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INTERACTIVE MECHANISMS BETWEEN PFAS AND SOIL CHARACTERISTICS: A REVIEW

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Abstract. Studies related to the interaction of PFAS (per- and polyfluoroalkylated) substances in soil media are essential for finding means of removal from the environment. This review focuses on the most important soil characteristics, such as pH, temperature, organic matter and nutrient content, potentially toxic elements, soil particle size distribution, total nitrogen content and cation exchange capacity, which determine the different interactions of PFAS in soil media, analysing the sorption mechanism at a deeper level. In addition, the analysis of the interaction of PFAS with soil properties is important for predicting the sorption of PFAS in soil. The requirement for an effective PFAS removal method and methodology with threshold values is greatly influenced by the properties of the medium, environmental conditions and PFAS characteristics. This review discusses the most important features of the interaction between PFAS and soil properties and the mechanisms of PFAS sorption in soil.

Keywords: PFAS, soil, sorption, potentially toxic elements.

1. Introduction

Aerogenic and edaphic pathways from stationary and diffusive contamination sources lead to the comprehensive dispersion of PFAS in the environment. Therefore, various types of soil for different uses are threatened by the PFAS effect. Moreover, PFAS becomes a co-contaminant to existing contamination load with inorganic (e.g., potentially toxic elements) and other organic compounds (e.g., PAH) (Feng et al., 2024; Sharma et al., 2024). Therefore, the interaction between the synergistic or antagonistic behaviour of PFAS and other media must be studied. Identifying each PFAS's structural peculiarities, including the structural allocation, functional group assignment, the orientation of each functional group, etc., is important to justify appropriate behaviour allocations in the soil matrix. C-F bonding formations in PFAS enhance the electronegative properties that contribute to the stability of PFAS in various environmental media and real-world industrial applications, characterised by distinct chemical and physical properties (Leung et al., 2023a; Wang et al., 2023). PFASs with functional groups consisting of carboxyl and sulphonic groups exhibit an acidic nature in antithetical existences. In contrast, some PFASs can be found with zwitterionic and cationic peculiarities, which have unique interactions. Apart from the functional group variations within the PFAS structure, other important structural arrangements should be considered to define the complex behaviour and interactions in different soils. PFAS can be further classified according to the carbon chain length, which is named short-chain PFAS with six or fewer perfluorinated carbon content, consisting of carboxylic functional groups and five or fewer perfluorinated carbon content, consisting of sulfonate functional groups. Major analytical concerns were fulfilled based on perfluorononanoic acid (C_9), perfluorodecanoic acid (C_{10}), perfluoroundecanoic acid (C_{11}), perfluorododecanoic acid (C_{12}), perfluorotridecanoic acid (C_{13}) and perfluorotetradecanoic acid (C_{14}), which consist of carboxyl functional groups and

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perfluorooctane sulfonic acid (C_8), which consists of sulfonate functional group. Several factors were considered when selecting the PFAS for comprehensive analysis and further justification. Long-chain PFAS have more interactive properties, such as hydrophobic nature and bioaccumulation concerns, which depend on the contaminants' presence in the soil. There is a requirement to fill the knowledge gap between the mixed PFAS existences in different soil media characteristics and those are highlighted considering the long-chain PFAS behaviour in different soil media (Feng et al., 2024; Leung et al., 2023a; Sharma et al., 2024; Wang et al., 2023).

Considerable investigations have been conducted to identify such behaviours of different long-chain PFAS in soil matrices with varying soil characteristics according to the above-mentioned directions. Soil pH, temperature, soil organic matter (SOM), soil nutrient content, potentially toxic elements, soil texture, total N content, cationic exchange capacity, and anionic exchange capacity are some important parameters to be considered to describe the selected range of PFAS interaction and retention in the soil. Different reviews and research articles (Feng et al., 2024; CHEM Trust, 2013; Mirabediny et al., 2023; OECD, 2024; Veciana et al., 2022; Verma et al., 2023; Wang et al., 2023; Zhang & Yazaydin, 2024) have highlighted the behaviour of different PFASs under varying soil allocations to describe the movements of PFAS in soil matrices with minimum contact levels in the presence of different adsorption materials. This review aims to identify and highlight the soil characteristics that induce strong interactions and retention of PFAS.

2. PFAS structure, behaviour, and related mechanisms

PFAS exhibit unique stability and are widely spread due to several properties. Structural characteristics and functional groups intervening in the interactions with the surrounding environment can be described using two main properties of PFAS. Major structural component arrangements, such as carbon chain length and the number of carbons present in the main structure of PFAS, must be identified with specific characteristics for describing the interactions with soil matrices and carbon-based adsorptions. Another important feature is based on the functional group present in the PFAS structure. This classification is important as several functional groups initiate other interaction derivations. Functional group interactions and presentation studies have been covering perfluoroalkyl acid (containing carboxyl groups), perfluoroalkyl sulphonamides (containing a sulfonic group), fluorotelomers (partially fluorinated carbon chains), and polymeric PFAS. Once the number of carbons present in the chain, PFAS can be categorised as long-chain and short-chain based on the number of carbons present in a PFAS structure (ITRC, 2020b; OECD, 2024). If the number of carbon atoms counts as seven or more than seven, it can be categorised as long-chain PFAS, and less than seven is categorised as short-chain PFAS.

This property is comprehensively important as it highlights information such as water solubility, interactions, and bioaccumulation strength in different media. The tail is said to be a perfluorinated part of the structure, and the functional group acts as the head of the structure (Feng et al., 2024; Leung et al., 2023a; Wang et al., 2023). These arrangements are critical in terms of interactions such as hydrophobic interactions, electrostatic interactions, ligand exchange interactions, hydrogen bonding interactions, etc. (Sharma et al., 2024; Zhang & Yazaydin, 2024). Fluorine is the most important and critically focused element in PFAS structure which acts as a protective shield and forms C-F bonds to have more stable structures chemically and physically (Feng et al., 2024; ITRC, 2023; Leung et al., 2023b; Wang et al., 2023). This positioning makes PFAS more polar than the other related hydrocarbons increasing the acidic conditions that impact the above-mentioned interactions with soil media conditions and other substances (Feng et al., 2024; Wang et al., 2023). The soil interactions with PFAS should be further evaluated due to the weak formation of hydrogen bonds. This confirms that the properties must be aligned following the structural allocations with soil characteristics. When comparing the fluorine atoms with hydrogen atoms, the fluorine atoms are bigger, which will induce the PFAS structure to be more stable than other hydrocarbons (Feng et al., 2024; ITRC, 2023). Due to the low degree of polarisation of PFAS, those structures do not bond, which directs toward higher volatilisation in the environmental media and other soil characteristics. The structural arrangements of PFAS further reduce surface tension and encourage it to spread across different media, exhibiting an amphiphilic nature (Trinh et al., 2022; Wang et al., 2023). This forms a fluorosis phase, characterised by interactions with water and organic solvents. Micelle formation is another phenomenon in PFAS that forms a spherical cluster, trapping other contaminants and facilitating their movement from one location to another (Leung et al., 2023b; Shahrokhi & Park, 2024). This study examines the behaviour of long-chain PFASs such as PFNA (C_9), PFDA (C_{10}), PFUnDA (C_{11}), PFDoDA (C_{12}), PFTrDA (C_{13}), PFTeDA (C_{14}), consisting of carboxyl functional group, and PFOS (C_8), which consists of sulfonic functional group, and have common chemical and physical characteristics

due to the existence of fully fluorinated carbon chains, while different in terms of chain length and functional group arrangements. Chain length increments in the above-mentioned PFASs justified more allocation towards higher hydrophobicity and intense interactions with different soil characteristics, especially soil with high organic content, such as peat and loamy soil, which confirms the hydrophobic interactions (Feng et al., 2024; Wang et al., 2023). Due to this, leaching is minimised towards the groundwater aquifers in such arrangements. Therefore, retention is high compared to other soil textures (Feng et al., 2024; Wang et al., 2023). All the hydrogen atoms are entirely replaced by the fluorine elements with high energy between C-F bonds, emphasising the resistance towards stability breakdowns under different conditions (Feng et al., 2024; Wang et al., 2023). Steric hindrance confirms the stability of the overall structure of PFAS, which is formed due to the fluorine's van der Waals radius becoming a more rigid structure (Barlow et al., 2019). This feature will influence the interactions with SOM when focusing on the functional groups influences of PFNA (C_9), PFDA (C_{10}), PFUnDA (C_{11}), PFDoDA (C_{12}), PFTrDA (C_{13}), PFTeDA (C_{14}), consisting of carboxyl groups, which confirm the interactions with positively charged ions $Ca^{2+,}$ Mg^{2+}, metal oxides, minerals, etc., due to the formation of negative charge from the deprotonation and exhibiting electrostatic interactions (Feng et al., 2024). PFOS(C_8) exhibits strong hydrophilic properties due to the sulfonate functional group, which promotes higher interactions with higher CEC conditions. The other important factor to be considered in the PFAS allocation is the molecular weight of each PFAS (Feng et al., 2024; Wang et al., 2023). Studies show that the adsorption nature increases with the PFAS substances' molecular weight (ITRC, 2020a; Wang et al., 2023). Further, the PFOS (C_8) functional groups generate ion and ligand exchange interactions to form more complexes, which enhance the adsorption procedures within the soil matrices (Feng et al., 2024; ITRC, 2020a; Wang et al., 2023; ECHA, 2023).

3. Soil matrix characteristics and dominance behaviours on PFAS

The adsorption nature of the PFAS in soil and related substances is comprehensively induced by several factors identified by different research studies (Feng et al., 2024; Wang et al., 2023). Those can be highlighted as soil pH, temperature, SOC, soil nutrient contents, potentially toxic elements, soil texture, total N content, and cationic exchange capacity. According to studies carried out by Feng et al. (2024) and Wang et al. (2023), soil organic matter (SOM) is a significant factor influencing adsorption processes. In organic materials-rich soils, more adsorptions are initiated due to stronger hydrophobic natures (Feng et al., 2024; Wang et al., 2023) Once the PFAS and interactions with soil consisting of humic substances in soil behaviour can be highlighted under three different components: humic acid, fulvic acid, and humin. Humin is a condition of SOM consisting of high molecular weight organic materials with minerals resulting in strong bond features and insoluble ones (Hayes et al., 2017). This feature increases the adsorption and retention capabilities while ensuring comprehensive interactions of long-chain PFAS in the soil media. Soil humin substances are more effective for the longest-chain PFAS interactions, where more hydrophobic and van der Waals forces fractions exist (Qi et al., 2022). Hydrogen bonding and hydrophobic interactions comprehensively act in the presence of humic acids due to the presence of aromatic structures and functional groups such as carboxyl, hydroxyl, etc. (Feng et al., 2024; Wang et al., 2023). Each selected long-chain PFAS exhibits strong hydrophobic interactions with SOM due to the long tails. In higher temperatures, the decomposition of SOM will reduce the organic content in the soil, which will cause a reduction in PFAS adsorption (Feng et al., 2024; Wang et al., 2023).

Soil pH has an impact on the surface charges of minerals and a direct impact on the ionisation of soil organic matter. Soil humic substances consist of functional groups such as carboxyl, hydroxyl, etc., which have more protonation capacities in low pH (acidic soil conditions), resulting in higher retention of PFAS in soil through electrostatic interactions. Higher pH conditions (alkaline soil conditions) cause deprotonation from the organic matter and mineral surfaces, resulting in repulsive forces with anionic PFASs, making PFAS more mobile in the media conditions. Acid buffering capacity and surface charge effects on adsorption conditions are important readings that can be gained through the pH measurements in soil media (Feng et al., 2024; Wang et al., 2023). Soil pH in water (pH_{H2O}) can be measured using distilled water, which reflects the actual pH of soil in natural conditions. Resulted values for the exchangeable acidity pH (pH_{HCL}) of the soil highlight the release capabilities of H^+ to soil media, which can be justified as higher PFAS mobility. In other words, if (pH_{H2O}) is higher than (pH_{HCL}), the negative charge of the soil media will increase, causing higher mobility of carboxylated and sulfonated PFAS in the media. If the measured values are similar, it emphasises that the soil conditions are more stable in retaining PFAS. These pH values give comprehensive insights into further studies on interactive behaviour of PFAS and soil. In acidic soil conditions, protonation will happen, resulting in more

cations retained in the soil, increasing CEC to hold negatively charged carboxylated and sulfonated PFASs, thus reducing the movements (Wang et al., 2019; Feng et al., 2024; Wang et al., 2023). CEC is another feature that determines the retention of PFAS in the soil media. It highlights the soil's ability to retain positively charged ions. Soils with more clay and organic matter content can retain positive cations (divalent and trivalent), which act as a bridge to connect PFAS and soil particles through the existing ions. These bridging conditions enhance the retention of PFAS in the soil media. Sandy soil has a lower capability to develop cation exchange capacities (CEC) due to the low existing nature of soil organic matter (SOM), which results in a reduction of PFAS retention in such soil. Therefore, the mobility of the PFAS increases in the low CEC soil (Feng et al., 2024; Wang et al., 2023). The soil's total nitrogen (TN) amount is another important parameter that determines the adsorption consequences of PFAS (Wu et al., 2023; Feng et al., 2024; Wang et al., 2023)Higher nitrogen contents in soil media justify the presence of organic and inorganic substances in soil media, enhancing the PFAS adsorption(Wu et al., 2023; Feng et al., 2024; Wang et al., 2023) Lower nitrogen content indicates less PFAS retention, which enhances the mobility of PFAS in the soil media (Feng et al., 2024; Wang et al., 2023; Wu et al., 2023). The addition of nitrogen with fertilisers in terms of ammonium ions replaces some spaces on the soil surface (Feng et al., 2024; Wang et al., 2023; Wu et al., 2023). Microbes exist in soil pore water and trap PFAS, making a layer that retains PFAS in soil (Feng et al., 2024; Wang et al., 2023; Wu et al., 2023). Therefore, PFAS can move with pore water within the soil (Feng et al., 2024; Wang et al., 2023; Wu et al., 2023). Loss on ignition (LOI) is a comprehensive parameter that can provide insights into PFAS adsorption (Feng et al., 2024; Wang et al., 2023; Wu et al., 2023) Organic matter and suitable carbonate contents can be gained through this parameter analysis. Higher LOI implies a higher SOM content in the soil, which indicates a higher adsorption of long-chain PFAS in that soil (Kikuchi et al., 2018). Potential toxic elements (PTE) such as Cd, Cr, Cu, Mo, Hg, V, Ni, Pb, Se, and Zn have recently become more investigated in different media due to several impacts on environmental conditions and human health (Huque et al., 2002; Wang et al., 2023). There are active sites in the mineral particles, soil, sediments, and adsorbents (Chen et al., 2009; Daniels et al., 2004). Competition exists for that active site under the different surrounding characteristics when considering different PFAS and PTEs (Feng et al., 2024). Cations are more direct towards negatively activated sites, along with cationic PFAS substances, which altered the PFAS adsorption rate related to the existing metallic nature (Feng et al., 2024). Clay minerals and SOM, except substances such as kaolin (positively charged), are negatively charged and interact with PTE, which are in cationic form, to cause the reduction of the negative charge while enhancing the adsorption of PFAS (Feng et al., 2024; Huque et al., 2002). As most of the PFASs are presented with negatively charged functional groups, those PTEs act as bridging components that enhance the adsorption and interactions with soil (Daniels et al., 2004; ECHA, 2023; Feng et al., 2024; Wang et al., 2023).

4. Conclusion

1. Soil characteristics such as pH, temperature, soil organic matter, soil nutrient contents, potentially toxic elements, soil texture, total nitrogen content, and cationic exchange capacity significantly influence long-chain PFAS retention due to several interactions, such as hydrophobic, electrostatic, ion exchange, and hydrogen bond formations.

2. Organic-rich fine-textured soil, acidic conditions (low pH), and higher cationic exchange potential promote higher sorption capacity of PFAS.

3. Potential toxic elements contribute in different ways to PFAS sorption by forming complexes and competitive adsorption on soil.

4. Therefore, multi-contaminated soil matrices that consist of potential toxic elements and total N content exhibit complex interactions between contaminants and PFAS, providing competitive sorption protocols.

5. Considering the above, a sustainable composite material allocation should be justified to mitigate competitive sorption while overcoming the soil physicochemical characteristics to ensure a comprehensive PFAS treatment.

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INTERAKTYVŪS PFAS IR DIRVOŽEMIO CHARAKTERISTIKŲ MECHANIZMAI: APŽVALGA

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Santrauka. Tyrimai, susiję su PFAS (per- ir polifluoralkilintų) medžiagų sąveika dirvožemio terpėje, yra būtini ieškant šalinimo iš aplinkos priemonių. Šioje apžvalgoje daugiausia dėmesio skiriama svarbiausioms dirvožemio savybėms, tokioms kaip pH, temperatūra, organinės medžiagos ir maistinių medžiagų kiekis, potencialiai toksiški elementai, dirvožemio granuliometrinė sudėtis, bendrasis azoto kiekis ir katijonų mainų geba. Šios savybės nulemia PFAS sorbcijos mechanizmus dirvožemyje ir padeda prognozuoti teršalų dinamiką. PFAS šalinimo efektyvumui įtakos turi ne tik dirvožemio savybės ir aplinkos sąlygos, bet ir pačių medžiagų charakteristikos. Apžvalgos tikslas – aptarti svarbiausius PFAS sąveikos su dirvožemiu bruožus bei sorbcijos mechanizmus.

Reikšminiai žodžiai: dirvožemis, PFAS, potencialiai toksiški elementai, sorbcija.