ng g^{-1}	(Blaine et al., 2014b)
Corn silage cultivated in a land contaminated with perfluorinated alkylacids (PFAAs), as a result of illegal waste disposal	Sheep	Germany	Liver - PFOS Sheep 1: 885 μg kg^{-1} Sheep 2: 1,172 μg kg^{-1} Control: 1.5 μg kg^{-1} Muscle tissue – PFOS Sheep 1: 24.4 μg kg^{-1} Sheep 2: 35.1 μg kg^{-1}	(Kowalczyk et al., 2012)
Municipal solid waste compost and wastewater treatment plant biosolids. Multi species soil systems (MS.3)	Soil invertebrates (*Eisenia andrei*)	Spain	Earth worms from biosolid-amended soils PFASs: 9.9–101 ng g^{-1} dw Control: 1.76 ng g^{-1} dw BAF: 2.2–198	(Navarro et al., 2016)
Biosolids-amended soils without any additional spiking to earthworms	Earthworms (*Eisenia fetida*)	China	The bioaccumulation factors PFOS: 1.54–4.12 PFOA: 0.52–1.34 g(soil)/g(worm) PFOS and PFOA concentrations exhibited positive influence on accumulation of PFOS and PFOA in earthworms	(Wen et al., 2015)
diPAPs and PFCAs present in WWTP and paper fiber biosolids, amended soil. (greenhouse microcosm)	*Medicago truncatula* plants.	Canada	Plant accumulation of endogenous PFCAs present in the biosolids (0.1–138 ng g^{-1}) and those produced from 6:2 diPAP degradation (100–58 000 ng g^{-1}) observed within 1.5 months of application	(Lee et al., 2014)
Municipal biosolids applied soil without any additional spiking to earthworms	*Earthworms (Eisenia fetida)*	USA	worm concentrations were also highest for PFOS (683 ng g^{-1}) and PFDS (28.1 ng g^{-1}) with lower concentrations of PFOA (4.76 ng g^{-1}) and PFNA (3.98 ng g^{-1}).	(Rich et al., 2015)
Compost amended soils	wheat and corn	Turkey	the total perfluorinated compounds migration in of wheat and corn decreased as follows: stalk > leaf > grain.	(Sungur et al., 2020)
Biosolid-amended fields	Grass samples	USA	Per fluorooctanoic acid (PFOA) was a major homologue (∼10 – 200 ng g^{-1} dry wt), followed by perfluorodecanoic acid (∼ 3 - 170 ng g^{-1}). PFOS in plants (1–20 ngg^{-1}) generally was less than or equal to most PFCAs.	(Yoo et al., 2011)
Biosolids-amended soils	Alfalfa, Lettuce, Maize, Mung bean, Radish, Ryegrass, Soybean.	China	PFOS concentrations in roots range from 212.4 to 723.6 ng g^{-1}, while the PFOA concentrations in roots range from 703.4 to 4310.3 ng g^{-1}.	(Wen et al., 2016)
Biosolids-amended soils	alfalfa, lettuce, maize, mung bean, radish, ryegrass, and soybean	China	The root concentration factors of N-EtFOSAA ranged from 0.52 to 1.37 (pmol/g$_{root}$)/(pmol/g$_{soil}$)	(Wen et al., 2018)
Biosolids-amended soils	Maize	China	The distribution of $\sum$PFOA followed the order of roots > leaves > straws, while that of $\sum$PFOS was roots > straws > leaves.	(Zhang et al., 2018)

through decreasing the concentration of PFAS in biowaste sources (i.e., source control). Mobilizing amendments including surfactants and desorbing agents, can be applied to increase the bioavailability and mobility of PFAS in biowastes and in soil treated with these biowastes (Blaine et al., 2014). The mobilized PFAS can, subsequently, be removed through phytoremediation, soil washing, or destruction. Immobilizing amendments, such as sorbent materials, can be used to decrease the bioavailability and mobility of PFAS, thereby minimising their uptake by plants and leaching to groundwater (Sørmo et al., 2021). In the case of mobilizing techniques, there is a potential for leaching of the mobilized PFAS in the absence of active plant growth for PFAS uptake. But, in the case of immobilization techniques, there is a need for regular

Table 4
Selected references on environmental toxicity of PFAS compounds.

Source	End point	Test organism	Toxicity	References
Test range (mg L^{-1}) PFBA 10–3000 PFOA – 3–1000 PFOS – 0.03–10	Toxicity	Zebrafish embryos	The EC_{50} in 144 h PFBA: 2200 mg L^{-1} PFOA: 350 mg L^{-1} PFOS: 1.5 mg L^{-1}	(Ulhaq et al., 2013)
Exposure to various PFOS concentrations (0–8 mg L^{-1}) from 6 to 120 h post-fertilization (hpf)	Embryo toxicity	Zebrafish embryos	120 hpf $LC_{50:}$ 2.20 mg L^{-1} $EC_{50:}$ 1.12 mg L^{-1}	(Huang et al., 2010)
After 24-hour exposure to PFC	Cytotoxicity	Human placental choriocarcinoma cell line JEG-3	EC_{50} - µM PFOS: 107–125 µM PFDoA and PFNA: 181–220 µM PFOA: 594–647 µM	(Gorrochategui et al., 2014)
Treated with perfluorinated acids in a range of concentration from 1 to 5000 µM for 4, 24 and 72 h.	Viability	Human colon carcinoma (HCT116) cells	EC_{50} (µM) in 24 h: PFOA: 937.1 ± 67.9 µM PFHxA: 4153.9 ± 14.6 µM PFHpA: 1386.0 ± 56.9 µM	(Kleszczynski et al., 2007)
Exposure to 0–1811 µM PFOA-F I agar plates	Shoot fresh weigh Root fresh weight Root length	Model plant species *Arabidopsis thaliana.*	EC_{50} (µmol F/L) PFOA in 21d: Shoot fresh weight: 316.7 ± 34.8 µM Root fresh weight: 401.0 ± 42.9 µM Root length: 904 ± 93.4 µM	(Yang et al., 2015)
Stock solutions of PFOS and PFOA were added to the six soils at seven different concentrations	Phytotoxicity	*Brassica chinensis*	EC_{50}- PFOS: 95 to > 200 mg kg^{-1} PFOA: 107–246 mg kg^{-1}	(Zhao et al., 2011)
25 mL solutions of 11 different PFOA levels: (0 (control), 0.02, 0.2, 2, 20, 200, 800 and 1600 mg kg^{-1}	Germination and seedling growth	Wheat (*Triticum aestivum* L.)	PFOA - EC_{50} (mg kg^{-1}) Germination rate: 819.0 mg kg^{-1} Height of plant: 500.3 mg kg^{-1}	(Zhou et al., 2016)
	L. gibba- wet weight *D. pulicaria and* *D. magna - immobility*	Green algae *Selenastrum capricornutum* and *Chlorella vulgaris,* the floating macrophyte *Lemna gibba,*and invertebrates *Daphnia magna* and *Daphnia pulicaria.*	PFOS: 50% inhibition of growth (IC_{50}) – *L. gibba*: 31.1 mg L^{-1} (ww). *D. pulicaria:* 134 mg L^{-1} *D. magna*: 67.2 mg L^{-1} Significant adverse effects ($p \le 0.05$) for all organisms: 134 mg L^{-1}	(Boudreau et al., 2003)
Sexually mature fish were exposed via the water for 21 d to 0 (control), 0.03, 0.1, 0.3, or 1 mg PFOS L^{-1}	Reproductive and developmental toxicity	Fathead minnow (*Pimephales promelas*)	A concentration of 1 mg PFOS L^{-1} was lethal to adults within two weeks EC_{50} (95% confidence interval) in 21 d: 0.23 (0.19–0.25) mg L^{-1}	(Ankley et al., 2005)
Exposed to concentrations of PFOS – 0, 6.25, 12.5, 25, 50, 100 mg L^{-1} PFOA – 0, 62.5, 125, 250, 500, and 1,000 mg L^{-1}	Acute and chronic toxicities	*Daphnia magna* and *Moina macrocopa* and *Oryzia slatipes*	EC_{50} in 48 h *D. magna* PFOS: 37 mg L^{-1} PFOA: 477 mg L^{-1} *M. macrocopa* PFOS: 18 mg L^{-1} PFOA: 200 mg L^{-1}	(Ji et al., 2008)
Measured concentrations 0.001–25 (mg L^{-1}) 6:2,8:2, and 10:2 saturated fluorotelomer carboxylic acids (FTsCA) and unsaturated (FTuCA) fluorotelomer carboxylic acids	Toxicity	Fresh water algae, *Chlorella vulgaris* and *Pseudokirchneriella subcapitata* and amphipod, *Hyalella azteca*	C. *vulgaris* most sensitive species, with EC_{50} – mg L^{-1} 6:2FTsCA: 26.2 mg L^{-1} 6:2FTuCA: 31.8 mg L^{-1} 8:2 FTuCA: 1.1 mg L^{-1} 10:2FTsCA: 4.2 mg L^{-1} H. *azteca* most sensitive to LC_{50} – mg L^{-1} 8:2FTsCA: 5.1 mg L^{-1} 10:2FTuCA: 3.7 mg L^{-1}	(Mitchell et al., 2011)
Tested concentrations root elongation - (mg L^{-1}) PFOS – 6.25–200 PFOA – 62.5–2000	Root elongation in plants	Green neon shrimp (*Neocaridina denticulate*), Planarian, Snail Cucumber, Pakchoi, Lettuce	96 h -LC_{50} Green neon shrimp PFOS: 10 mg L^{-1} PFOA: 454 mg L^{-1} Planarian PFOS: 23 mg L^{-1} PFOA: 337 mg L^{-1} Snail PFOS: 178 mg L^{-1} PFOA: 672 mg L^{-1} Cucumber, Pakchoi, Lettuce EC_{50} (mg L^{-1}) PFOS: 99 to > 200 PFOA: 263 – 1254	(Li, 2008)
	Acute and subchronic toxicity	Adult male C57BL/6J mice		(Xing et al., 2016)

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Table 4 (*continued*)

Source	End point	Test organism	Toxicity	References
Exposure to subchronic oral toxicity of PFOS at 2.5, 5, 10 mg PFOS kg^{-1} BW day^{-1} for 30 days			Perfluorooctane sulfonate (PFOS) - acute oral LD_{50} value 0.579 g kg^{-1} (BW)	
Cells exposed to nine different concentrations (2 µM to 20 mM) of perfluorinated acids	Cytotoxicity	Two different mammalian cell lines promyelocytic leukemia rat cell line (IPC-81) and the rat glioma cell line (C6) and one marine bacteria *Vibrio fischeri*	EC_{50} (µM) IPC-81 Perfluorohexanoic acid: 3715.4 ± 85.6 Perfluoroheptanoic acid: 1778.3 ± 41.9 Perfluorooctanoic acid: 676.1 ± 46 C6 Perfluorohexanoic acid: 7943.3 ± 365.9 Perfluoroheptanoic acid: 3981.1 ± 275.2 Perfluorooctanoic acid: 676.1 ± 46.7 Vibrio fischeri Perfluorohexanoic acid: 4265.8 ± 393.5 Perfluoroheptanoic acid: 3020.0 ± 69.5 Perfluorooctanoic acid: 1380.4 ± 138.8	(Mulkiewicz et al., 2007)

examination of the long-term stability of the immobilized PFAS (Bolan et al., 2021). Some of the selected reports on the application of various PFAS remediation technologies, which include source control, soil flushing, phytoremediation, immobilization, and destruction or degradation, are summarized in SI Table 2, Table 5, and Fig. 4.

Table 5

Selected references on the remediation of soil contaminated with PFAS compounds derived from biowastes.

PFAS Sources	PFAS input	Remediation techniques	Observation	References
Biosolids	PFAAs 0 ~ 434 ng g^{-1}	Phytoremediation	Preferential uptake of PFCAs over PFSAs; Accumulation of shorter chain PFAAs over longer chain PFAAs; Accumulation in the plant tissues.	(Blaine et al., 2013)
Biosolids	PFAAs 0 ~ 434 ng g^{-1}	Phytoremediation	Fruit crops accumulate fewer long-chain PFAAs than shoot or root crops.	(Blaine et al., 2014)
Reclaimed water from raw sewage	PFAAs	Phytoremediation	Chain-length-dependency trends were evident in both lettuce shoot and strawberry fruit, with decreasing concentrations associated with increasing chain length.	(Blaine et al., 2014b)
Wastewater treatment plant sludge	PFCAs and 6:2 diPAP 0.1 ~ 83 ng g^{-1}	Biodegradation and phytoremediation	Biodegradation of 6:2 diPAP to its corresponding fluorotelomer intermediates and C4-C7 PFCAs, and substantial plant accumulation of endogenous PFCAs present in the biosolids and those produced from 6:2 diPAP degradation, especially for the short-chain PFCAs (C4-C6).	(Lee et al., 2014)
Biosolids	9 PFCAs and 3 PFSAs 41.4 ~ 220 ng g^{-1}	Phytoremediation	PFCA concentrations in grain increased logarithmically with increasing PFCA concentrations in soils ($P < 0.01$) while PFSAs in grain were correlated linearly with PFSA concentrations in soils ($P < 0.01$).	(Wen et al., 2014)
Biosolids	10 PFCs 2 ~ 483 ng g^{-1}	Chemical leaching	The leaching potential of PFCs decreases with increasing chain length.	(Sepulvado et al., 2011)
Biosolids	PFOS and PFOA 1.21 ~ 43.2 ng g^{-1}	Bioremediation	The concentration of PFOS accumulated in earthworms was higher than that of PFOA, and the accumulation may not be a process of partitioning equilibrium.	(Wen et al., 2015)
Biosolids	16 PFASs 17.5 ~ 120 ng g^{-1}	Bioremediation	PFAS bioaccumulation factor (BAF) values in earthworms correlated linearly with total PFAS concentrations in biosolids; An effective bioaccumulation of long chain perfluorinated carboxylic acids in earthworms.	(Navarro et al., 2016)
Biosolids	PFOA and PFOS	Phytoremediation	PFOA generally has higher concentrations than PFOS in the edible parts of crops; Leaf vegetables and root vegetables showed generally higher concentrations of PFOA and PFOS than fruit vegetables or fruits.	(Xiang et al., 2018)
Drying sludge and compost	20 PFASs	Bioremediation	A predominant accumulation of long-chain PFASs in the roots and shorter-chain ones in the aerial plant organs; Transfer and bioaccumulation factors were higher for PFASs than PBDEs in all crop plants and earthworms.	(Navarro et al., 2017)
Compost	PFOA, PFOS and FOSA	Phytoremediation	The highest bioconcentration factors of carrot for PFOA and PFOS were determined in the leaves, while lower values were calculated in the core and the peel; FOSA was totally degraded in the presence of carrot.	(Bizkarguenaga et al., 2016)
Sewage sludge	PFOA and PFOS	Phytoremediation	The highest transfer factors were found for the vegetative plant compartments with average values for PFOS below those for PFOA; Transfer of PFOA and PFOS into potato peelings exceeded the carryover to the peeled tubers.	(Lechner and Knapp, 2011)

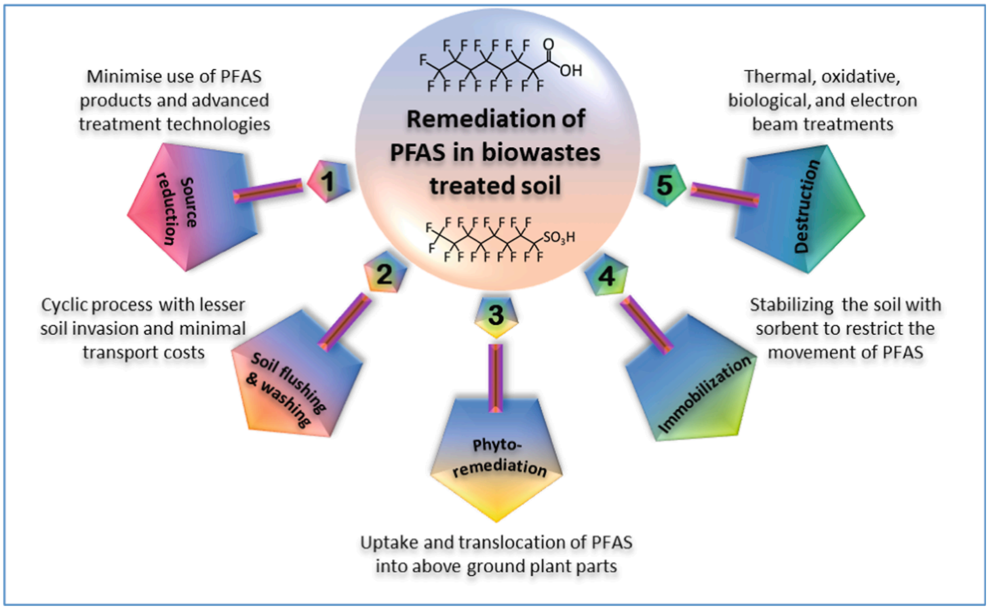

Fig. 4. Remediation options for PFAS in biowastes and soils treated with biowastes.

5.1. *Source control*

Efforts to control PFAS exposure must rely on reducing the use of PFAS-containing products, which range from household to industrial scale. However, the reduction in exposure is not straightforward and requires the combined efforts from a wide range of sectors, including policymakers and regulatory authorities. Soil and water serve as the natural sinks for the majority of the wastes and, hence, focusing on the sources of PFAS is a beneficial approach. Once PFAS enter soil or wastewater streams, biosolids, composts, or manures will not be free from PFAS contamination. Advanced technologies with high-end results, such as chemical oxidation and electron beams, could be applied to degrade PFAS in biowastes into harmless products, which would ensure their safe application to soil and, subsequently, they would have minimal effect on environmental and human health (Mahinroosta and Senevirathna, 2020).

One approach to minimise the risk from PAFS contamination in biowaste-treated soils could be to modify the biowaste in such a way that the PFAS are either decomposed or eliminated before soil application. This type of source control of PFAS still poses several challenges, because the PFAS need to be completely changed into harmless chemical species and removed. Pre- and post-treatment of commercially sourced biosolids, using technologies such as heat treatment, composting, blending, or thermal hydrolysis, barely had any effect on reducing the level of PFAA (Lazcano et al., 2019). A detailed analysis confirmed that the high-temperature heat treatment increased the PFAA load. Thermal hydrolysis did not affect the PFAA load, while blending of the biosolids with maple sawdust and aged bark tended to dilute the concentration of

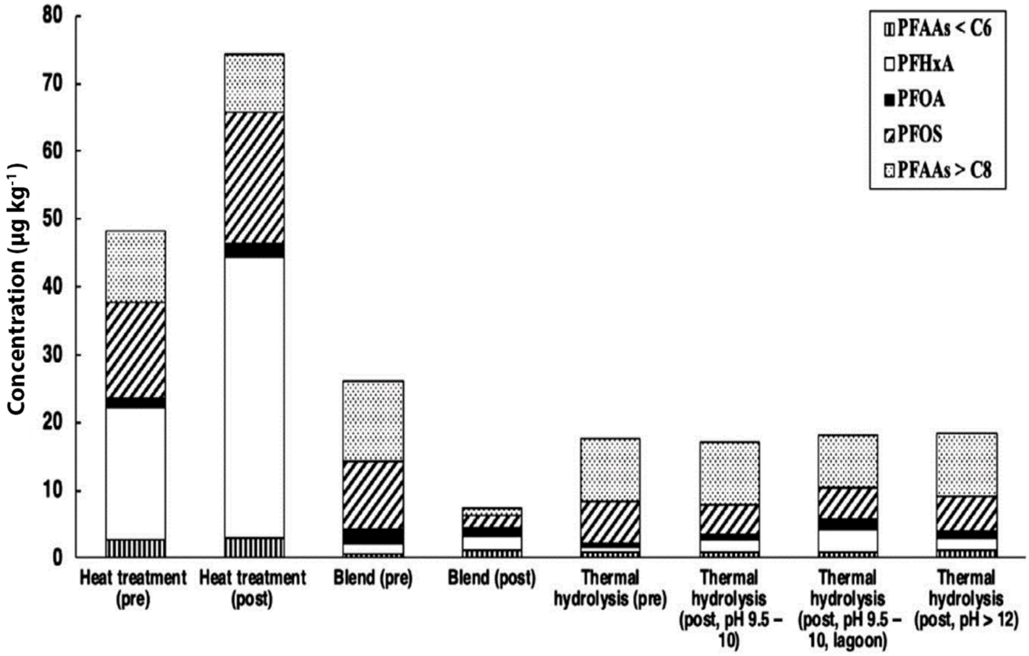

Fig. 5. PFAA loads (μg kg^{-1}, dw) for the < 2 mm particle size fraction of the samples. Pre: before post-treatment process (the Class A or B biosolids) and post: after post-treatment process. PFAAs < C6 include PFBA and PFBS, and PFAAs > C8 include PFNA, PFDA, PFUdA, PFDoA, PFTrDA, and PFTeDA. (Lazcano et al., 2019).

PFAA, which led to a reduced content in the biosolids (Fig. 5). The source reduction could serve as a preventive strategy to control the PFAS contamination at early stages which implies that there is relatively lesser probability of the commination. Since the source reduction is a significant aspect of lowering the PFAS contamination, it could be classified under the remediation measures.

Heating biowastes containing PFAS at high temperatures to alter the chemistry of the C-F bond could be a viable approach for addressing PFAS contamination. However, the evolution of potentially toxic volatiles at elevated temperatures could pose a more serious environmental concern than the PFAS themselves. One study highlighted that the addition of a liming agent, such as $Ca(OH)_2$, to the PFOS-spiked sludge sample caused the C-F bond to break, which resulted in the formation of either CaF_2 or $Ca_5(PO_4)_3F$, depending upon the conditions employed during the heating process (Wang et al., 2020) The formation of CaF_2 is favoured at a low temperature of 400 °C, while $Ca_5(PO_4)_3$ dominates at a high temperature of 900 °C, and the transformation ratio of fluorine to these two compounds is dependent on the Ca/F molar ratio used (Fig. 6). The various pathways for the mineralisation of fluorine into calcium salts during high-temperature treatment are shown in Figure SI 1.

Another point that needs to be considered is the cost. Because biowastes are cheap and readily available for use in huge quantities, their treatment before addition to the soil would incur a high cost, which will vary depending on the type of treatment used. Another concern is the addition of the PFAS-contaminated biowaste to soil. It presents an additional challenge, because it can be leached into groundwater and pose a more serious concern. More environmentally friendly approaches, such as the use of the renewable and biodegradable products for household and industrial purposes and advanced treatment technologies for PFAS remediation, are required to address this problem.

5.2. Soil flushing and washing

The in situ removal of PFAS is much cheaper and easier to implement as compared to ex-situ adsorptive removal using carbon-based materials or other types (Aly et al., 2019). Soil flushing is an in situ method of remediating pollutants, wherein an influx of fluid containing desired additives is forced through the soil to dissolve and separate the contaminants (Ramadan et al., 2018). The type of soil and the nature of the extracting fluid are critical for maximising the removal of the contaminants, and soils with coarse texture and loosely bonded contaminants present good conditions for effective soil flushing. As it is a cyclic process, the extracted fluid containing the contaminants needs to be purified and fed back into the system for continuous removal. Because the need for excavation and transport costs are eliminated, soil flushing is an attractive proposition. PFAS are a complicated class of organic contaminants, and soils contaminated with them usually encompass a large area or a large field, such as airports and defence sites. Hence, soil flushing is expected to be a good option, wherein the extracting fluid in the form of organic solvents, such as methanol, ethanol, or propanol, could be inserted into the ground at a specified place for the remediation of soil (Senevirathna et al., 2020). However, the complexity of the PFAS and their existence in several chemical forms pose challenges, because the same extracting fluid might not be suitable for all the range of PFAS. Moreover, the flushed/extracted PFASs need to be dealt with by using appropriate measures such as degradation or complete destruction to achieve their remediation and prevent any further contamination.

Soil washing is another cost-effective technique that could be utilized to remediate soils containing PFAS. However, in comparison with soil flushing, the bulk soil needs to be brought onto a specific site and treated with an extracting fluid in an ex-situ type of process. The removal efficiency of various PFAS depends upon their solubility in the extracting fluid, and the most common PFAS, such as PFOS, PFOA, and PFHxS, show good solubility in water. Hence, these can be easily removed from soil using water washing (Mahinroosta and Senevirathna, 2020). Once removed with water, the PFAS are treated with an aqueous phase for further treatment with adsorbents such as activated carbon. This latter operation is expected to have a significant impact on increasing the overall cost for the removal of PFAS using a soil washing process. It needs to be mentioned here that one of the crucial parameters that will determine the efficacy of soil flushing and washing is the removal rate of the PFAS, which is dependent upon the characteristics such as the amount and flow conditions of the fluid employed for extraction. Overall, soil flushing and soil washing are promising technologies for PFAS removal, with each having its pros and cons in terms of process complexity, type of soils treated, and the cost involved.

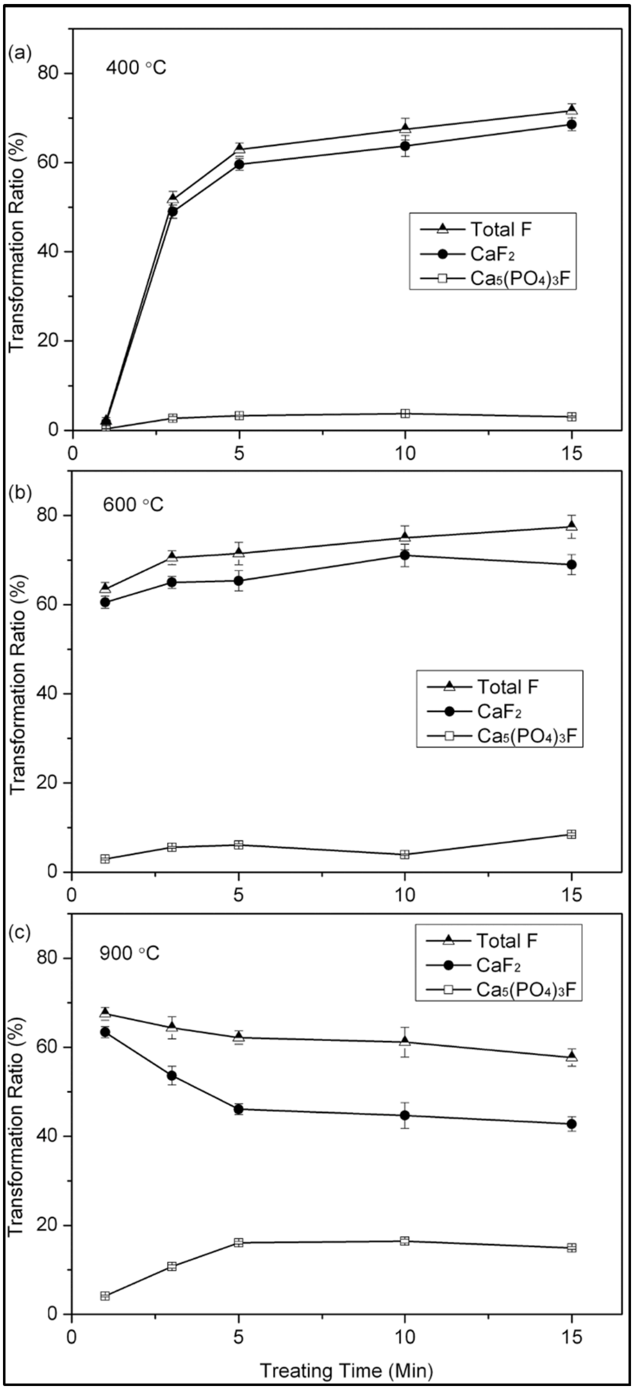

Fig. 6. Effect of thermal treatment time on the fluorine transformation efficiencies. The transformation ratio (TR) values for the sludge + PFOS + $Ca(OH)_2$ samples (Ca/F molar ratio of 2:1) were obtained by heating samples at 400 °C, 600 °C, and 900 °C with different retention times (Wang et al., 2013).

5.3. Phytoremediation

Uptake, accumulation, and metabolism of PFAS in plant species present a potential risk of exposure to humans causing harmful effects. Nevertheless, plant uptake of PFAS offers the benefit of using this mechanism as a tool to remediate PFAS-contaminated soils. Plants can readily accumulate short-chain PFAS in their leaves, whereas the roots adsorb more of the long-chain compounds (Jiao et al., 2020). Most of the literature focussing on plant uptake and accumulation of PFAS from biowaste-amended soils deals with food crops, which are not suitable to use in phytoremediation of PFAS. Plant species that have been used for remediating other persistent organic pollutants (e.g., petroleum hydrocarbons, PCBs, pesticides) and other emerging contaminants (e.g., pharmaceuticals) could be tried for phytoremediation of PFAS (Hoang et al., 2021, Thijs et al., 2016).

However, plant species that can remediate other organic compounds cannot perform in a similar manner with PFAS, because these compounds have very different behaviours in the soil–water-plant ecosystem (Blaine et al., 2014). PFAS are recalcitrant compounds, having unique chemical structures, and they are expected to be less degradable or nondegradable. Hence, extrapolation of previous results with other compounds is not appropriate in the study of phytoremediation of PFAS. Therefore, in-depth investigations are required concerning remediation of PFAS by plant species. Field studies with non-food crops are more relevant, to investigate the ability of plants to remediate PFAS present in bio-waste treated soils. Careful strategies need to be considered when disposing plant parts following phytoremediation of PFAS. Simple landfilling or unplanned disposal of the plant parts might pose a risk of returning the PFAS back into the environment. A recycling pathway of the PFAS-containing plant parts could be to convert them into biochar via high temperature pyrolysis, which is likely to eliminate PFAS through thermal degradation (Du et al., 2019, Zhang et al., 2020), and, at the same time, give a value-added product. Studies should also focus on the transformation or degradation of PFAS inside plants, about which no information is currently available. Most previous studies have investigated PFAS in plants grown near a specific point source, such as industrial manufacturing sites, fire training fields, landfills, or WWTPs. Field studies related to soil–plant relationships of PFAS in agricultural soils with biosolids are scarce, and the topic warrants future research (Wang et al., 2020).

5.4. Immobilization

Immobilization techniques help to reduce leaching of PFAS from soils into groundwater. A wide range of sorbent materials, such as activated carbon (powder or granular form), resins, minerals, biomaterials including biochar, polymers, amine functionalised materials, and modified clays, has been developed and tested for PFAS removal (Bolan et al., 2021, Mahinroosta and Senevirathna, 2020). Immobilization has been tested under many conditions, including laboratory-scale and field-scale trials, and the technique has been tried under both ex-situ and in situ conditions. The method has the potential of being a significant solution for PFAS contamination (Darlington et al., 2018). Still, it is not a cost-effective remediation technique, because a large quantity of absorbents may be required. For example, Das et al. (2013) used adsorbents added at a rate of 10% (w/w) to immobilize PFOS, and they can cause a significant increase in the cost of remediation. In addition to the high cost, the immobilization technique affects soil characteristics such as texture, thus limiting the final land use. In soils with added clay-based sorbents, the permeability and shear strength were changed (Das et al., 2013). A study carried out at an airport firefighting training ground in Norway showed a reduction of PFOS of 94–99% using montmorillonite as an amendment (Aly et al., 2019). Sörengård et al. (2020) observed a change in the compressive strength of soil when it was mixed with powdered activated carbon (PAC) and RemBind® for immobilization of contaminants. Immobilization often cannot be the best option because contaminants will not be degraded or destroyed completely, but rather bind or immobilize them. The sorbed contaminants will be left in place; however, they can be desorbed over time which makes the treated soil a potential source of toxicity (Xiao et al., 2019). This desorption could cause leaching of the contaminants to subsurface soils and groundwater (Sarsby, 2000). Results from the study of Silvani et al. (2019) demonstrate the application of biochar as a sustainable and environmentally-friendly sorbent for PFAS and metals in contaminated soils. In their study, PFAS-contaminated soils with varying total organic matter contents (1.6 and 34.2%) were amended with six doses of biochar, and the biochar reduced the concentration of PFOS over 90%. Aside from this study, the immobilization technique has been used only at a few military sites or airports or industrial sites. Laboratory-scale experiments have been conducted with contaminated soils collected from airports or military sites or industrial sites. The potential for the immobilization technique to remove PFAS contamination originating from biosolids application is currently not well understood, especially for the level of PFAS contamination found in biosolids and how much it contributes to current background levels. Hence, investigations are required using soils amended with PFAS-containing biosolids to evaluate the efficacy of the immobilization technique for PFAS removal (Mahinroosta and Senevirathna, 2020).

5.5. Destruction

Destruction is the most desirable remediation option for contaminant removal. Destruction techniques are thermal treatment, chemical oxidation, biological remediation, electron-beam treatment, and ball milling. However, destruction is not easy in the case of PFAS remediation due to a strong C–F bond and the high melting point of PFAS. So far, destruction techniques have been applied only at the laboratory scale. Field-scale application of these techniques has not yet been successful (Mahinroosta and Senevirathna, 2020).

Studies conducted so far have reported that the high melting point of PFAS makes thermal-treatment methods difficult. There are few reports about this treatment process, which suggests that thermal treatment is not feasible and sustainable due to high energy consumption and harmful effects of heat on soil (Mahinroosta and Senevirathna, 2020).

Chemical oxidation is one of the options to degrade PFAS, and it can be either ex-situ or in situ. The commonly used oxidants for in situ chemical oxidation are ozone, hydrogen peroxide, potassium/sodium permanganate, and sodium persulfate. Dombrowski et al. (2018) compiled details about the chemical-oxidation technique for PFAS removal and reported that heat-activated persulfate efficiently degraded PFCA compounds. Persulfate treatment removed PFOA when injected repeatedly with a low dosage, but the efficiency of PFOS removal was less. The chemical oxidation method needs to have a thorough investigation of the balance of free radicals, reaction kinetics, and radical scavengers.

Bioremediation using microbes and plants is theoretically a promising destructive technique for remediation of PFAS (Shahsavari et al., 2021). In practice, however, it is not feasible for PFAS removal due to the non-degradable nature of PFAS (Cousins et al., 2020). Microbial degradation of PFAS has not been studied extensively, and the limited literature on the topic provides conflicting outcomes (Kucharzyk et al., 2017). Conversely, abiotic methods such as electron-beam treatment and ball milling are able to degrade PFAS. For example, Turner et al. (2020) conducted mechanochemical ball milling experiments on soils from a Canadian firefighting training area, which indicated that PFOS concentrations can be lowered by up to 96% in the contaminated soils. Electron beam (eBeam) technology utilizes electron accelerators to generate extremely large numbers of highly energetic electrons from electricity. Recently, Pillai (2020) evaluated eBeam technology to treat PFAS contaminated soils and noticed that 2000 kGy dose reduced PFOS and PFOA concentration in Wurtsmith Air Force Base soils by $>$ 99.9%

and 53.7%, respectively. These novel techniques can be used to achieve a complete defluorination of PFAS in biowastes and biowaste-treated soils.

The literature survey concerning different remediation techniques for PFAS removal showed that there are no well-developed remediation methods for PFAS removal, especially for biowaste-treated soils. In addition, most of the literature documenting destruction techniques targets only PFAS contamination in aqueous systems, and less attention has been paid to the removal of PFAS in soil ecosystems (Cui et al., 2020, Tenorio et al., 2020). Hence more studies are needed concerning the enhanced degradation of PFAS in contaminated soil. PFAS removal from biosolids-amended soil requires special attention, because application of biosolids is inadvertently increasing the load of PFAS in soil.

6. Summary and conclusions

Solid biowastes, including biosolids, animal and poultry manures, and composts, are major sources of PFAS input into the environment. Plant uptake of PFAS through the application of these biowastes is an important pathway of animal and human exposure to PFAS. Various approaches have been used to remediate PFAS in biowastes and soil treated with these biowastes. Source control to prevent PFAS reaching the environment through biowaste application can be achieved by removing PFAS during the wastewater treatment process and avoiding PFAS containing organic wastes during the composting process. Removal of PFAS in biowastes might be achieved by thermal and chemical oxidation techniques, but they are difficult to carry out. Mobilizing amendments including surfactants and desorbing agents, can be applied to increase the bioavailability and mobility of PFAS in biowastes and in soil treated with these biowastes. The mobilized PFAS can subsequently be removed through phytoremediation or soil washing. Immobilizing amendments, such as sorbent materials, can be used to decrease the bioavailability and mobility of PFAS, thereby minimising their uptake by plants and leaching to groundwater. One of the limitations of the mobilizing technique is susceptibility to leaching of the mobilized PFAS in the absence of active plant uptake of PFAS. Similarly, in the case of the immobilization technique, the long-term stability of the immobilized PFAS needs to be monitored.

7. Future research priorities

Given the present understanding on the distribution of PFAS in solid biowastes and contamination of soil resulting from the application of these biowastes, we propose the following future research priorities:

- Identification of PFAS sources reaching biowastes used for soil application, and development of guidelines to reduce these sources reaching the biowastes (i.e., source control).
- Greater understanding on the distribution of various PFAS in different components of solid biowastes used for soil application.
- Examination of transformation of PFAS during treatment and conversion of solid biowastes to other products, such as conversion of biosolids to biochar.
- More in situ field studies to demonstrate the effect of a wide range of mobilizing agents in removing PFAS and of immobilizing agents in reducing the mobility and bioavailability of PFAS from biowastes and soil treated with these biowastes.
- Development of methods to demonstrate the effectiveness of mobilization techniques on the subsequent uptake of PFAS by plants and biota from soil receiving biowastes.
- Development of analytical techniques to examine the in situ long-term stability and effectiveness of immobilization of PFAS in biowastes and soil receiving these biowastes.
- In situ field studies to monitor phytotoxicity and eco-receptor end-points to demonstrate the environmental risk reduction derived from the application of amendments to manage PFAS in biowastes and soils treated with these biowastes.
- Development of regulatory acceptance guidelines to reduce PFAS input into soil from biowaste application.

Declaration of Competing Interest

None.

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Appendix A. Supplementary material

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