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Remediation of poly- and perfluoroalkyl substances (PFAS) contaminated soils — To mobilize or to immobilize or to degrade?

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Abstract

Poly- and perfluoroalkyl substances (PFASs) are synthetic chemicals, which are introduced to the environment through anthropogenic activities. Aqueous film forming foam used in firefighting, wastewater effluent, landfill leachate, and biosolids are major sources of PFAS input to soil and groundwater. Remediation of PFAS contaminated solid and aqueous media is challenging, which is attributed to the chemical and thermal stability of PFAS and the complexity of PFAS mixtures. In this review, remediation of PFAS contaminated soils through manipulation of their bioavailability and destruction is presented. While the mobilizing amendments (e.g., surfactants) enhance the mobility and bioavailability of PFAS, the immobilizing amendments (e.g., activated carbon) decrease their bioavailability and mobility. Mobilizing amendments can be applied to facilitate the removal of PFAS though soil washing, phytoremediation, and complete destruction through thermal and chemical redox reactions. Immobilizing amendments are likely to reduce the transfer of PFAS to food chain through plant and biota (e.g., earthworm) uptake, and leaching to potable water sources. Future studies should focus on quantifying the potential leaching of the mobilized PFAS in the absence of removal by plant and biota uptake or soil washing, and regular monitoring of the long-term stability of the immobilized PFAS.

Keywords

PFAS; Aqueous firefighting foam; Soil remediation; Biosolids; Mobilization and immobilization

1. Introduction

The substances of both organic and inorganic origin containing at least one fluorine (F) atom are generally termed as fluorinated substances or fluorochemicals or fluorinated chemicals. Among them, a specific sub-group of chemicals are known as perfluoroalkyl and poly-fluoroalkyl substances (PFASs) that contain a perfluoroalkyl moiety in their structures (Banks et al., 1994; Buck et al., 2011). The perfluoroalkyl moiety, generally represented by C_nF_{2n+1}, is a one or more carbon (C) atom-containing fluorinated aliphatic chain in which most of its hydrogen (H) atoms are substituted by F atoms (Banks et al., 1994). To be classified as a PFAS, the substance should have at least one perfluoroalkyl moiety in its chemical structure. The key groups of PFASs include perfluorinated carboxylic acids (PFCAs), perfluorinated sulfonic acids (PFSAs), and perfluorinated phosphonic acids (PFPAs), whereas polyfluorinated compounds include fluorotelomer alcohols (FTOHs), fluorotelomer sulfonic acids (FTSs), polyfluorinated alkyl phosphates (PAPs), perfluorooctane sulfonamine (PFOSA), and their derivatives (Buck et al., 2011). It is estimated that >4000 synthetic fluorinated compounds belong to the complex PFAS family.

The perfluorinated or polyfluorinated compounds are anthropogenic in nature and released into the environment due to human and industrial activities (Buck et al., 2011). Owing to their strong C—F bond strengths, these compounds are remarkably resistant to external environmental conditions such as temperature, water or oil status of the medium, and microbial attack. As a result, PFASs have found widespread applications in various industries including plastic manufacturing, textiles and leather industries, surfactants preparation, and even in medical applications (Prevedouros et al., 2006). Daily-life applications of PFASs extend to food wrapping materials, drink can-lining materials, non-sticky cookware, water-resistant fabrics and clothing, grease/oil resistant papers and surfaces, and firefighting foam (Darrow et al., 2013; Mahinroosta and Senevirathna, 2020; Yeung and Mabury, 2016).

Due to their widespread use in industries and daily-life products, PFAS have entered the soil and water environments, and now they are found in microorganisms, plants, higher animals, and humans globally, including in the Arctic and Antarctic ecosystems (Ahrens et al., 2016; Dreyer et al., 2009; Sunderland et al., 2019). Due to their unique chemical structures and stability, several PFASs have been proven to be bioaccumulative and toxic in higher animals including humans (Ahrens et al., 2019; Lau et al., 2007; Xiao et al., 2017). Among various usages, aqueous film forming foam (AFFF) is recognised as one of the key sources of PFAS entry into the soil and water environments. Perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), and perfluorohexane sulfonate (PFHxS) are the most commonly found PFASs in AFFF. The Class B fluorine-based AFFF is used to extinguish flammable liquid-caused fires, and firefighting training sites at and around fire brigades, airports, and defence establishments have been reported to contaminate the surrounding soil, water bodies, and groundwater with PFAS (Cousins et al., 2019). In addition to such point sources of PFAS, diffused pollution of soil and groundwater has taken place through transportation of these contaminants from household products and activities via biosolids, wastewater treatment plants, and landfill leachates (Sinclair and Kannan, 2006; Bolan, 2019).

Because of the high chemical, thermal, and biological stability of PFAS compounds, and also due to their existence as mixtures in environmental matrices, their remediation both in water and soil/sediment is extremely challenging. Approaches taken to remediate PFAS in solid and liquid media can be different. For example, they can be removed from a solid medium (e.g., soil or biosolid) following mobilization using selective chemical species, or their concentration can be reduced by plant uptake and by chemical or microbial destruction (Jeon et al., 2011; Simon et al., 2019). In the case of liquid media, PFAS compounds are removed mostly by using adsorbents or chemical destruction (Merino et al., 2016). PFAS immobilization in solid media has also been reported by adsorbents or fixing agents (Darlington et al., 2018). In terms of biotic degradation of PFAS compounds in soil and water, available information is limited (Wei et al., 2019).

A few review articles have concentrated on strategies for removing PFAS compounds from water, including sorption on various adsorbents (Du et al., 2014; Zhang et al., 2019; Vo et al., 2020), focussing on field testing of selected adsorbents (Espana et al., 2015), and advanced defluorination and degradation (thermal and non-thermal) methods (Ahmed et al., 2020; Vo et al., 2020), but review articles critically analysing soil PFAS remediation are scarce in the literature. Only two reviews have been written on this topic in the recent past

(Ross et al., 2018; Mahinroosta and Senevirathna, 2020). However, more information is needed concerning the scientific and technological soundness of PFAS remediation approaches, especially in soils. So, the current review paper aims to present information on the remediation of PFAS contaminated soils by critically reflecting on the pros and cons of contaminant mobilization, immobilization, and destruction strategies using a wide range of soil amendments. The mobilizing amendments help to desorb PFAS compounds that are bound to soil minerals or organic matter, and hence increase their bioavailability and mobility (Milinovic et al., 2015, 2016; Pan et al., 2009; Tang et al., 2017). In contrast, the immobilizing amendments adsorb or fix the PFAS compounds in soils reducing their bioavailability and mobility (Aly et al., 2019; Das et al., 2013; Hale et al., 2017). This review discusses all the above strategies by first presenting an overview of various soil PFAS sources, PFAS interactions with soil components and bioavailability, soil PFAS remediation through manipulating the bioavailability using conventional and advanced soil amendments along with some exemplary case studies, and highlighting their respective techno-economic advantages and disadvantages.

2. Sources of PFAS in soil

Soil and water environments receive PFAS contaminants mainly through discharge of AFFF, effluent discharge from wastewater treatment plants and landfills, and contaminated wastes such as biosolids (Table 1; Fig. 1).

2.1. Firefighting foams

As an important point source, PFAS from AFFF can be introduced into the terrestrial and aquatic environments during storage, handling, use, and post-use cleaning stages of these chemicals (Cousins et al., 2019). For example, a small PFAS volume can be released in the form of a concentrated foam at the time of storage, careless handling during transfer of containers, and usage and calibration of equipment. However, a more occasional release than the above occurs when a large volume of PFAS enters into the environment during real firefighting operations (Houtz and Sedlak, 2012, 2013). Additionally, downward leakage and/or lateral overflow from temporary ponds storing AFFF-contaminated water following fire training operations can also become an important diffused source of contamination in the surrounding areas (Eschauzier et al., 2013; Houtz and Sedlak, 2012; 2013).

Soil and groundwater contamination resulting from the use of firefighting foams at defence sites, airports, and fire brigade training sites has been noticed in many countries including Australia and the USA. For example, currently around 90 sites in Australia are being investigated for PFAS contamination from the regular use of firefighting foam (Australian Defense, 2019). Similarly, around 26,000 PFAS contaminated sites exist across the USA, impacting more than six million people through drinking water contamination (Darlington et al., 2018). Both in Australia and USA, the impacted areas are located around and near defence facilities where AFFF is used either for real fire extinguishing or training purposes. There are sources of PFAS other than AFFF that contaminate the environment, but given the great extent of PFAS contamination (i.e., high PFAS concentration and toxic chemical constituents) in and around defence sites globally, these sites warrant immediate risk assessment and remediation actions.

2.2. Wastewater effluents and sludges

Municipal waste disposal sites, landfills, wastewater treatment facilities, and biosolids provide major diffused sources of PFAS contamination of soil and water. For example, household wastewater containing PFAS chemicals in low concentration can reach municipal wastewater treatment plants and finally accumulate in biosolids (Bossi et al., 2008; Campo et al., 2014; Chen et al., 2012a; Gallen et al., 2018; Guo et al., 2010; SI Fig. 1). A number of PFAS compounds including PFOA and PFOS were found in Australian biosolids in recent years (Gallen et al., 2016, 2017; 2018). Higgins et al. (2005) reported total PFAS concentrations ranging from 55 to 3370 ng/g in domestic sludge in the USA. Venkatesan and Halden (2013) measured PFAS concentration in 113 biosolid samples collected from 94 waste water treatment plants in USA, and obtained a mean concentration of PFOS = 403 ± 127 ng/g, PFOA = 34 ± 22 ng/g, and PFDA = 26 ± 20 ng/g. Similarly, Sun et al. (2010) reported total PFAS concentrations in digested sewage sludge in Switzerland ranging from 28 to 637 ng/g, while total PFOS concentrations ranged from 15 to 600 ng/g.Kallenborn et al. (2004) demonstrated that Nordic countries had relatively low PFAS concentrations (0.6–15.2 ng/g) in sludges. Levels of PFAS contamination in Swedish sludge ranged from 0.6–23.9 ng/g and 1.6–54.8 ng/g for PFOA and PFOS, respectively (Haglund and Olofsson, 2009). Other estimates suggested that annually around 2749–3450 kg of total PFAS was present in biosolids across the USA, and around 1375–2070 kg PFAS ended up in agricultural land through soil applications (Venkatesan and Halden, 2013). Sepulvado et al. (2011) found that PFASs were found to concentrate to the 120 cm soil depth and reached a concentration up to 483 ng/g in field soils that had received long term biosolid applications. Washington et al. (2010) investigated sludge-applied soils in proximity to a wastewater treatment plant handling sewage waste from PFAS industries. The PFOS and PFOA concentrations in the sludge applied soil reached a maximum of 408 μg/kg and 312 μg/kg, respectively. Sludge from this wastewater treatment plant was found to contain PFOA concentrations up to 1875 ng/g.

PFAS can enter the sewage system through a variety of industrial sources that include PFAS manufacturing, fluoropolymer manufacturing, and AFFF manufacturing (Prevedouros et al., 2006). Many studies reported the degree of PFAS pollution in wastewater sludge (Table 1). The PFAS issue arises in sewage sludge because conventional wastewater and sewage treatment methods cannot efficiently eliminate these recalcitrant compounds from the system. The increase in concentration of some PFAS compounds such as perfluoroalkyl acid (PFAA) in sewage effluents over that in the influent is attributed to the degradation of more complex PFAA precursors during activated sludge treatment (Houtz and Sedlak, 2012; 2013). For example, wastewater treatment plants could show 9–352 % increase in PFOA concentration in effluents over influents (Schultz et al., 2006). However, PFOS often could exhibit a decrease in concentration in the effluent, attributed to high K_d values causing retention of PFOS in the sludge and lowering final PFOS concentrations in effluents (Yu et al., 2009). Becker et al. (2008) observed a 20-fold increase in PFOA concentrations from influents to effluents, and an additional 10 and 50 % PFOA and PFOS, respectively, adsorbed in the sludges.

2.3. Landfill leachate

Household wastes composed of goods containing hydrophobic and stain-resistant coatings (e.g., carpets) can release PFAS chemicals when dumped into the landfill (Gallen et al., 2016; Wei et al., 2019; Yan et al., 2015; Lang et al., 2017). As a result, PFAS can be released into the groundwater through contaminated leachates from landfills, or laterally move to surrounding land areas if appropriate lining is not in place (SI Fig. 2). In addition to sewage and wastewater, industrial wastes such as fabrics, building and coating materials can be a diffuse source of PFAS and their related chemicals (Janousek et al., 2019) when they are disposed in landfill sites. For example, long-chain PFAAs (e.g., PFOA and its precursors) were detected in landfill sites where the above types of wastes were dumped off for disposal. Knutsen et al. (2019) warned that short-chain PFAS compounds released from a range of household and industrial wastes could dominate over long-chain compounds in the leachates of historic landfill sites. The removal of such short-chain PFAS contaminants from water can be extremely challenging, which, in addition to the general challenges of landfill leachate treatment methods, underscores the PFAS contamination issue worldwide in ground and surface water resources from landfill sites.

3. Dynamics of PFAS in soils

PFAS compounds that reach soil through various sources (Table 2) can undergo sorption, partition, and complexation reactions that enable them to be retained in the soil (Zhang et al., 2019). Sorption refers to electrostatic interaction of PFAS with charged clay and organic matter surfaces, whereas partition refers to hydrophobic interaction of PFAS with organic substrates such as soil organic matter. Complexation involves partition of PFAS with dissolved organic matter forming soluble PFAS-organic matter complexes (; Zhu and Kannan, 2019). In contrast, plant uptake, leaching, degradation/transformation, and volatilization potentially can remove these chemicals from contaminated soils (Fig. 2). The dynamics of PFAS compounds and their fate in the soil depend on the characteristics of PFAS compounds (e.g., solubility and chain length), soil properties (e.g., organic matter and pH), and environmental factors (e.g., precipitation) (Milinovic et al., 2015). PFAS compounds can be adsorbed on soil particles through hydrophobic interaction and/or electrostatic attraction (Johnson et al., 2007; Wei et al., 2019). The major PFAS compounds such as PFOA and PFOS tend to exist as dissociated anions of acids under natural soil environmental conditions. PFAS compounds in the soil system can show both hydrophilic and hydrophobic characteristics. While the long-fluorinated alkyl chain of PFAS compounds confers on them hydrophobic properties, the sulfonate and carboxylate functional groups provide them hydrophilic characteristics (Darlington et al., 2018; Ross et al., 2018). Vapor state mobility of PFAS compounds may rarely occur under soil environmental conditions, because of the low to very low vapor pressures of most of the PFAS compounds (Kucharzyk et al., 2017), or in other words these compounds have high molecular weight and are highly stable in nature.

3.1. Sorption/desorption process

This section covers both sorption as indicated by electrostatic interaction of PFAS with charged clay and organic matter surfaces, and partition as indicated by hydrophobic interaction of PFAS with organic substrates such as soil organic matter. Because of simultaneously having hydrophobic fluoroalkyl long chains and hydrophilic ionizable functional groups, PFAS compounds show complex behaviours in the environment in terms of their sorption and desorption processes (Ahrens, 2011; Kannan, 2011). The sorption of PFAS in soils has been shown to increase with an increase in the chain length of PFAS compounds and also with an increase in the fraction of organic components (f_{oc}) in the soil (Brusseau, 2018; Milinovic et al., 2015). Simultaneously, PFAS sorption is also influenced by the soil pH and soil solution ionic strength. It has been shown that PFAS sorption in soil increases with an increasing electrolyte concentration (i.e., ionic strength) and/or due to the presence of higher valent cations in the soil solution (Wang and Shih, 2011). A decreasing pH of the soil (i.e., dominance of protons (H^+) on the soil surface) also increases the sorption of PFAS compounds, which is attributed mainly to an increase in positive charge with decreasing pH (Du et al., 2014; Jeon et al., 2011; Bolan et al., 1999). Therefore, two key mechanisms can be identified for PFAS sorption in the soil environment: (1) hydrophobic interaction with soil particles rich in aromatic hydrophobic components, and (2) surface electrostatic interaction with charged soil minerals (Fig. 3; Hellsing et al., 2016).

The sorption of PFAS compounds in soils and sediments has been studied extensively. In general, long chain PFAS compounds are adsorbed in soils and sediments dominantly via hydrophobic attraction, and short chain compounds via polar-polar interaction (e.g., electrostatic attraction) (Zhao et al., 2012). Soils and sediments containing high contents of organic carbon (OC) or black carbon particles thus tend to show high sorption of PFAS compounds. The physicochemical behaviours of PFAS compounds, especially the net hydrophobicity evolving from their chemical structures are critical to predict the sorption strengths of these chemicals to soils and sediments. For example, Milinvic et al. (2015) reported that among three studied PFAS compounds, namely PFOS, PFOA and perfluorobutane sulfonic acid (PFBS), PFOS was the most strongly adsorbed by six different soils. The authors attributed the strong interaction of PFOSs with soil particles to hydrophobic interaction, as indicated by a strong correlation between the log K_{ow} values of the three PFAS compounds and the log K_{oc} values of the soils (Fig. 4). Chen et al. (2013a, 2013b) demonstrated that PFOS and PFOA sorption (K_d value) increased with an increase of the organic carbon fraction and ionic strength of five different soils, whereas the sorption decreased with an increasing humic acid (HA) concentration in the solution. The authors suggested that HA or other dissolved organic matter might form complexes with PFAS compounds in the soil solution and inhibit sorption of those chemicals on to soil components such as clay minerals and particulate organic matter.

Hellsing et al. (2016) found that a negatively charged silica surface was not able to adsorb anionic PFAS compounds such as perfluorohexanoic acid (PFHxA), PFOA, PFOS, and perfluorononanoic acid (PFNA). On the contrary, positively charged alumina surface adsorbed significant amounts of these compounds, indicating that an electrostatic mechanism might come into partial effect for adsorbing PFAS compounds on electrically charged soil components (Higgins and Luthy, 2006). Johnson et al. (2007) suggested that PFOS sorption mechanisms to the surfaces of minerals such as kaolinite, goethite, high iron sand, and Ottawa sand could be dominantly controlled by electrostatic attraction when surfaces of these minerals were OC free. The presence of organic carbon of the mineral surfaces drives the sorption mechanism toward hydrophobic interaction. According to their charge characteristics, the above minerals adsorbed PFOS in the order: goethite > kaolinite > high iron sand > Ottawa sand. Tang et al. (2010) observed that pH, ionic strength, and Ca^{2+} concentration of solutions significantly influenced the sorption of PFOS by goethite, but their effects were only marginal when sorption occurred on silica. In the case of goethite, low pH values and high Ca^{2+} concentrations enhanced PFOS sorption via possible electrostatic attraction. Likewise, Ferrey et al. (2012) suggested that at around solution pH = 7, iron oxides adsorbed PFOS and PFOA predominantly through electrostatic attraction rather than hydrophobic interaction.

Literature suggests that PFAS sorption and desorption studies are mostly concentrated on soils and sediments, where point source pollution occurred mainly surrounding AFFF handling and storage facilities. However, due to the high mobility of these compounds, reports now exist that agricultural soils globally are also contaminated or vulnerable for contamination by these toxic compounds (Yao et al., 2015). The sorption and desorption behaviour of PFAS in agricultural soils can be different than that of other soils, because agricultural soils receive a continuous supply of amendments, fertilizers, and irrigation water. Therefore, understanding the retention of PFAS in agricultural soils requires future research attention, and the fate and behaviour of these contaminants should be studied using advanced biogeochemical prediction models.

3.2. Leaching

Most PFAS compounds are relatively more soluble in water than other persistent organic pollutants such as PAHs (Post et al., 2017). Hence, PFASs are liable for leaching, especially in soils with low sorption capacity (e.g., sandy soils). The sorption of the PFAS compounds in soils influences their leaching behaviour through the soil profile (Gellrich et al., 2012). Gellrich et al. (2012) observed that in groundwater, PFASs with short chain lengths (< 7 fluorinated carbon atoms) predominate in concentrations. The short chain PFASs can be less toxic than long chain ones (e.g., PFOS and PFOA); short chain PFASs display a higher mobility but lower bioaccumulation potential in the environment (Das et al., 2008; Newssted et al., 2008; Gellrich et al., 2012). However, despite the restricted use of PFAS, the concentrations of PFOA and PFOS in water and other environmental matrices are likely to increase because of (1) the continuous desorption of PFOS and PFOA that are still bound to soil particles, and (2) the slow transformation of precursors of these compounds in environmental compartments (Frömel and Knepper, 2010). Gellrich et al. (2012) detected PFOA in the leachate percolating through a soil column until about four months after the experiment began, but PFOS was found below the detection limit (1 ng/L) even after 2.5 years. Similarly, Stahl et al. (2013) reported that PFOA moved rapidly with water through a soil lysimeter, while PFOS travelled slowly (SI Fig. 3). Thus, the leaching behaviour of PFAS is in part influenced by the extent to which the PFAS is adsorbed onto soil and sediments during transport. The sorption of PFAS onto soil and sediments during transport facilitates partial removal of PFAS from aqueous media, which potentially retards PFAS flow velocity relative to the velocity of water, thereby attenuating the concentration of PFASs over distance and time across the stream (Higgins and Luthy, 2006; Li et al., 2019; Ross et al., 2018).

3.3. Plant uptake

Unlike nonpolar contaminants (e.g., petroleum hydrocarbons) that tend to accumulate on plant root surfaces via lipid partitioning, polar contaminants including PFAS may enter into the transpiration stream of plants and move across the whole system (Ahrens et al., 2009; Blaine et al., 2013; Dalahmeh et al., 2018; Garcia-Valcarcel et al., 2014). For those contaminants that enter the transpiration stream, the ratio of concentrations of the solute in the transpiration stream to the soil solution is defined as the transpiration stream concentration factor (TSCF) (Felizeter et al., 2012).

TSCF = [Concentration of solute in the transpiration Stream/Concentration of solute in the soil solution]

Plant uptake of PFAS compounds is dependent on PFAS chain length and the sorption behaviour in soils. Being relatively hydrophilic, the short chain PFAS are expected to have high TSCF values. Since ionisable contaminants such as PFAS are soluble and non-volatile, a high concentration of PFAS can potentially accumulate in plants (Ghisi et al., 2019). PFAS accumulation occurs predominately in the leaves, because water along with PFAS enters into the roots and then translocates to the leaves before getting evaporated, resulting in the accumulation of PFAS in the leaves (Ghisi et al., 2019). The bioaccumulation factor (BAF) of PFAS can be calculated as follows (Liu et al., 2017):

BAF = [PFAS concentration in the plant (μg/kg plant)/PFAS concentration in the soil (μg/kg soil)]

Only a few studies have documented the bioaccumulation potential of PFAS, particularly PFOA and PFOS, into food crops (SI Fig. 4). In an artificially contaminated soil, Stahl et al. (2009) observed an uptake of PFOS and PFOA in maize, wheat, potato, and oats, and particularly high concentrations accumulated in the vegetative portions of the plants. Lechner and Knapp (2011) also reported similar PFOA and PFOS uptake patterns in the vegetative portions of carrot, cucumber, and potato. Blaine et al. (2013) found that the dry weight concentrations of PFBA and PFPeA in plants grown in a PFAS-contaminated biosolid-amended soil under glasshouse conditions reached 266 and 236 μg/kg in lettuce, and 56 and 211 μg/kg in tomato, respectively. PFBA showed the highest BAF (56.8) in the case of lettuce, while the highest BAF for PFPeA was 17.1 in the case of tomato. Under field conditions, while no PFBA and PFPeA were detected in corn grains, concentrations of the chemicals in the corn stover were ultralow (Blaine et al., 2013). In another study, Blaine et al. (2014) found that among various PFAS compounds present in a contaminated biosolid-amended soil, crops such as radish, celery, and pea accumulated the highest concentrations of PFOA (67 μg/kg), PFBA (232 μg/kg), and PFBA (150 μg/kg), respectively. However, an increasing chain length of PFAS compounds significantly decreased the shoot-soil concentration factor (SCF) for all the crops. Under greenhouse conditions, Blaine et al.

(2013) also monitored the entry of PFAS into the human food chain via irrigating food crops (lettuce and strawberry) with reclaimed water (PFAS concentration = 0.2–40 μg/L). PFBA and PFPeA (short-chain PFAS) showed the overall highest accumulation of any PFAS in the edible parts of both the crops. The authors also reported that the OC content of soils had an inverse relationship with the bioaccumulation of PFAS. Zhu and Kannan (2019) conducted a field study within a one-mile radius of a five-decade old fluoropolymer (PFCA) industry and observed that, while the soil samples and plant tissues contained mainly PFOA, the earthworms accounted for higher proportions of long-chain PFCAs (e.g., PFUnDA and PFDoDA). The biota-soil accumulation factor in earthworms and root-soil accumulation factor in plants/grasses increased with an increasing chain length of the PFCAs. Thus, the bioaccumulation of PFAS in crops from soils would depend on PFAS concentration, soil properties, crop species, and the specific PFAS analyte, and it is not clearly understood. Also, reports concerning PFAS bioaccumulation in beneficial soil animals such as earthworms are scant (Karnjanapiboonwong et al., 2018; Zhao et al., 2013; Zhu and Kannan, 2019), and, given the importance of these animals in ecosystem functions, it warrants future studies.

3.4. PFAS transformation

The major transformation processes of PFAS include abiotic and biotic degradation with a limited extent of volatilization. Transformation of various precursor substances can provide an indirect source of PFAS input to soils (Ruan et al., 2015). For example, fluorotelomer alcohols (FTOHs; $F(CF_2)_nCH_2CH_2OH$) are some of the major indirect sources of PFAS input in soil (Dinglasan et al., 2004; Yuan et al., 2016). Studies showed that FTOHs might undergo degradation and produce secondary chemicals such as polyfluoroalkyl carboxylic acids (PFCAs), fluorotelomer aldehydes, and secondary polyfluorinated alcohols (Liu et al., 2007, 2010; Zhao et al., 2017). For example, an aerobic biotransformation of 6:2 FTOH ($F(CF_2)_6CH_2CH_2OH$) was reported to produce PFCAs including PFBA, PFPeA, and PFHxA, *x*:3 acids, such as 5:3 acid ($F-(CF_2)_5CH_2CH_2COOH$), and 4:3 acids, such as ($F-(CF_2)_4CH_2CH_2COOH$) (Zhao et al., 2013a). The profiles of FTOH degradation products varied depending on the bacterial strains involved. For example, mixed bacterial strains yielded equal amounts of PFCAs and x:3 acids, and a small amount of other transient intermediates (Liu et al., 2010; Zhao et al., 2013a, b). In contrast, a single strain of *Pseudomonas* sp. transformed 6:2 FTOH yielding a high quantity of transient intermediates and low quantities of PFCAs and x:3 acids (Kim et al., 2012). Similarly, Tseng et al. (2014) indicated that a white-rot fungus (*Phanerochaete chrysosporium*) degraded 6:2 FTOH yielding mainly 5:3 acids.

FTOHs biodegradation rates and pathways differed among aerobic/anoxic/anaerobic conditions, with the first-order rate constants in the following decreasing order: aerobic > anoxic > anaerobic conditions (Yu et al., 2016). The anaerobic biodegradation of FTOH was found inefficient to produce PFCAs, but might form polyfluorinated acids (Zhang et al., 2013b). FTOHs could also be transformed by terrestrial plants and animals. For example, Zhao and Zhu (2017) observed that 10:2 FTOH was biotransformed to PFDA, PFNA, and PFOA by soil microorganisms, PFDA, PFHxA and PFPeA by wheat roots, and PFDA and PFNA by earthworms (*Eisenia foetida*). In the atmosphere, peroxy radical reactions could degrade FTOHs producing a series of homologous PFCAs (Ellis et al., 2004), which also contributes to widespread contamination of PFCAs in soil. Other fluorotelomer derivatives, such as fluorotelomer sulfonate, polyfluoroalkyl phosphate, fluorotelomer acrylate, and fluorotelomer stearate monoester, could act as precursors of FTOHs, which could subsequently be subjected to the same degradation pathways as mentioned above to form PFCAs (Lee et al., 2010; Russell et al., 2008; Butt et al., 2014; Wang et al., 2011; Lewis et al., 2016; Zhang et al., 2016; Liu and Liu, 2016; Dasu et al., 2012).

The potential PFAS precursors from electrochemical fluorination (ECF) include mixtures of linear and branched isomers of perfluorooctane sulfonamide (FOSA), sulfonamido ethanol (FOSE), FOSE-based phosphate diester (SAmPAP diester), and perfluoroalkyl sulfonamide derivatives (e.g., *N*-methylperfluorooctane sulfonamidoethanol (MeFOSE), *N*-ethylperfluorooctane sulfonamidoethanol (EtFOSE), *N*-ethylperfluorooctane sulfonamide (EtFOSA), and *N*-methylperfluorooctane sulfonamidethylacrylate (MeFOSEA)) (Ruan et al., 2015). A few studies confirmed that biotransformation of sulfonamide derivatives was an indirect source of PFOS in soils. For example, Benskin et al. (2013) reported that SAmPAP diester was persistent in marine sediments with an approximate half-life of >380 days at 25 °C, whereas EtFOSE was transformed by bacteria to a number of products, including *N*-ethyl perfluorooctanesulfonamido acetic acid (EtFOSAA), perfluorooctane sulfonamide acetate (FOSAA), EtFOSA, FOSA, and PFOS (Benskin et al., 2013). Rhoads et al. (2008) proposed a transformation route of EtFOSE as follows: EtFOSE→EtFOSAA→EtFOSA→FOSA→perfluorooctane sulfinate (FOSI)→PFOS. The proposed biodegradation pathways of PFAS in the soil system largely follow those reported in activated sludge and sediments. For instance, Mejia-Avendaño and Liu (2015) investigated aerobic biotransformation of EtFOSE and EtFOSA in soil. PFOS was identified from the biotransformation products of EtFOSA (4.0 mol%) after 182 days of aerobic incubation, which demonstrated that EtFOSE and EtFOSA were precursors of PFOS in the soil environment. No further degradation of PFOS was reported in the soil. Similarly, Zhao et al. (2016b) found that EtFOSE was sequentially transformed to EtFOSAA, FOSAA, FOSA, and PFOS in an earthworm-soil system. Zhao et al. (2018) further reported that FOSA could be degraded to PFOS in soil-wheat and soil-plant-earthworm systems. Only a few studies to date have examined biotransformation of perfluoroalkyl sulfonamide derivatives in soil. The available studies have not fully identified intermediate products, suggesting that the proposed pathway of transformation of PFAS is still uncertain. Considering the role of soil as a sink for PFAS in the environment, future studies are necessary to examine the biotransformation of legacy and emerging PFASs in the soil environment.

4. Remediation of PFAS in soil

Remediating PFAS contaminated solid and aqueous media can be extremely challenging because of the following key reasons: (1) high chemical and thermal stability of PFAS compounds, (2) frequent occurrence of complex mixture of PFAS compounds in the contaminated environment, (3) unique physicochemical properties of PFAS compounds (i.e., both hydrophobic and oleophobic behaviours), and (4) extremely persistent nature (i.e., limited or no biodegradation). Although various methods have been reported to remove

PFAS compounds from aqueous media (Carter and Farrell, 2010; Ding and Peij-nenburg, 2013; Du et al., 2014; Wagner et al., 2013; Zhang et al., 2011), these methods may not be readily applicable for remediating PFAS contaminated soils or waste materials (e.g., biosolids) (Darlington et al., 2018). Two broad approaches, namely mobilization and immobilization, using soil amendments, as discussed below, may prove logistically and economically viable for the remediation of PFAS contaminated soil. While an immobilization approach can be used to reduce mobility and bioavailability of PFAS, the mobilization approach can be used to remove PFAS through soil washing and phytoremediation. Both these two approaches can facilitate the destruction of PFAS from soil through abiotic and biotic degradation processes.

4.1. Mobilization of PFAS compounds

The key mechanisms involved in the mobilization of contaminants in soils/sediments include solubilisation, desorption, and complexation reactions. Mobilization processes can be used to remove contaminants from soil through washing (i.e., soil flushing) and plant uptake (i.e., phytoremediation). In the case of organic contaminants such as PFAS, mobilization processes can be facilitated using various soil amendments (Table 3), which lead to the complete destruction of these contaminants through abiotic and biotic degradation reactions.

4.1.1. Soil flushing and soil washing—Soil flushing is an *in-situ* process that involves injection of a flushing solution into the ground for extracting contaminants (Hale et al., 2017; Yao et al., 2015). The main advantage of soil flushing is that large quantities of soil can be treated *in-situ* without the need for excavation and transport (Jawitz et al., 2000; Svab et al., 2009; Zheng et al., 2012). Surfactants having both hydrophobic and hydrophilic structural groups are used to facilitate the desorption of persistent organic pollutants (POPs) and subsequent soil flushing. However, many PFAS, including PFOS and PFOA, are themselves surfactants, which can make PFAS behaviour difficult to predict during PFAS mobilization. For example, Pan et al. (2009) found that a cationic surfactant (cetyltrimethylammonium bromide (CTAB)) was able to significantly enhance the sorption of PFOS to sediments due to the initial sorption of CTAB to sediments, thereby exposing CTAB’s hydrophobic tails to adsorb PFOS. However, an anionic surfactant (sodium dodecyl-benzene sulfonate (SDBS)) showed a concentration-dependent effect where a SDBS concentration <4.34 mg/l increased PFOS sorption to sediments, but SDBS concentration >21.7 mg/l increased PFOS desorption (Pan et al., 2009). Guelfo and Higgins (2013) found that an anionic surfactant (sodium dodecyl sulphate (SDS)) at low concentration decreased the sorption of PFOS, PFNA, and PFDA, but increased the sorption of long chain PFAS, such as PFBA, PFPeA, PFHxA, PFHpA, and PFBS. In general, anionic surfactants could enhance the solubility of PFAS in water, which would decrease PFAS sorption to soils/ sediments and, thereby, facilitate PFAS mobilization.

Other common soil flushing additives such as organic/inorganic acids/bases and solvents such as methanol or ethanol might be suitable for removing PFAS from soils. For example, Schröder (2003) applied organic solvents (e.g., ethyl acetate (EtOAc), dimethylformamide (DMF), pyridine, tert-butyl methyl ether (MTBE), 1,4-dioxane, or tetrahydrofuran (THF)) to determine the ability of a solvent or combination of solvents for sludge PFAS extraction.

Under pressurised solvent extraction at 150 °C and 143 bar, a sequential flushing with a mixture of EtOAc and DMF followed by methanol modified with phosphoric acid appeared to be the most effective extractants of sludge PFAS. Omitting DMF from the flushing mixture made the procedure greener without significantly reducing the PFAS extraction efficiency.

Advanced methods such as reverse osmosis (RO) and supercritical fluid (SCF) assisted extraction using comparatively non-toxic organic solvents were also attempted for removing PFAS from aqueous and solid matrices, respectively. For example, Tang et al. (2006) found that isopropyl alcohol increased the solubility of PFOS during reverse osmosis (RO), but decreased the membrane flux. Chen et al. (2012a) reported that at a critical point of CO_2 (50 °C, 20.3 MPa), HNO_3 (16 N) first suppressed the polarity of PFOS and PFOA molecules increasing their solubility in supercritical CO_2, and then methanol led to increased mobilization of PFOS and PFOA from a sand matrix with 59 and 77 % extraction efficiencies, respectively, and from paper and fabric with 80 and 100 % efficiencies, respectively.

The first stage of soil washing would concentrate PFAS of soils/sediments into a solution. Once the contaminant is flushed out and collected, the solution could be decontaminated using water treatment technologies such as sorption, ion-exchange, or filtration including RO technique for reuse or safe disposal. Special research in the future should be given to developing environmentally benign treatment methods, such as using natural and green adsorbents and supercritical CO_2 extraction.

4.1.2. Phytoremediation—

Unlike other POPs, PFAS are relatively soluble and remain in the soil solution, leading to their ready uptake by plants and subsequent removal using phytoremediation technology. The plant uptake of PFAS compounds is dependent on their chain length and the sorption behaviour of soils. Promising PFAS compounds suitable for phytoremediation include those with relatively low log K_{ow} and a small C chain. For example, the mass uptake of PFBA was calculated to be high at 11.27 mg/m^2/year, assuming log K_{ow} (0.001), concentration (0.1 mg/L), transpiration (500 L/m^2/year), and fractional water use (0.3) (Austin et al., 2017). The carbon chain length (6 or less) is possibly a more appropriate screening metric for phytoextraction than log K_{ow}, as the short chain PFASs show the highest TSCF values (Austin et al., 2017).

Huff et al. (2019) considered the bioconcentration factor (BCF) as a key metric in assessing the suitability of plant species for phytoremediation of contaminated sites. They demonstrated hyperaccumulation of multiple PFAS compounds; a greater than 10-fold soil to leaf translocation of PFAS in above-ground plant tissues was exhibited. The BCFs of PFOA hyperaccumulating species ranged from 11.5–46.5, and that of PFOS ranged from 10.3–17.9. They also showed higher plant uptake of PFOA and PFOS (44–344%) with the application of a proprietary soil amendment than without the amendment. Similarly, Gobelius et al. (2017) reported successful uptake of 26 PFAS compounds in plants from contaminated soils around a firefighting training site in Stockholm.

Phytoremediation of a PFAS contaminated site can be a slow process, but it involves low capital cost and almost no maintenance cost. Thus, phytoremediation remains the most economic and sustainable green technology available for the remediation of PFAS contaminated sites.

4.2. Immobilization of PFAS compounds

The immobilization technique redistributes PFAS contaminants from the solution to solid phase, thereby reducing their mobility and bioavailability. A range of amendments have been tested to enhance the immobilization of PFAS contaminants in soils and sediments (Table 4).

4.2.1. Sorption—The materials used for sorbing PFAS in soil and water mainly include carbon-based and clay-based materials, ionic surfactants, and anionexchange resins (Ahrens and Bundschuh, 2014; Yu et al., 2009; Senevirathna et al., 2010). Carbon-based materials include activated carbon (AC), multi-walled carbon nanotubes (CNTs) and biochars (Darlington et al., 2018; Liu et al., 2019). Of these, CNT and AC were reported to show remarkable PFAS sorption capacity (Ochoa-Herrera and Sierra-Alvarez, 2008; Sarkar et al., 2018; Wagner et al., 2013; Xiao et al., 2017). The non-polar functional groups of carbon-based materials enable them to be highly useful for hydrophobic PFAS sorption.

Powdered or granulated AC (GAC) was effective in removing PFOA and PFOS in soil and water (Cummings et al., 2015). A small number of commercial adsorbents (e.g., RemBind™, PefluorAd) available on the market could be used as a complement or as an alternative of AC (Birk, 2016). Clay mineral candidates, such as montmorillonite (Zhou et al., 2010; Zhang et al., 2014), kaolinite (Zhang et al., 2014; Zhao et al., 2014), hematite (Zhao et al., 2014), alumina (Wang and Shih, 2011), and boehmite (Wang et al., 2012), were used for batch-scale removal of PFAS compounds, specially focussing on PFOS. For example, Hale et al. (2017) tested AC, compost soil, and montmorillonite to immobilise PFAS in contaminated soils. PFAS leaching was reduced by 94–99.9, 29–34, and 28–40 % for AC, compost, and montmorillonite amendments, respectively. Natural clay minerals have a hydrophilic surface, which is intrinsically negatively charged, rendering them ineffective for hydrophobic and anionic PFAS removal. However, when modified with a surfactant, its hydrophobic alkyl chain enhances PFAS sorption through hydrophobic partitioning. Additionally, in contrast to natural clay minerals, quaternary ammonium surfactant-modified products generate positive surface charge (Sarkar et al., 2011, 2012), which could attract anionic PFOS and PFOA via electrostatic interaction. Das et al. (2013) reported the immobilization and reduced leaching of PFOS (>90 %) from four AFFF-contaminated soils using a palygorskite-based organoclay prepared with oleylamine. Like organoclays, a swellable organically modified silica was shown to outperform GAC for PFAS sorption (Fig. 5), including short-chain PFAA compounds (Stebel et al., 2019). However, care should be taken in choosing the organic agents for modifying mineral materials, because chemicals such as oleylamine or hexadecyltrimethyl ammonium (HDTMA) can be extremely toxic to native soil micro- and macro-organisms (Sarkar et al., 2010, 2013).

Some liquid-based amendments were also used to facilitate PFAS immobilisation in contaminated solid media. For example, Aly et al. (2019) observed a six-fold increase of

PFAS retention in contaminated soils by applying a commercially available coagulant. Similarly, Pan et al. (2009) calculated the thermodynamic index of irreversibility (TII) for PFOS to sediments in the presence of a cationic surfactant, cetyltrimethylammonium bromide (CTAB), to quantify the degree of sorption irreversibility caused by CTAB, with a value of 0 representing a highly reversible system and 1 representing irreversible sorption. A value of 1 was approached for CTAB concentrations of 18.1 and 36.1 mg/L, indicating its potential use for PFOS immobilization. The cationic surfactant could be delivered to the source of pollution using *in situ* percolation or injection.

Many factors including media characteristics, PFAS characteristics, and adsorbent characteristics would influence the overall effectiveness of PFAS remediation via sorption treatments (Darlington et al., 2018). The pH of the solid media (e.g., soil, biosolid), as well as their concentrations of inorganic and organic ions, significantly control the sorption efficiency. PFAS sorption decreases with increasing pH of the medium. While natural organic matter in the soil does not reduce the PFAS sorption capacity of activated carbon and clays, high ionic strength (i.e., high concentration of inorganic salts) adversely affects the sorption capacity of organoclays through the adsorbents' charge reversal behaviour (Das et al., 2013; Jeon et al., 2011; Sarkar et al., 2012). The presence of sulfonate functional groups can lead to strong sorption of PFAS. Physicochemical characteristics of adsorbents (e.g., SSA, pore size distribution, cation exchange capacity, zeta potential) also have a remarkable effect on PFAS sorption (Du et al., 2014; Jeon et al., 2011). Adsorbents with small pore size and high SSA can lead to high PFAS sorption capacity. Similarly, adsorbents with a basic or positively charged surface tend to show high PFAS sorption capacity through the combined mechanism of hydrophobic interaction and electrostatic attraction (Lu et al., 2016).

4.2.2. Stabilization and solidification—Stabilization and solidification (S/S) of contaminants including PFAS can be achieved by applying cementitious binders and additives into the contaminated soil, sediment, and waste media (Bates et al., 2000; Fagerlund et al., 2019; Sörengård et al., 2019a,b). The stabilizing/solidifying agents immobilize contaminants via physical and/or chemical protection. In the case of physical protection, contaminant leaching is prevented by reducing the hydraulic conductivity of the system. In the case of chemical protection, contaminants are stabilized by reducing their aqueous solubility through precipitation, redox alteration, and sorption reactions. The S/S technique undertaken *in situ* or *ex situ* can prove efficient in terms of treatment performance and costs when contamination over a large area requires remediation (Fagerlund et al., 2019; Sörengård et al., 2019a,b). A limited number of reports are available on PFAS site remediation using the S/S method. Sörengård et al. (2019a) examined seven additives, namely pulverised activated carbon (PAC), Rembind®, powdered zeolite, chitosan, hydrotalcite, bentonite, and $CaCl_2$ at 2% application rate, for stabilizing a total of 14 PFAS compounds in an aged-contaminated soil. The PAC and Rembind® additives performed the best with, respectively, 70 and 94 % reduction of leaching achieved for all the concerned PFAS compounds except perfluorobutane sulfonate (PFBA). Highly persistent and bioaccumulative long-chained PFAS (e.g., PFOS) was stabilized by 99.9 % by PAC or Rembind® application. The chain length and functional groups present in the PFAS compounds influenced the additives' PFAS stabilization efficiency. The stabilization capacity increased by 11–15 % per CF_3-moeity, and the stabilization of perfluorosulfonates (PFSAs) was 49 % higher than perfluorocarboxylates (PFCAs). During the soil PFAS stabilization process, PAC and Rembind® did not show any significant impact on the physical matrix stability (Orengarda et al., 2019).

4.3. Destruction of PFAS compounds

A complete remediation of PFAS contaminated soils can be achieved by complete destruction of PFAS compounds through biotic (e.g., biodegradation) and/or abiotic (e.g., thermal oxidation, chemo oxidation, ball milling) degradation processes, as discussed below.

4.3.1. Bioremediation—Biodegradation via microorganisms can be an effective and efficient method to remediate soils and groundwater contaminated with many organic pollutants (Fahid et al., 2020; Wang et al., 2019). An aerobic environment promotes the production of perfluorinated compounds (PFCs) from relevant precursors through microbial transformation (Dasu and Lee, 2016; Liu and Mejia-Avendaño, 2013). However, PFOS and PFOA are reported to be strongly resistant to microbial transformation under aerobic environments, and only a few reports are available on this topic (Liu and Mejia-Avendaño, 2013; Pasquini et al., 2013; Ochoa-Herrera et al., 2016; Liu et al., 2010; Chetverikov et al., 2017).

Biodegradation of PFAS in soil depends on the nature of microbial composition. For instance, Pasquini et al. (2013) discovered that *E. coli* was not able to biodegrade PFOS and PFOA. In contrast, *P. plecoglossicida* (Chetverikov et al., 2017), *P. parafulva* (Yi et al., 2016), *Acidimicrobium sp.* (Huang and Jaffé, 2019) and *P. aeruginosa* (Kwon et al., 2014) led to a significant reduction of the PFOS concentration. Beskoski et al. (2018) reported that chemoorganoheterotrophic bacteria, as well as yeast and molds, could reduce PFOA and PFOS moderately. A few other studies investigated the degradation potential of various PFCs using microbial cultures of sludges, industrial site sediments, contaminated and uncontaminated soils, and wastewater treatment plants; however, the exact bacterial composition was often not clarified (e.g., Ochoa-Herrera et al., 2016; Liu et al., 2010; Mejia-Avendaño et al., 2016). Apart from the bacterial composition, environmental factors that incude pH and soil solution composition are important for the optimal degradation rate of PFAS compounds.

Several studies reported that the degradation of PFCs is limited to the non-fluorinated moiety; therefore, defluorination by biodegradation seems not possible (Mejia-Avendaño et al., 2016; Dimitrov et al., 2004). Defluorination is an important process to turn PFC molecules largely harmless. Various studies detected the release of fluorine ions (Beskoski et al., 2018; Chetverikov et al., 2017; Liu et al., 2010; Ochoa-Herrera et al., 2016). Ochoa-Herrera et al. (2016) reported a 3% release of the total fluorine ions of the PFOS concentration with an aerobic microbial treatment. They speculated that the release of fluorine ions was due to the degradation of fluorinated impurities caused by the low purity of PFOS (96 %). Another study found a low release of fluorine ions during the degradation of PFOA (96 % purity) with anaerobic microorganisms (Liu et al., 2010).

To date, the defluorination of PFCs by microbial biodegradation has not been reliably verified or falsified, but this does not imply an entire biodegradation of PFCs. Mejia-Avendaño et al. (2016) examined the aerobic biotransformation of perfluorooctane sulfonamide quaternary ammonium salt (PFOSAmS) and perfluorooctaneamido quaternary ammonium salt (PFOAAmS), whereby the PFOAAmS concentration was reduced to 43 % of the initial amount while PFOA increased. A parallel run without microorganisms, which showed no change in PFOAAmS or PFOA concentration, indicated that these results were related to biodegradation. In the experiment with microorganisms PFOAAmS decreased and PFOA increased and without microorganisms the PFOAAmS and PFOA concentration remained the same. Mejia-Avendaño (2016) explained this phenomenon with the biodegradation of PFOAAmS to PFOA. This study demonstrated that biodegradation was possible, although this only involved the breakdown of the non-fluorinated moiety. However, to our knowledge, no field or *in situ* experiments have been conducted on PFC biodegradation. Therefore, further research should focus on *in situ* implementation of PFC biodegradation, as previous studies were only conducted at the laboratory level. Furthermore, in order to understand fully the whole process, all degradation pathways and their environmental impact should be investigated in the future.

4.3.2. Chemical and thermal treatment—In general, PFOS and PFOA destruction using common water oxidative/disinfection methods (e.g., chloramination, chlorination, ozonation, chemical oxidation and ultraviolet treatment) was found ineffective in most cases (Higgins and Dickenson, 2016; USEPA, 2016a,b). However, the removal of PFAS compounds from soil, waste, and water sources can be achieved through chemical and thermal redox reactions. For example, laboratory scale destruction of PFAS was achieved through catalytic and electrocatalytic oxidation using anodic mixed metal oxides (e.g., Ti/RuO_2) (Lin et al., 2012). However, the presence of naturally occurring DOM might significantly restrict the degradation rate of PFAS in soil via chemical oxidation (Buxton et al., 1988). The low reduction potential of fluorine ($E < -2.7$ V) as such makes the defluorination reaction thermodynamically unfavorable. However, sub-critical reduction of certain elements (e.g., Fe) at high temperature and pressure could lead to PFOS oxidation at least in laboratory and bench scales, but it is not so feasible for *in situ* application.

In-situ chemical oxidation (ISCO) using a peroxydisulfate process has been previously used for the remediation of pollutants such as chlorinated ethenes and benzenes, oxygenates, benzene, toluene, ethylbenzene, xylenes (BTEXs), and PAHs from soil (Nadim et al., 2006; Tsitonaki et al., 2010). Formation of the activated persulfate radicals can be accomplished through UV exposure, heat, high pH (alkaline conditions), hydrogen peroxide, and a variety of transition metals (Watts and Teel, 2006). Thus, persulfate first can be delivered to the contaminated soil subsurface in an inactive form, and then activated when it comes in contact with the contaminated zone. Activation by heat can be accomplished using steam injection or thermal energy production using electrodes (Heine and Steckler, 1999).

Hori et al. (2008) found that persulfate oxidation, activated by hot water, was effective at degrading PFOA to below the detection limit after 6 h of treatment at 80 °C. Lee et al. (2012) were able to achieve a complete persulphate-induced degradation of PFOA at pH = 2.5 after 72 h at 40 °C, and 215 h at 30 °C. Hawley et al. (2012) examined the activation of persulfate oxidation with the goal of soil and groundwater treatment. Activators that were able to degrade PFOS by more than 97.5 % included the following: Fenton's reagent, peroxide- activated persulfate, and heat-activated persulfate. Strong reducing agents such as sodium dithionite and sodium hypophosphate were also tested, but only partial degradation of PFOS was observed. They attempted a 'Smart Combination *In-situ* Oxidation/Reduction (SCISOR)' technique involving a combination of redox agents and activators, which achieved 60 % PFOS removal after one contact phase. However, Place and Field (2012) expressed concern regarding the use of *in-situ* chemical oxidation (ISCO) for the remediation of AFFF-impacted sites, because advanced oxidation techniques have been known to facilitate PFAS and PFCA formation from the more complex precursors present in AFFFs. Pancras et al. (2013) demonstrated that the combination of SCISOR and soil washing techniques was able to achieve > 99 % removal of PFOS from contaminated soil. Similarly, electrochemical oxidation involving a specific anode-electrolyte combination was found effective in degrading PFAS at <200 mg/l concentrations in a bench-scale reactor (Niu et al., 2016).

Although chemical treatments involving chloramination, chlorination, ozonation and oxidation reactions have been found to be effective in the removal and destruction of PFAS compounds, it may not be applicable to large-scale remediaion of PFAS contaminated soils under field conditions. It involves expensive chemicals and results in residual chemicals which may cause environmental degradation and issues with safe disposal (Higgins and Dickenson, 2016).

Thermal treatments of PFAS contaminated soil include both complete degradation of PFAS requiring high temperature (900–1100 °C) (Watanabe et al., 2016), and thermal desorption (Lim et al., 2016). The complete thermal degradation is covered under *'Vitrification or incineration'* (Section 4.3.3). The thermal desorption technique involves *ex situ* or *in situ* heating of PFAS contaminated soils, and the subsequent removal of vaporised PFAS compounds through air filtration. For example, Sörengård et al. (2020) have been able to achieve 71–99 % thermal desorption of PFAS in a field contaminated soil at 550 °C, and >99 % desorption in a PFAS fortified soil.

4.3.3. Vitrification or incineration—Temperatures required for vitrification of PFAS compounds range between 1600 °C–2000 °C. An advantage of this process in the context of PFAS is the lack of by-products generated, because all organic contaminants are fully degraded or destroyed. For example, Yamada et al. (2005) tested whether fluorotelomer-treated textiles and paper, after being destroyed under municipal incinerator conditions, would form PFOA as a degradation product. There was no PFOA detected in the samples after incineration. Even if PFOA was formed during incineration, it must have also been destroyed in the process, meaning degradation of precursors during incineration would not be a significant PFOA source in the environment. There are several processes available to reach vitrification temperatures: electrical, thermal, and plasma. The electrical process is *in situ* and involves construction of a zone surrounded by graphite electrodes inserted in the ground which pass energy through the soil. The thermal process is *ex situ* and is generally carried out in a rotary kiln. Plasma processes are only necessary when temperatures of up to

5000 °C are required. Electrical or thermal processes could be used to target the combustion of PFAS on site without using more extreme temperatures.

4.3.4. Ball milling—

Mechanochemical destruction (MCD), or high energy ball milling, has recently gained attraction in the soil remediation sector due to its ability to destroy effectively POPs with no requirement for toxic solvents, extreme temperatures, harmful additives, or high pressures (Cagnetta et al., 2016). The mechanisms involved in mechanochemical transformations tend to be complex and dissimilar to reactions observed in other remediation processes, i.e., thermal, photochemical, and chemical oxidation. While heat is generated under ball milling conditions, it was disregarded as the sole reaction initiation mechanism because of the progression of mechanochemical reactions even at very low temperatures (i.e., 77 K) (Beyer and Clausen-Schaumann, 2005). Rather, the formation of free radicals and matrix defects by strong mechanical activation (i.e., grinding) are the main reaction initiators (Sohma, 1989). These ‘mechanoradicals’ are capable of accelerated chemical transformation and destruction of POPs by high energy ball milling. Centrifugal and planetary ball mills are commonly utilised in laboratory-scale research (Li et al., 2017; Nomura et al., 2012; Zhang et al., 2013a).

Although POP degradation pathways by ball milling are complex and not fully understood, most published trials show high rates of destruction between 99–100 % for a wide range of POPs and organic contaminants subjected to mechanochemical conditions (Cagnetta et al., 2016). While the focus of MCD has historically been directed toward chlorinated POPs, several papers have recently been published related to PFAS degradation by high energy ball milling. Work by Zhang and colleagues (2013a) revealed degradation efficiencies of 100 % for PFOA at 180 min and 99.88 % for PFOS at 360 min. The destruction of PFOS and PFOA was carried out in a range of supporting matrices including CaO, SiO_2, Fe-Si mix, NaOH, and KOH. The most complete PFAS degradation was obtained with KOH as the support matrix, showing conversion of organic fluoride (C—F) to negatively charged monovalent fluoride (F^-). Zhang and colleagues (2016a) repeated the same trial with a Chinese PFOS alternative known as F-53B (6:2 chlorinated polyfluoroalkyl ether sulfonate), and they achieved a more rapid rate of degradation due in part to the substitution of a single fluorine with a chlorine on the terminal carbon and the introduction of an ether group. Further work by Lu et al. (2017) successfully destroyed 6:2 fluorotelomer sulfonate within 60 min, again using KOH as the support. Citing the concerns over the amount of KOH required for matrix support, Cagnetta et al. (2017) degraded a range of PFAS compounds and switched the support matrix to La_2O_3. While effective, La_2O_3 cannot be used for soil remediation due to its high cost, and low concentration of PFAS in soil would lead to insignificant amounts of lanthanum oxyfluoride formed.

As a non-thermal technology requiring only mechanical energy input, ball milling presents a compelling treatment option for PFAS-impacted soil and hazardous waste. Further research is required to determine its effectiveness as a *bona fide* remediation technology, especially related to PFAS degradation mechanisms, kinetic reaction progression, fluoride fate, and scaling factors.

5. Case studies of field application of remediation technologies

In this section, selected 5 selected case studies involving the field application of mobilization (soil washing and phytoremediation), immobilization (stabilization/solidification), and destruction (thermal oxidation and chemical oxidation) techniques of remediating soil contaminated with PFAS compounds are given. The reduction of the mobility and bioavailability, and ultimate removal, of PFAS compounds are discussed.

5.1. Case study 1: Mobilization and soil washing

In this method, water is used to extract PFAS from contaminated soil without any use of additional chemicals. The method is being applied in several projects using mobile equipment operated by a Swedish company, Svevia. Contaminated soil is excavated and loaded to a container where it is mixed with water. A high-pressure water stream is used to scrub the fines from coarser soil particles and dissolve some of the contaminants. Fines are then separated from coarser fractions for further management. The coarse (washed) particles are ejected and placed back to the pit after concentration of contaminants is checked through sampling and analysis. The method was tested on a pilot scale (10 tonnes) where 96 % separation of PFOS from soil particles was achieved (Swedish EPA, 2018).

Soil washing was then applied on a full scale to soil from a site in Kalmar, Sweden, where firefighting exercises have been carried out for 10 years. The aim was to wash the soil in order to separate PFAS from soil particles, place cleaned particles back to the site, and clean the washing solution. Because PFAS are readily water soluble, it was expected that most of PFAS from soil could be removed and that the remediation goals could be reached. Fifty tonnes of soil were washed per batch. In the first two batches the achieved PFOS concentration in washed soil was 17 μg/kg (remediation goal was set to 29 μg/kg). In the following two batches the remaining concentration in washed soil was above the remediation goal. The washing solution was first cleaned using ozonation, but it was not sufficient to reduce PFAS concentrations and active carbon filters were applied. Only 10 % (150 of 1500 tonnes) of the planned soil volume got treated. The main obstacle to proceed with the clean-up was a high fraction of clayey soil particles that hampered the equipment (Johansson, 2019).

5.2. Case study 2: Mobilization and phytoremediation

Gobelius et al. (2017) conducted a major study on the plant uptake of PFAS at a contaminated site of fire training facility at Stockholm Arlanda airport, to assess PFAS phytoremediation potential of a range of plant species under field conditions. The extent of contamination of soil and groundwater and the uptake of 26 PFASs by plants were evaluated. Based on the bioaccumulation factor and biomass production, they proposed three scenarios to estimate the PFAS phytoextraction efficiency and remediation period. The first scenario comprised a shelter wood of mixed silver birch (~66 %) and Norway spruce (~33 %) stands. Considering this system with frequent harvest of shoot and birch sap, and an understory of ground elder, they estimated that it was possible to remove annually 1.4 g of Σ26PFASs/ha. The second scenario was the regular coppicing of birches, in which the tree trunks were left in the field but the twigs and leaves were harvested in every 3 – 5 years of rotation. Annually 5 tonnes/ha of biomass composed of twigs and leaves could be generated, leading to annually 0.65 g of extractable Σ26PFASs for birches. The third scenario was the preservation of a meadow composed of plant species with high PFAS uptake potential. Accordingly, the long beech fern (*Phegopteris connectilis*) and ground elder (*Aegopodium podagraria*) are practical choices, extracting annually 0.55 g/ha of Σ26PFASs if mowed regularly.

Results of this field study suggested that, while plant uptake thresholds for plant species investigated in this study remained uncertain, the first scenario with a shelter wood appeared an ideal remediation option with the highest PFASs uptake. Taking into consideration the decreasing concentrations of PFOS over time and the threshold values for PFOS in soils (i.e., 0.003 μg/kg for sensitive land use and 0.02 μg/kg for non-sensitive land use), the time period required for remediating the soil at the contaminated site was estimated. It was estimated that the uptake of PFOS by spruces and birches would require 48,000 years and 160,000 years, respectively, to reach the threshold value for sensitive land use or 18,000 years and 58,000 years, respectively, for the non-sensitive land use. Assuming similar toxicity of the other 25 PFASs as for PFOS, the threshold values for Σ26PFASs would be 0.078 μg/kg for sensitive land use and 0.52 μg/kg for non-sensitive land use. Thus, the uptake of Σ26PFASs by birches and spruces would require <45 years for the remediation of the site due to the greater uptake potential, in particular for PFBA, PFNA, PFHxS, and 6:2 FTSA.

5.3. Case study 3: immobilization and stabilization/solidification

The method was tested in Sweden for stabilization of PFAS in a soil matrix using various binders, such as combination of Portland cement, fly ash, and ground granulated blast-furnace base slag (9:1 soil to binder ratio), as well as commercially available stabilizers such as pulverised activated carbon, Rembind®, powdered zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride (Sörengård et al., 2019a). The leaching of various PFAS compounds decreased on average by 70 % and was larger for longer carbon chains. For PFOS the decrease was 99.9 %. Six tonnes of PFAS contaminated soil (Σ11 PFAS = 160 μg/kg) from an industrial site were then treated on a pilot scale using 10 % Portland-fly ash cement with an addition of 2% granular activated carbon. Solidified soil monoliths (1.2 m^3) are being leached with water simulating 15 years precipitation. According to the preliminary results, the leaching of PFOS and PFOA decreased by 98 % (Kleja et al., 2020).

In a recent study, two soils affected by AFFF training activities were treated with a combination of no-treatment, soil plus granular activated carbon (GAC), or soil plus GAC and general purpose Portland cement and aged 30 days before being subjected to simulated acidic leaching by a Synthetic Precipitation Leaching Procedure (SPLP) (USEPA, 2019). A total of 24 analytes were monitored in a soil leachability testing, including perfluoroalkyl acids (PFAAs), such as perfluorinated carboxylates (PFCAs) and perfluorinated sulfonates (PFSAs), and precursors/intermediates such as perfluorooctyl sulfonamide (FOSA), N-methyl/ethyl perfluorooctyl sulfonamidoacetic acids (N-MeFOSAA, N-EtFOSAA), and fluorotelomer sulfonates (FTS) (4:2 FTS, 6:2 FTS and 8:2 FTS). For most of the PFAS analytes, addition of GAC to the soils dramatically decreased the leachability of the PFAS compounds present in the soils (stabilization), and further reduced the leachability of PFAS compounds with the addition of cement as a binder (solidification) for about one third of the PFAS present in these contaminated soils. Overall percent immobilization of PFAS that were detectable in the leachate from treated soils ranged from 87.1%–99.9%.

In another case study in 2015, RemBind Pty Ltd (https:// https://rembind.com//) investigated the potential value of stabilization techniques to remediate 1000 tonnes of PFAS contaminated soil originated from two airport sites. In this work, RemBind® material was applied to stabilize PFAS in soil, thereby reducing its leachability to allow for safe disposal of the contaminated soil to landfill with regulatory approval. The lab-scale tests demonstrated that 5% (w/w) RemBind® addition was effective in reducing the leachate PFAS concentrations to below the target criteria of <0.2 μg/L.

After treatment of the contaminated soil with RemBind®, the approval for safe disposal validation results indicated that the leachate PFAS concentrations reduced to the level of reporting (LOR; 0.01 μg/L). Based on these results, permission was obtained for the safe disposal of treated soil to a lined landfill without any requirements for remediation or management. For the safe disposal of the PFAS contaminated soil, a 100 cm layer of pure RemBind® was spread in the bottom as an additional level of risk mitigation (SI Fig. 5). The RemBind® amended soil was spread on a layer of the RemBind® bottom liner and covered with another 100 cm layer of pure RemBind®. This is one of the major large scale (1000 tonnes) PFAS soil disposal projects completed in Australia with EPA regulatory approval. This field-based study provided a proof of concept for the application of immobilization agents as a cost-effective remediation technology for the sustainable management of PFAS contaminated soil. Since this initial work, RemBind® has been used at full commercial scale to treat 1,000′s of tonnes of PFAS contaminated soil in Sweden (https://rembind.com/projects/remediation-of-pfas-impacted-soil-at-a-fire-station-in-sweden/) and, more recently, at the Townsville RAAF Base in Queensland, Australia (https://rembind.com/projects/full-scale-pfas-remediation/).

5.4. Case study 4: Destruction by thermal oxidation

Endpoint (2017) introduced a new technology named as Vapor Energy Generator (VEG) and applied it practically on a small scale for thermal treatment of PFAS contaminated soil. The instrument delivered thermal energy via recycled water and propane to produce steam at 1100 °C for PFAS treatment (700 °C for other chemicals). Endpoint Consulting Inc. (http://www.endpoint-inc.com/contact_us.htm) in collaboration with the Colorado School of Mines conducted a VEG trial on PFAS contaminated spiked soil to examine the treatment effectiveness at various temperature ranges. Operation at 950 °C for 30 min removed several groups of PFAS together with PFOS with 99 % efficiency (Endpoint, 2017). In response to the increasing PFAS concentration in the environment, Clean Earth (https://cleanearthinc.com/locations) has started and applied thermal desorption as a feasible method to eliminate PFAS from soils. This method has been applied by Clean Earth since 1992 to remove diverse groups of organic contaminants from the environment. Under the guidance of the New York State Department of Environmental Conservation (NYSDEC), 22.6 tonnes of contaminated soil at Clean Earth’s Fort Edward, New York facility were decontaminated in December 2018 and February 2019. Based on this opening trial of thermal desorption technology for treatment of PFAS-contaminated soil, Clean Earth is continuously working with state agencies to launch standards to establish the effectiveness of this technology and its optimization (https://cleanearthinc.com/what-we-handle/pfas-contaminated-soils). Enviropacific is an Australian (https://enviropacific.com.au/about-us/) owned industry and a leader in providing waste management services, which have wide-ranging thermal treatment abilities for PFAS-contaminated soil. Recently thermal treatment trials by Enviropacific showed an effective PFAS removal in 20 soil samples, with >99.9 % reduction in PFOS (from 172 µg/kg to 0.004 µg/kg) and PFOA (from 2.73 µg/kg to <0.0005 µg/kg) (https://www.enviropacific.com.au/wp-content/uploads/2016/09/Enviropacific_Treatment-of-PFAS.pdf) (SI Figure 6). In addition, Ventia (https://www.ventia.com/projects) has established a joint venture with Suez (https://www.suez.com.au/en-au) to construct an efficient soil decontamination facility in Dandenong South, Victoria, Australia to provide a clean, consistent, and economical PFAS treatment service. This service has been functional since November 2018 and is situated next to the only dumping site licensed to collect Category B waste (i.e., industrial wastes with medium levels of contamination) in Victoria (Ventia, 2018). According to recent report, Ventia has treated more than 525,000 tonnes of polluted soil (including PFAS-contaminated soil) using thermal treatment technologies. This figure of 525,000 tonnes represents more than 72 % of all soils that have been thermally treated in Australia.

5.5. Case study 5: Destruction by chemical oxidation

In Canada, a bench-scale treatability trial on the removal of PFOS in AFFF-impacted water and soil systems was directed, with an emphasis on the application of oxidation processes. Results of the bench-scale treatability trial revealed that 84–97 % PFAS removal occurred in 30 days from soil/tap water systems. The technology involved a cautious balance of free radicals, rate of reaction, and radical scavengers (Mahinroosta and Senevirathna, 2020). However, this is considered as a relatively non-economical technique, due to the high cost and greater quantities of chemical consumption (Australian Defense, 2019). Apart from Environment and Climate Change Canada (ECCC) and Emergencies Science and Technology Section (ESTS) of Canada reports, there are only a few reports available and only on the laboratory scale for removal of PFAS from contaminated water systems via chemical oxidation or advanced oxidation processes. Recently AECOM (https://www.aecom.com/au/) launched a new electrochemical oxidation technology DE-FLUORO™ for permanent removal of PFAS from the environment in Adelaide; this trial was based on removal of PFAS from contaminated water (https://www.aecom.com/press-releases/aecom-to-launch-pfas-solution-de-fluoro-at-cleanup-2019-in-adelaide/).

6. Summary and conclusions

This review examined mobilization, immobilization, and destruction techniques for the remediation of soil contaminated with PFAS from various sources. Soil amendments which are effective in the mobilization of PFAS compounds via desorption and complexation reactions can be applied to enhance the mobility and bioavailability, and subsequent removal through plant uptake and soil washing. Anionic surfactants can be effective in the mobilization of PFAS compounds, and their subsequent removal through soil washing. However, one of the environmental issues with the application of mobilization techniques is that the mobilized PFAS compounds are subject to leaching, particularly in the absence of plant uptake and soil washing, thereby leading to groundwater contamination. Mobilization techniques can be used for the complete removal of PFAS compounds through abiotic and biotic degradation. Although most of the PFAS compounds are recalcitrant to undergo biotic degradation, the potential value of abiotic degradation through thermal and chemical redox reactions for the removal of PFAS compounds has been demonstrated.

Stabilization and solidification remediation treatment has been found to be an effective immobilization technology aimed at reducing leaching and bioavailability of PFAS compounds. However, a major intrinsic issue liked to immobilization techniques is that, although the PFAS compounds become less mobile and bioavailable, their total mass in soils remains unaffected. The immobilized PFAS compounds may be solubilized and become bioavailable with time through breakdown of organic-PFAS compounds complexes. Complete removal of PFAS compounds from solid media, including soil and biosolid waste, can be achieved through biotic and abiotic degradation and decomposition. Biodegradation of PFAS is slow and not readily applicable under field conditions to treat large volumes of contaminated matrix. However, abiotic degradation involving thermal and chemical oxidation and ball milling can be applied under field conditions to treat large volumes of contaminated matrix.

Given the present understanding on the mobilization, immobilization, and destruction techniques with regard to managing the mobility and bioavailability of PFAS compounds, and subsequent remediation of contaminated soils, we propose the following future research priorities:

- Remediation technologies are primarily focused on PFOS and PFOA, which are the major PFAS compounds detected in environmental matrices; however, many other PFAS may be present in environmental matrices including soil, wastewater, and biosolids, and technologies that are able to treat PFOS and PFOA effectively may not be appropriate for other PFAS with different properties.
- More *in situ* field studies are necessary to validate the beneficial effect of a wide range of mobilizing and immobilizing agents in remediating soil contaminated with PFAS.
- Co-contaminants including hydrocarbons and chlorinated solvents may also be present in PFAS contaminated soil. Oxidative technologies including *in situ* chemical oxidation that are often applied to these co-contaminants may transform PFAS compounds into PFOS and PFOA. Hence field studies are necessary to evaluate the impact of soil amendments on the mobilization of associated co-contaminants.
- It is necessary to develop methods to demonstrate effectiveness of mobilization and immobilization techniques to manage the mobility and bioavailability of PFAS. For example, advanced spectroscopic methods can be applied to examine the *in-situ* long-term stability and effectiveness of immobilization.

- *In situ* field studies are also necessary to determine bioavailability, phytotoxicity, and ecoreceptor endpoints to demonstrate risk reduction derived from the application of soil amendments to manage PFAS.
- It is important to enhance regulatory acceptance of these methods through sound scientific advances and demonstrations.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Appendix A.: Supplementary data

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Abbreviations:

AC	activated carbon
AECOM	architecture, engineering, consulting, operations, and maintenance
AFFF	aqueous film-forming foam
BAF	bioaccumulation factors
BCF	bioconcentration factor
CFR	chlorinated flame retardants
CNT	carbon nanotubes
CTAB	Cetyltrimethylammonium bromide
dAQ	aquifer into a deeper section
DDT	dichlorodiphenyltrichloroethane

diPFCA	polyfluoroalkyl phosphate diesters
DOM	dissolved organic matter
dw	dry weight
EC50	effective Concentrations$_{50}$
ECCC	environment and climate change Canada
EDL	environmental decontamination limited
FCSAP	federal contaminated sites action plan
ESTS	emergencies science and technology section
FOSA	perfluorooctane sulfonamide
FTCA	fluorotelomer carboxylic acids
FTS	fluorotelomer sulfonates
FTuCA	unsaturated fluorotelomer carboxylic acids
GAC	granulated activated carbon
HCB	hexachlorobenzene
ISCO	*in-situ* chemical oxidation
Kd	soil specific distribution coefficient
Koc or Kom	partition coefficients with the specific organic particle or organic component of the soil
Kow	octanol-water partition coefficient
LC50	Lethal Concentration$_{50}$
LD50	Lethal Dose$_{50}$
LOQ	instrumental limit of quantitation
LOR	level of reporting
L-PFBS	perfluorobutane sulfonate
L-PFOA	non branched perfluorooctanoic acid
MCD	mechanochemical destruction
MEP	multiple extraction procedure
MTBE	methyl-tert-butyl ether
NAPL	nonaqueous phase liquid

NOEC	no observable effect concentration
OC	organic carbon
PAC	powdered activated carbon
PAH	polycyclic aromatic hydrocarbon
PBDE	polybrominated diphenyl ethers
PCB	polychlorinated biphenyls
PFAA	perfluoroalkyl acid
PFAS	perfluorinated alkyl substances
PFBA	perfluorobutanoic acid
PFBS	perfluorobutane sulfonic acid
PFC	perfluorinated compounds
PFCA	perfluorocarboxylic acids
PFDA	perfluorodecanoic acid
PFDDA	perfluorododecanoic acid
PFDoDA	perfluorododecanoic acid
PFHpA	perfluoroheptanoic acid
PFHxA	perfluorohexanoic acid
PFHxS	perfluorohexane sulfonate
PFNA	perfluorononanoic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonate
PFOSA	perfluorooctane sulphonamide
PFPeA	perfluoropentanoic acid
PFSA	perfluorosulfonic acids
PFTeDA	perfluorotetradecanoic acid
PFUnDA	perfluoroundecanoic acid
POP	persistent organic pollutants
RO	reverse osmosis
sAQ	aquifer into a shallow section

SCF	supercritical fluid
SCISOR	Smart combination *In-situ* oxidation/reduction
SDBS	sodium dodecylbenzene sulfonate
SOM	soil organic matter
SPLP	Synthetic precipitation leaching procedure
SSA	specific surface area
S/S	stabilization and solidification
THF	tetrahydrofuran
TII	Thermodynamic index of irreversibility
TSCF	transpiration stream concentration factor
WWTP	wastewater treatment plant
VEG	vapor energy generator
UCS	uniaxial compressive strength

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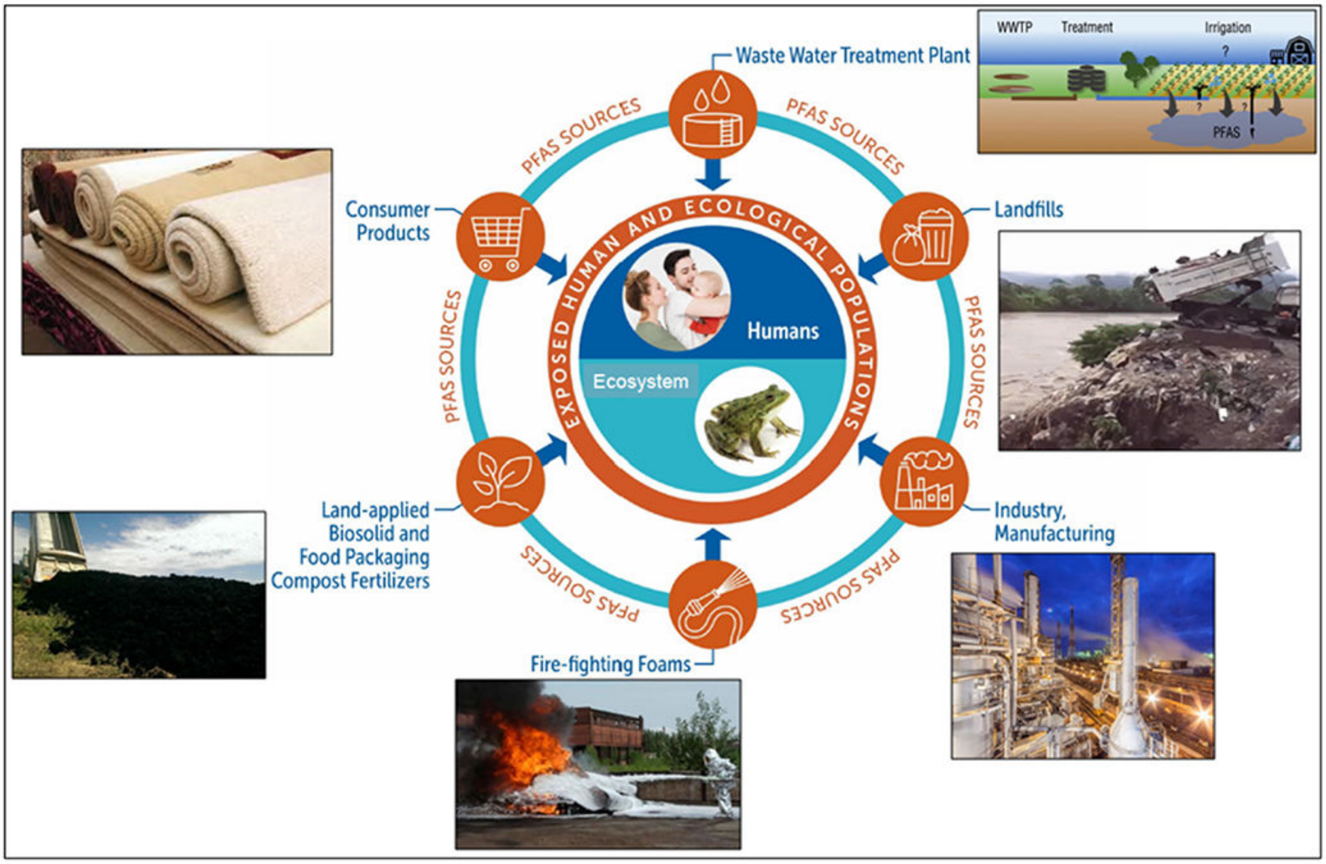

Fig. 1.

Schematic diagram on the sources of PFAS compounds (Ghisi et al, 2019).

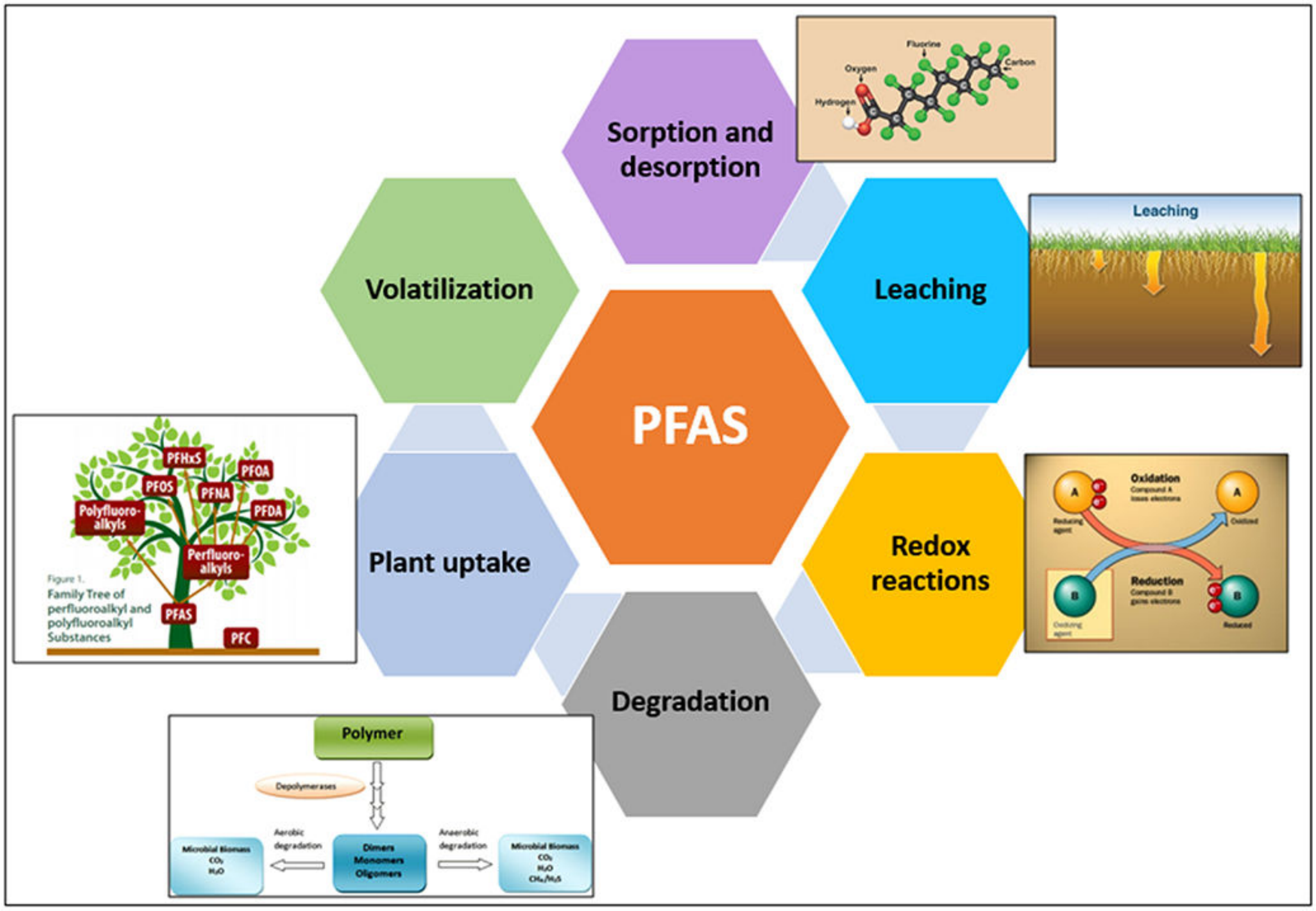

Fig. 2.

Schematic diagram on the dynamics of PFAS in soil.

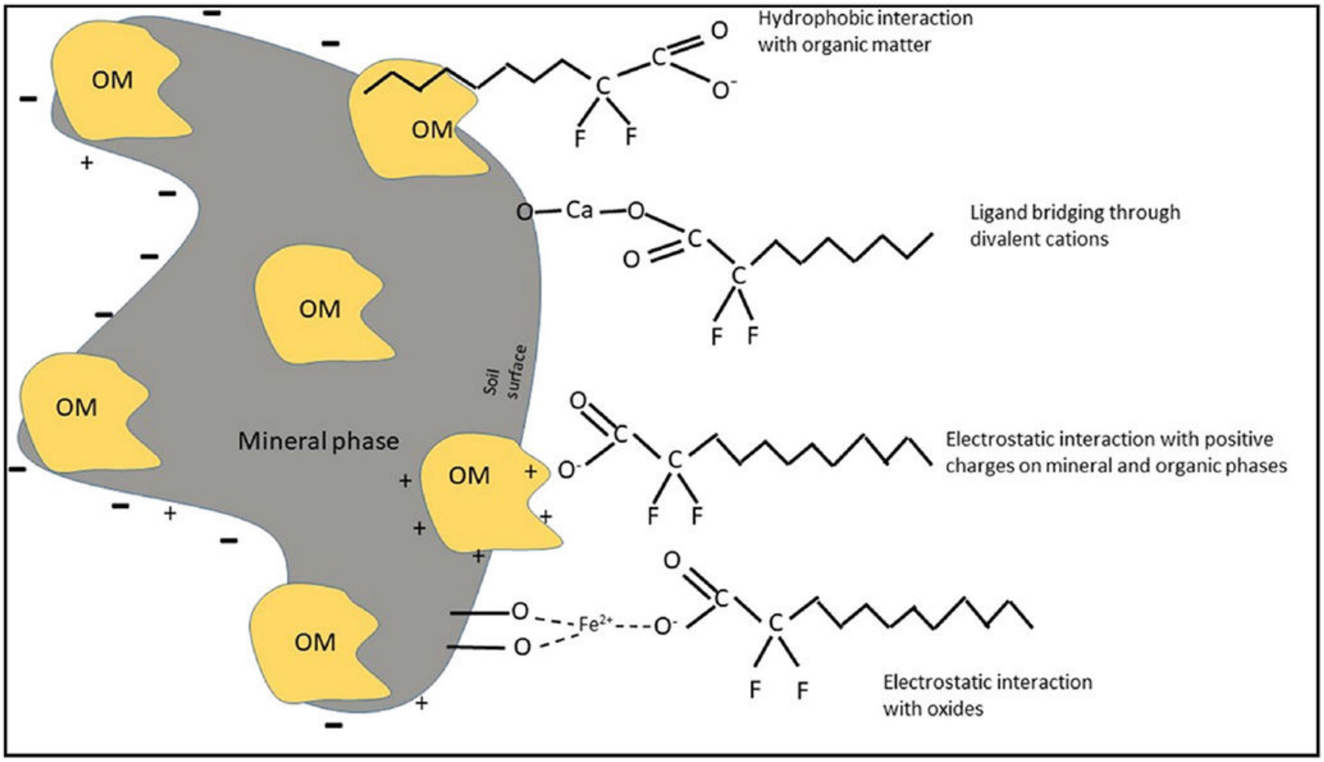

Fig. 3.

Sorption interactions of PFAS in soil (Ross et al., 2018; Li et al., 2018b).

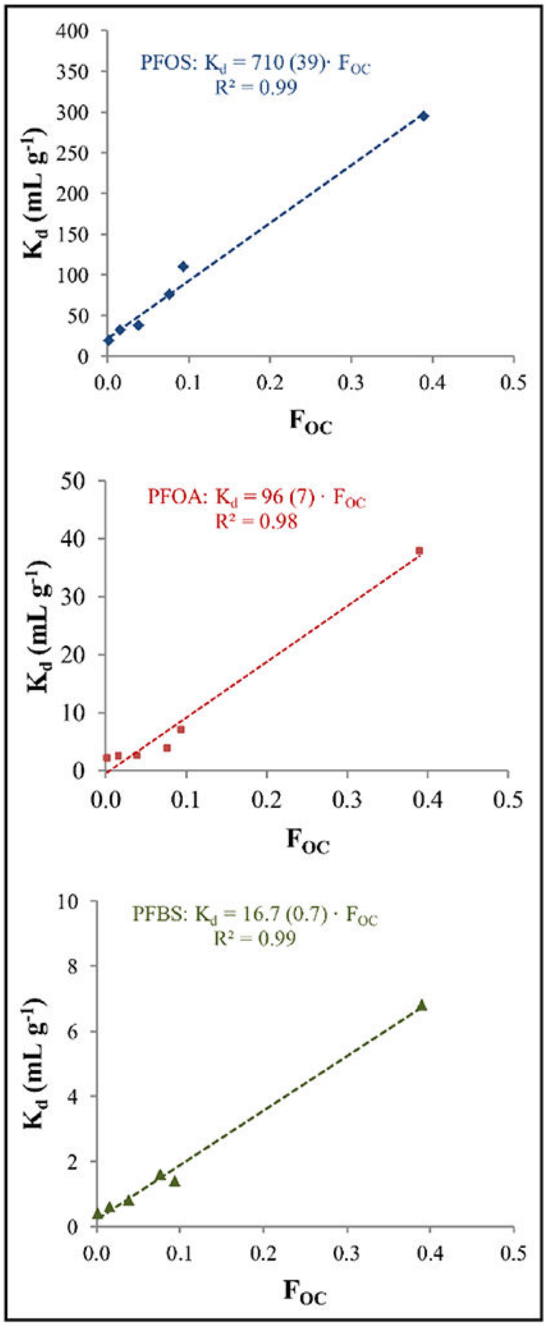

Fig. 4.

Relationship between organic matter and PFAS sorption by soils (Milinovic et al., 2015).

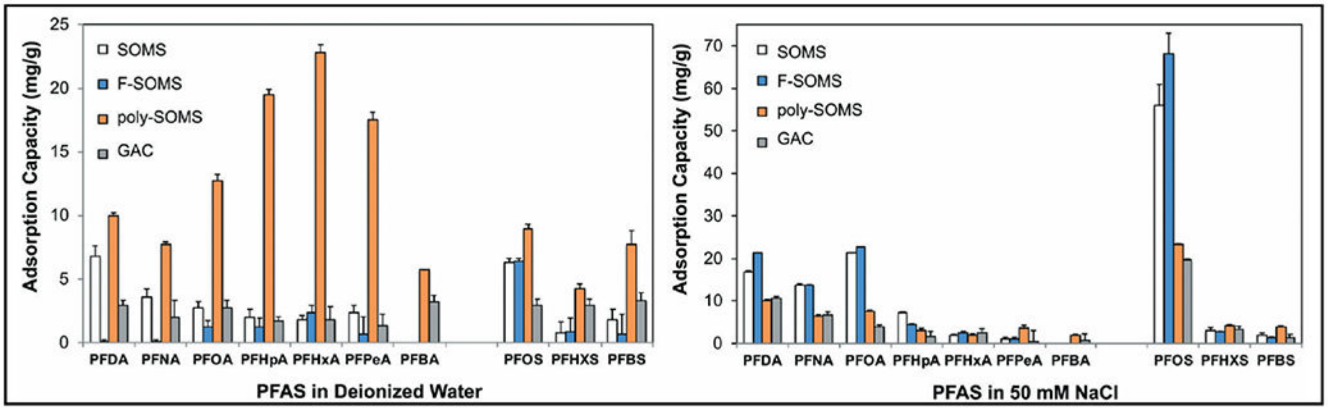

Fig. 5.

Sorption of PFAS by organo silicate compounds (SOMS = Swellable organically modified silica; F-SOMS = Fluoroalkyl-modified organosilica; poly-SOMS = entrapped cationic polymer-modified organosilica; GAC: Granulated activated carbon) (Stebel et al., 2019).

Table 1

Selected references on PFAS contamination in water and wastewater resources.

Source	Country	PFAS content	Reference
Surface and well water from areas associated with application of fluorochemical industry impacted biosolids	Alabama, USA	PFOA ranged from < LOQ (i.e., 10 ng/L) to 11,000 ng/L in well and surface water PFOA ranged from 594 to 2070 ng/l in drinking water samples PFOS ranged from < LOQ to 151 ng/L in well and surface water PFNA ranged from 12.4–286 ng/L in well and surface water PFDA ranged from 54.2 to 838 ng/L in well and surface water	(Lindstrom et al.,2011)
Groundwater, surface water, sewage treatment plant effluents and landfill leachates	Länsstyrelser,Sweden	Drinking water source area - average Σ_{26} PFAS 8.4 ng/L Landfill leachates - average Σ_{26} PFAS 487 ng/L Surface water - average Σ_{26} PFAS 112 ng/L Groundwater - Σ_{26} PFAS 49 ng/L Sewage treatment plant effluents - Σ_{26} PFAS 35 ng/L Background screening lakes - Σ_{26} PFAS 3.4 ng/L	(Ahrens et al., 2016)
Effluent and sludge in wastewater treatment plants	New York, USA	PFOA ranged from 58 to 1050 ng/l in effluent PFOS ranged from 3 to 68 ng/l in effluent PFOA ranged from 18 to 241 ng/g oven dry weight in sludge PFOS ranged from <10 to 65 ng/g oven dry weight in sludge	(Sinclair and Kannan, 2006)
Biosolids/ soil mixture exposed to ambient outdoor conditions	Maryland, USA	PFOA showed 24.1 ng/g dry weight PFUnDA showed 18.4 ng/g dry weight PFDA showed 17.4 ng/g dry weight Increased PFDA, PFDoDA and PFOSA over time from unidentified precursors	(Venkatesan and Halden, 2014)
Biosolids	Columbia and 32 Sates, USA	PFOS showed 403 ng/g dry weight in biosolids PFOA showed 34 ng/g dry weight in biosolids PFDA showed 26 ng/g dry weight in biosolids Mean load of ΣPFAS in biosolids estimated 2749–3450 kg/year	(Venkatesan and Halden, 2013)
Water, suspended particulate matter and sediment	Tokyo, Japan	ΣPFAS ranged from 16.7 to 42.3 ng/L in water column ΣPFAS ranged from 6.4 to 15.1 ng/g dry weight in suspended particulate matter ΣPFAS ranged from 0.29 to 0.36 ng/g dry weight in surface sediment	(Ahrens, Taniyasu et al. 2010)
Direct sources - Landfill leachate, water draining from a nearby military base/ urban area Indirect source — Infiltrated rainwater	Central part, The Netherland	Groundwater within landfill leachate plume-L-PFOA showed 1.8 μg/L PFBA showed 1.2 μg/L ΣPFAA showed 4.4 μg/L Maximum concentration of ΣPFAA in groundwater originating military base showed ~17 ng/L Maximum concentrations of L-PFOA and PFBA in groundwater halfway the landfill and public supply well field showed 29 and 160 ng/L, respectively L-PFOA and PFBA showed 0.96 and 3.5 ng/L, respectively in groundwater pumping wells	(Eschauzier, Raat et al. 2013)
Raw and treated landfill leachate	China	ΣPFAA ranged from 7280 to 292,000 ng/l in raw leachate ΣPFAA ranged from 98.4 to 282,000 ng/L in treated leachate	(Yan, Cousins et al.2015)
Surface sediment and sediment core samples	Lake Ontario,Canada	PFOS ranged from 0.684 to 51.8 ng/g dry weight in sediment samples PFDA ranged from 0.044 to 4.06 ng/g dry weight in sediment samples PFOS ranged from 0.492 to 30.1 ng/g dry weight in sediment core samples PFDA ranged from 0.024 to 1.676 ng/g dry weight in sediment core samples	(Yeung, De Silva et al. 2013)
Landfill leachates	USA	PFAA precursors (i.e., PFOSA and FTCA) ranged from ~4 to 36 μg/l ΣPFAS	(Benskin, Li et al.2012)

Source	Country	PFAS content	Reference
		ΣPFAS estimated to leave from 8.5 to 25 kg/yr the landfill via leachate for subsequent treatment at a wastewater treatment plant	
WWTP derived sludge-applied soils	Alabama, USA	PFDA showed ≤990 ng/g PFDDA showed ≤530 ng/g PFOA showed ≤320 ng/g PFOS showed ≤410 ng/g	(Washington, Yoo et al., 2010)
Biosolids amended soils	Beijing, China	Σ_9PFCA ranged from 18 to 113 ng/g dry weight Σ_3PFSA ranged from 23.4 to 107 ng/g dry weight ΣPFAA ranged from 41.4 to 220 ng/g dry weight	(Wen, Li et al. 2014)
Soil improver produced by industrial waste	Sauerland,Germany	PFOA showed > 0.5 µg/Lin drinking water ΣPFOS + PFOA showed <160 µg/L in creek water	(Wilhelm, Kraft et al. 2008)
Landfill leachate, biosolids	Australia	PFHxA ranged from 12 to 5700 ng/l in landfill leachate Decabromodiphenyl ether (BDE-209) ranged from <0.4 to 2300 ng/g in biosolids PFOS ranged from < LOD to 380 ng/g in biosolids	(Gallen, Drage et al. 2016)
Primary sludge, waste biological sludge, treated biosolids	Canada	PBDE ranged from 230 to 82,000 ng/g in primary sludge PBDE ranged from 530 to 8800 ng/g in waste biological sludge PBDE ranged from 420 to 6000 ng/g in treated biosolids	(Kim, Guerra et al. 2013)
Lime stabilized biosolids	Mid-Atlantic region, USA	PFNA showed 25.1 ng/g dry weight in sewage sludge PFOA 23.5 ng/g dry weight in sewage sludge PFOS 22.5 ng/g dry weight in sewage sludge	(Armstrong, Lozano et al. 2016)
Sewage sludge	Shanghai, China	ΣPFA ranged from 126 to 809 ng/g dry weight PFOA ranged from 23.2 to 298 ng/g dry weight	(Yan, Zhang et al. 2012)
Sludge	Nigeria	PFCA ranged from 10 to 597 pg/g in sludge PFSA ranged from14 to 540 pg/g in sludge PFOS showed 539.6 pg/g in hospital sewage sludge	(Sindiku, Orata et al. 2013)
Wastewater, sludge	Korea	PFOS ranged from 3.3 to 54.1 ng/g in sludge PFOA ranged from 2.3 to 615 ng/L in wastewater PFOA ranged from 3. 4 to 591 ng/L in influent and effluent wastewater	(Guo, Sim et al. 2010)
Influent, effluent, sludge	Spain	PFHxA showed 5.60 µg/L in water L-PFBS showed 0.31 µg/L L-PFOS showed 1.79 µg/g dry weight in sludge PFBA showed 1.88 µg/g dry weight in sludge	(Campo, Masia et al. 2014)
Biosolids	Canada	diPAP ranged from 4 to 83 ng/g dry weight PFCA ranged from 0.1 to 19 ng/ g dry weight	(Lee, Tevlin et al.,2014)

Table 2

Selected references on the sources of PFAS compounds in soil.

Sources	Country	Observations	Reference
Fluoropolymer manufacturing facility	USA	Both atmospheric deposition and groundwater recharge have contributed to the sources of the site contamination	(Zhu and Kannan, 2019)
Sulfluramid use in agriculture	Brazil	Soil contained ΣPFAS concentrations of up to 5400 pg/g with PFAS profiles generally dominated by PFOS and FOSA	(Nascimento et al., 2018)
AFFF	Australia	PFOS and PFHxS were the most dominant PFAA in all soil samples, with concentrations of PFOS reaching 13,400 ng/g	(Bräunig et al., 2019)
Mega-fluorochemical industrial park	China	Hotspots of contamination by PFAA were found near the mega-fluorochemical industrial park with the maximum concentrations of 641 ng/g in agricultural soil	(Liu et al., 2017)
PFSA manufacturing facility	China	total concentrations of PFAA ranged from 1.30 to 913 ng/g on a dry weight basis	(Gao et al., 2019)
AFFF	Sweden	PFOS and PFOA concentrations in the soil were ranging from 2.18 to 8520 ng/g and from <0.12 to 287 ng/g dry weight, respectively	(Filipovic et al., 2015)
PFAS-production facility	USA	12.2 ng PFOS/g dry weight and 8.0 ng PFOA/g dry weight	(Xiao et al., 2015)
3 M fluorochemical plant	Belgium	The mean concentrations of PFOA and PFOS in soil were 0.89 and 6.77 ng/g respectively	(Groffen et al., 2019)
Sludges generated from WWTP	USA	Highest concentrations were PFDA (≤990 ng/g), PFDDA (≤530 ng/g), PFOA (≤320 ng/g), and PFOS (≤410 ng/g)	(Washington et al., 2010)
AFFF	USA	PFOS was the PFAS species detected at the highest concentration on nearly every soil (median 2400 μg/kg) and aquifer solid (median 270 μg/kg) sample	(Houtz et al., 2013)
Biosolids generated from WWTP	Canada	Biosolid-amended soil exhibited increased concentrations of PFCA (0.1–19 ng/g dw)	(Lee et al.2013)
AFFF	Norway	PFOS accounted for 96 % of the total PFAS concentration in the soil with concentrations ranging from <0.3 μg/kg to 6500 μg/kg	(HØisæter et al., 2019)
AFFF	Australia	PFOS is the dominant PFAS measured, with concentration varying from 10 to 200 μg/g	(Baduel et al., 2015)
AFFF-impacted fire fighter training area	USA	PFOS surface soil levels reached 36,000 μg/kg around the burn pit	(McGuire et al., 2014)
Biosolids	Spain	Biosolid amendment increased concentrations 1.5–14-fold for PFAS	(Navarro et al., 2016)
Wastewater	Kampala, Uganda	ΣPFAS ranged from 1700 to 7900 pg/g dry weight in soil	(Dalahmeh, Tirgani et al., 2018)
Firefighting foams	France	Median value for ΣPFAS showed 12,112 ng/g in area #2 Median value for ΣPFAS showed 8701 ng/g in area #6 Mean value for ΣPFAS ranged from 1110 to 2237 ng/g and the maximum concentration observed <4300 ng/g in area #3 and #4	(Dauchy, Boiteux et al., 2019)
Biosolids	Australia	Annual load of PFOA in agricultural soils estimated 2.2 kg Annual load of PFOS in agricultural soils estimated 13 kg	(Gallen, Drage et al. 2016)
Biosolids	Columbia and 32 Sates, USA	Mean load of ΣPFAS 1375–2070 kg applied to agricultural land Estimated PFOA of 85.8–129 kg/year applied to agricultural land Estimated PFOS of 1026–1545 kg/year applied to agricultural land	(Venkatesan and Halden, 2013)

Sources	Country	Observations	Reference
Biosolids/ soil mixture exposed to ambient outdoor conditions	Maryland, USA	PFOA showed 24.1 ng/g dry weight PFUnDA showed 18.4 ng/g dry weight PFDA showed 17.4 ng/g dry weight Increased PFDA, PFDoDA and PFOSA over time from unidentified precursors	(Venkatesan and Halden, 2014)
WWTP derived sludge-applied soils	Alabama, USA	PFDA showed ≤990 ng/g PFDDA showed ≤530 ng/g PFOA showed ≤320 ng/g PFOS showed ≤410 ng/g	(Washington, Yoo et al. 2010)
Biosolids amended soils	Beijing, China	Σ_9PFCA ranged from 18 to 113 ng/g dry weight Σ_3PFSA ranged from 23.4–107 ng/g dry weight ΣPFAA ranged from 41.4–220 ng/g dry weight ΣPFAA in wheat root, straw, husk and grain ranged from 140 to 472, 36.2 to 178, 6.15 to 37.8, and 7.32 to 35.6 ng/g, respectively	(Wen, Li et al. 2014)
Sewage sludge	Shanghai, China	Predicted PFOA in agricultural land was 1.08 ng/g dry weight Predicted PFOA in agricultural land was 7.53 ng/g dry weight	(Yan, Zhang et al. 2012)
Biosolids	Chicago, USA	PFOS ranged from 2 to 483 ng/g in biosolids amended soil	(Sepulvado, Blaine et al. 2011)
Biosolids	Canada	Total diPAP showed up to 300 ng/ g dry weight in soil Total PFCA showed up to 50 ng/g dry weight in soil	Lee, Tevlin et al., 2013
Biosolids	China	PFOS ranged from 1.44 to 43.2 ng/g PFOA ranged from 1.21 to 28.5 ng/g	(Wen, Zhang et al. 2015)

Table 3

Selected references on the potential value of soil amendments in the mobilization of PFAS compounds in soils.

Amendments	PFAS compounds	Observations	Reference
0.01 mol/L $CaCl_2$ and 0.03 g of NaN_3	PFOA, PFBS and PFOS	Desorption yields of PFOA, PFBS and PFOS were 15-19%, 18-27% and <4%, respectively	Milinovic et al. (2016)
Acetic acid	PFCAs and PFSAs	Desorption experiments indicated desorption became difficult as the chain length increased, and PFSAs were harder to be desorbed than the corresponding PFCAs	Zhao et al. (2012)
Rainwater	PFOA and PFOS	Of the 360 g of PFOA and 367.5 g of PFOS applied to the soil, loss from the soil plot through leachate amounted to 3.12% for PFOA and 0.013% for PFOS. Short-chain PFASs and PFOA pass through the soil much more quickly than PFOS	Stahl et al. (2013)
Water	PFCs	The short chain PFC could pass through the soil without retention and were likely to be carried away easily with surface runoff	Gillrich et al. (2012)
Cationic surfactant, CTAB, and an anionic surfactant, SDBS	PFOS	While CTAB remarkably enhanced the sorption of PFOS on the sediment, SDBS increased the desorption of PFOS	Pan et al. (2009)
Oxalate and root exudates	PFOS	Oxalate increased PFOS desorption by 1.43- to 17.14-fold, effects of root exudates were similar to those of oxalate	Tang et al. (2017)
Methanol with ammonium acetate	86 PFASs	Methanol with hydrochloric acid provided excellent recoveries for most cationic and zwitterionic PFASs	Munoz et al. (2018)
100 mM of $CaCl_2$	6 PFCs	The desorption was lower than Adsorption, the soil matrix may act as a protective barrier towards extensive groundwater contamination	Enevoldsen and Juhler (2010)
Supercritical carbon dioxide (Sc-CO_2)	PFOS and PFOA	The extraction efficiencies (with double extractions) were approximately 77%-100% for PFOA and 59%-80% for PFOS	Chen et al. (2012b)
Ethanol	PFOS	The regeneration percent of PFOS from spent activated carbon was 84% after 0.5 h and 98% after 23 h using 50% ethanol solution at 45 °C	Deng et al. (2015)
Ethanol and NaCl in methanol solution	PFCAs	50% ethanol at 45 °C and 1% NaCl in 70% methanol solution were suitable for the desorption of PFCAs from the bamboo-derived activated carbon and resin IRA67, respectively	Du et al. (2015)
Ethyl acetate—dimethylformamide and methanol—phosphoric acid	PAEO, PFOS and PFOA	The sequential use of ethyl acetate—dimethylformamide and methanol—phosphoric acid in combination with pressurised liquid extraction resulted in exhaustive extraction of fluorinated anionic and non-ionic surfactants in sewage sludge	Schröder (2003)
NAPL, anionic surfactant sodium decyl sulfate (SDS) and *n,n*-dimethyldodecylamine *n*-oxide (AO)	PFAAs	NAPL, SDS and AO led to decrease in the sorption of PFOS at lower PFOS concentrations (1 μg/L)	Guelfo and Higgins (2013)

Table 4

Selected references on the potential value of soil amendments in the immobilization of PFAS compounds in soils.

Amendments	PFAS compounds	Observations	Environmental media	Reference
Activated carbon Montmorillonite clay Compost soil	PFOS	Leaching was reduced by 94–9.9% for AC, 29–34% for compost soil and 28–40% for the montmorillonite amended samples	Soil	Hale et al. (2017)
Powdered activated carbon (PAC), Rembind®, pulverized zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride	14 PFAS	Standardized leaching tests on S/S-treated soil revealed that leaching of 13 out of 14 target PFASs (excluding perfluorobutane sulfonate (PFBA)) was reduced by, on average, 70% and 94% by adding PAC and Rembind®		Sörengård et al. (2019a)
Poly(diallyldimethylammonium) chloride, poly(dimethylamine-co-epichlorohydrin)	PFOS	below the EPA health advisory level of 0.1 nM using poly (diallyldimethylammonium) chloride concentrations as low as 0.16 g/L	Soil	Anderson et al. (2019)
Palygorskite-based material modified with oleylamine	PFOS	a negligible release (water extractable) of PFOS (only 0.5 to 0.6 %) with a year incubating with amendment (10 % w/w)	Soil	Das et al. (2013)
Fine powdered, coal-based, activated carbon (AC)	PFOS, PFOA and PFHxS	4% (w/w) of the AC to contaminated soils resulted in almost complete removal of PFCs from the water phase	Soil	Kupryianchyk et al. (2016)
Modified clay adsorbent	PFOS	Soil treated with Clay adsorbent (10 % w/w) for a year, a negligible release of the contaminant (only 0.5 to 0.6 %)	Soil	Das et al. (2013)
Cationic polymers polydiallyldimethyl ammonium chloride and polyamine	PFBS, PFHxS, PFOS, PFBA, PFOA and PFNA	For all PFAS the retention on the column showed hysteresis where only 1 to 20% of the PFAS was recovered from the column after flushing with 30 pore volumes of simulated groundwater	Soil	Aly et al. (2019)
Colloidal activated Carbon	PFOS, PFOA	all PFASs were below their respective method detection limits in all post-injection samples	groundwater	McGregor (2018)
Colloidal activated carbon (PlumeStop ®)	14 PFAS	1% (w/w) colloidal AC treatment, PFAS leachability decreased by up to 81 % for i.e. PFOA	Soil	Sorengard et al. (2019b)
Laccase	PFOA	PFOA was degraded 24 % in water after 36 d, 40 % in soil slurry after 140 d	soil slurry	Luo et al. (2018)
Soil organic matter (SOM)	5 PFAS	peat soil with high fraction of organic carbon (foc, 59 %), the PFAAs were completely sequestrated in the soil	Soil	Zhao et al. (2016a)
SOM	PFOS	SOM in soils promoted sorption of PFOS; ferric oxides had the opposite effect	Soil	Qian et al. (2017)
Iron oxides, alumina and SOM	PFOS	Iron oxides, alumina and SOM were positively correlated to sorption KF	Soil	Wei et al. (2017)
Poly(dimethylamine-co-epichlorohydrin), poly-(diallyldimethylammonium)	PFOS	PFOS concentrations can be reduced below the EPA health advisory level of 0.1 nM using poly(diallyldimethylammonium) chloride concentrations as low as 0.16 g/L	Soil	Anderson et al. (2019)
Corn straw biochar	PFOS	The mobility of PFOS was significantly reduced after the addition of 5% biochar	sediments	Guo et al. (2019)
TiO_2	PFOS	The addition of TiO_2 NPs increased PFOS sorption by altering the sediment surface properties	sediments	Li et al. (2018)