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Sustainable remediation strategies for emerging PFAS contaminants in water

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At present, the decline in drinking water quality may pose risks to human health. Various types of organic as well as inorganic contaminants in drinking water are reported globally. Among emerging organic contaminants, poly- and perfluoroalkyl substances (PFAS) have been identified in water as major concerns for humans due to their toxic health effects. Anthropogenic activities are considered the dominant source of PFAS addition in the hydrosphere. PFAS toxicities are largely chronic in nature, as they may be carcinogenic and can lead to central nervous system disorders and reproductive problems. This review provides a critical discussion of the different remediation techniques for PFAS, such as chemical redox reaction, adsorption, electrochemical treatment, bioremediation, membrane filtration, and the treatment-train approach. Despite these techniques, extensive future research is required to develop better techniques for PFAS remediation.

1. Introduction

Since the 1950s, per- and polyfluoroalkyl substances (PFAS) have been employed as synthetic materials in industry throughout the world. They are persistent in nature, and therefore have numerous applications in industrial as well as consumer settings as surfactants and external protectors, and are being extensively used throughout the world.[1–4] According to Buck *et al.*[5] highly fluorinated aliphatic substances are a subset of fluorinated substances. These substances may contain one or more C atoms in which all H substituents of the corresponding non-fluorinated analogues are replaced by F atoms, resulting in the perfluoroalkyl moiety C_nF_{2n+1}. As these substances possess higher stability as well as hydrophilic and lipophilic characteristics, they have numerous industrial applications, such as in fire-resistant fabric, non-stick cookware, waterproofing, food packaging, and firefighting foams.[6–8]

Owing to their stability, these substances are not readily degraded and can accumulate in the atmosphere, leading to their global distribution. Nearly every individual in the world has been in contact with PFAS and has traces of PFAS in their blood.[9,10] Some of these substances enter the ecosystem and food chain, where they become bioaccumulated in the organs of both humans and animals. Due to the persistent and bioaccumulative nature of perfluorooctanesulfonic acid (PFOS), its salts, and perfluorooctane sulfonyl fluoride (PFOSF), these substances were added to the persistent organic pollutants (POPs) list under the Stockholm Convention on Persistent Organic Pollutants in 2009. In addition, C_8–C_{14} perfluoroalkyl carboxylates (PFCAs) and perfluorobutane sulfonate (PFH_xS), and their sodium and ammonium salts have been included in the candidate listing of Substances of Very High Concern (SVHC) in the EU. Meanwhile, the production of AFFFs (aqueous film-forming foams) consisting of long-chain PFAAs is banned in Australia.[11–14]

The regular use of AFFFs is a crucial legacy problem. AFFF has been broadly applied for fire protection personnel, particularly where petroleum hydrocarbons are stored and used, at airfields and in the petroleum industry.[15–17] During regular training of firefighters and testing of fire protection systems, large amounts of AFFF have been released into the environment, and it has been observed at many sites, such as air force bases, metropolitan and rural fire training grounds, and in the petroleum industry at refineries and oil terminals.

The ingredients of PFAS in the foam are persistent in nature and can be adsorbed in soil and groundwater, ultimately moving several kilometers away from the source.[18,19] There are some sites where many PFAS are present at higher concentrations in groundwater as well as surface water.[20] As fluoro-alkyl compounds are stable under usual atmospheric conditions,

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they persist in the environment for several years once released.[21] Long-term exposure to PFAS in an individual leads to their accumulation in organs, resulting in severe toxicity.[22,23]

The term PFAS represents a broad group that contains about 4700 compounds, and their characteristics correspond to the multiple fluorine atoms attached to alkyl chains of variable length. They are categorized as short-chain and long-chain PFAS. Short-chain PFAS, the most persistent of the two groups, are highly stable. Long-chain PFAS are also stable but can degrade into short-chain PFAS.[24–26] Initially, PFAS were believed to be nonreactive and non-toxic, and little attention was paid to their environmental fate or their effects on the health of humans as well as the ecosystem. However, PFAS have become a global concern owing to their omnipresence, high stability, and increasing reports of toxicity in both animals and humans. PFAS comprise thousands of compounds, with perfluorooctanoic acid (PFOA) and perfluoro-octane sulfonic acid (PFOS) being the most extensively studied.[26,27]

PFAS are a group of organofluorine substances with a hydrophilic functional group (carboxylic acid or sulfonic acid) head along with a hydrophobic fluorinated carbon chain. The main families of PFAS comprise perfluoroalkyl sulfonic acids (PFSA), perfluoroalkyl carboxylic acids (PFCA), and the potential PFCA and PFSA precursor compounds. PFCA has a perfluorinated carbon chain, typically containing up to 16 carbon atoms, along with functional groups such as carboxylic acids. PFSA is a perfluorinated compound with a sulfonic acid functional group. Table 1 compiles the chemical structures of PFAS that are commonly used worldwide.

Since the 1960s, PFAS have been used globally in a broad range of products due to their water- and oil-repellent properties. They are generally employed as stain protectors for products in the textile, upholstery and carpet industries.[29,30] Additionally, they are used as specific surfactants in the fluoropolymer industry. In addition to this, they have been used in the production of important firefighting foams. Moreover, PFAS are also used to make precise extinguishing materials as well as to store large amounts of flammable chemicals.[12–31] Due to their widespread use, traces of PFAS have been detected in industry facilities, living organisms, wastewater, commercial household products and food items.[32,33] The exposure of humans to PFAS (at small concentrations) may result in accumulation in the blood of individuals.[34] In this review, the authors discuss the detrimental effect of PFAS contamination in water on human health. Furthermore, diagnosis and remediation techniques are also extensively reviewed.

2. Transport of PFAS in an aqueous environment

Lower PFAS concentrations have been found in aqueous solutions between pg L^{-1} and μg L^{-1} levels.[35,36] Long-chain compounds, such as perfluorooctanoic acid (PFOA), as well as perfluorooctane sulfonic acid (PFOS), are the widely assessed PFAS.[37] Numerous studies have confirmed the large presence of PFAS in aqueous environments (river water, storm water, lake water, groundwater, wastewater, and drinking water).[38–42] A portion of PFAS remains in wastewater treatment plants and is subsequently released into the receiving environment, contributing to their presence in aqueous systems. A recent study suggests that the potential impact of PFAS on groundwater is linked to the use of treated wastewater for irrigation.[43] Insufficient data are available for the removal efficiency of conventional water treatment plants and the transformation of PFAS precursor compounds; therefore, their occurrence and distribution are only partially understood.[38,42] Applications of PFOS and PFOA have decreased because of their detrimental effects on the health of the public, but there has been no decrease in the total quantity of PFAS released into the environment, due to the replacement of long-chain compounds by short and ultra-short PFAS.[41] Short and ultra-short PFAS have lower bioaccumulation as well as bioconcentration *via* trophic levels in contrast to long-chain species; nevertheless, they are environmentally persistent.[44,45]

3. Sources of PFAS

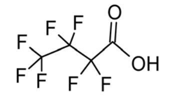
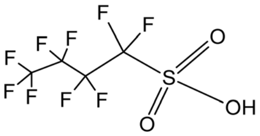
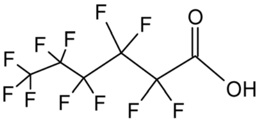
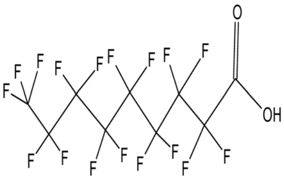
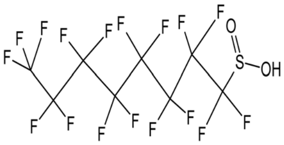
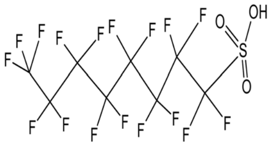
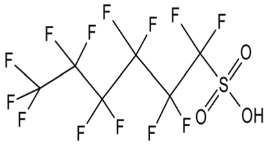
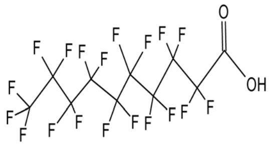
Recently, approximately 80% of electronic waste (e-waste) has been disposed of directly in landfills or indirectly ended up there due to inadequate recycling practices.[46] Regardless of the alarming consequences, e-waste substances as sources of PFAS in the environment around processing sites and landfills are continuously overlooked.[47–49] Because of limitations in analytical methods for detecting PFAS, relatively low concentrations in mixed substances, a lack of specific regulatory policy measures, non-specific e-waste processing, and the high costs for treating e-waste, PFAS compounds are often not properly detected. Fig. 1 shows the major sources of PFAS in aqueous environments along with their transport routes.

E-waste leachates have been reported as sources of hazardous waste because of the occurrence of alkanes, halogenated plasticizers, heavy metals, and dioxins; nevertheless, the occurrence of PFAS is overlooked.[50,51] The leachate generated by treatment at landfills is either treated at on-site treatment plants or transported to wastewater treatment plants, and none of the treatment trains put in place are adequate to remediate or destroy PFAS compounds. Therefore, e-waste treatment plants, landfills, and wastewater-treatment plants are considered the point sources of PFAS pollution in an aqueous environment.[52,53] Additionally, PFAS manufacturing and processing facilities, airports, and military installations that use firefighting foams are some of the main sources of PFAS.[54,55]

4. Harmful effects of PFAS

Proliferated peroxisome expression is the primary mechanism by which PFAS are known to exhibit their toxicity. Mammals, such as rodents, have been used in experiments to try and understand the harmful consequences of PFAS. However, because these organisms differ in how peroxisome proliferation is expressed, it is not fully acceptable to extrapolate the findings of this research to humans.[56] A possible correlation has been suggested by numerous case studies examining the negative

Table 1 Structure of some important PFAS (modified from Ahrens *et al.*[28])

Name	General formula	IUPAC name	Chemical structure
Perfluorobutanoic acid (PFBA)	$C_4HF_7O_2$	Heptafluorobutanoic acid hydrate (1 : 1)	
Perfluorobutanesulfonic acid (PFBS)	$C_4HF_9O_3S$	1,1,2,2,3,3,4,4,4-Nonafluorobutane-1-sulfonic acid	
Perfluorohexanoic acid (PFHxA)	$C_6HF_{11}O_2$	Undecafluorohexanoic acid	
Perfluorononanoic acid (PFNA)	$C_9HF_{17}O$	Heptadecafluorononanoic acid	
Perfluorooctanoic acid (PFOA)	$C_8HF_{15}O_2$	1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-Heptadecafluorooctane-1-sulfinic acid	
Perfluorooctanesulfonic acid (PFOS)	$C_8HF_{17}O_3S$	1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,8-Heptadecafluorooctane-1-sulfonic acid	
Perfluorohexanesulfonic acid (PFHxS)	$C_6HF_{13}O_3S$	1,1,2,2,3,3,4,4,5,5,6,6,6-Tridecafluorohexane-1-sulfonic acid	
Perfluorodecanoic acid (PFDA)	$C_{10}HF_{19}O_2$	Nonadecafluorodecanoic acid	

effects of PFAS exposure in humans, between PFAS compound exposure and the emergence of diseases such as hypertension, cancer, thyroid disease, ulcerative colitis, *etc.*[57,58] Children are more vulnerable to PFAS exposure than adults, according to a study.[59] This may be related to children's faster metabolisms and constantly changing and developing physiological systems. Epidemiologic data linking prenatal and/or childhood exposure to polyfluoroalkylamides with immunity, infection, asthma, cardio-metabolic problems, changes in neurodevelopment, thyroid issues, renal issues, and puberty onset were recently reviewed.[59] Food and drinking water are the primary pathways that expose human beings to PFAS, among several other routes.[60] A group of Chinese women's plasma PFAS concentrations were positively correlated with fish and crustacean consumption; however, their plasma PFAS concentrations were negatively correlated with the consumption of drinking water and soy products.[61]

4.1 Cancer

PFAA carcinogenic effects have mostly been investigated in human subjects. Communities consuming PFAS-contaminated water and chemical industry personnel exposed to PFOA and

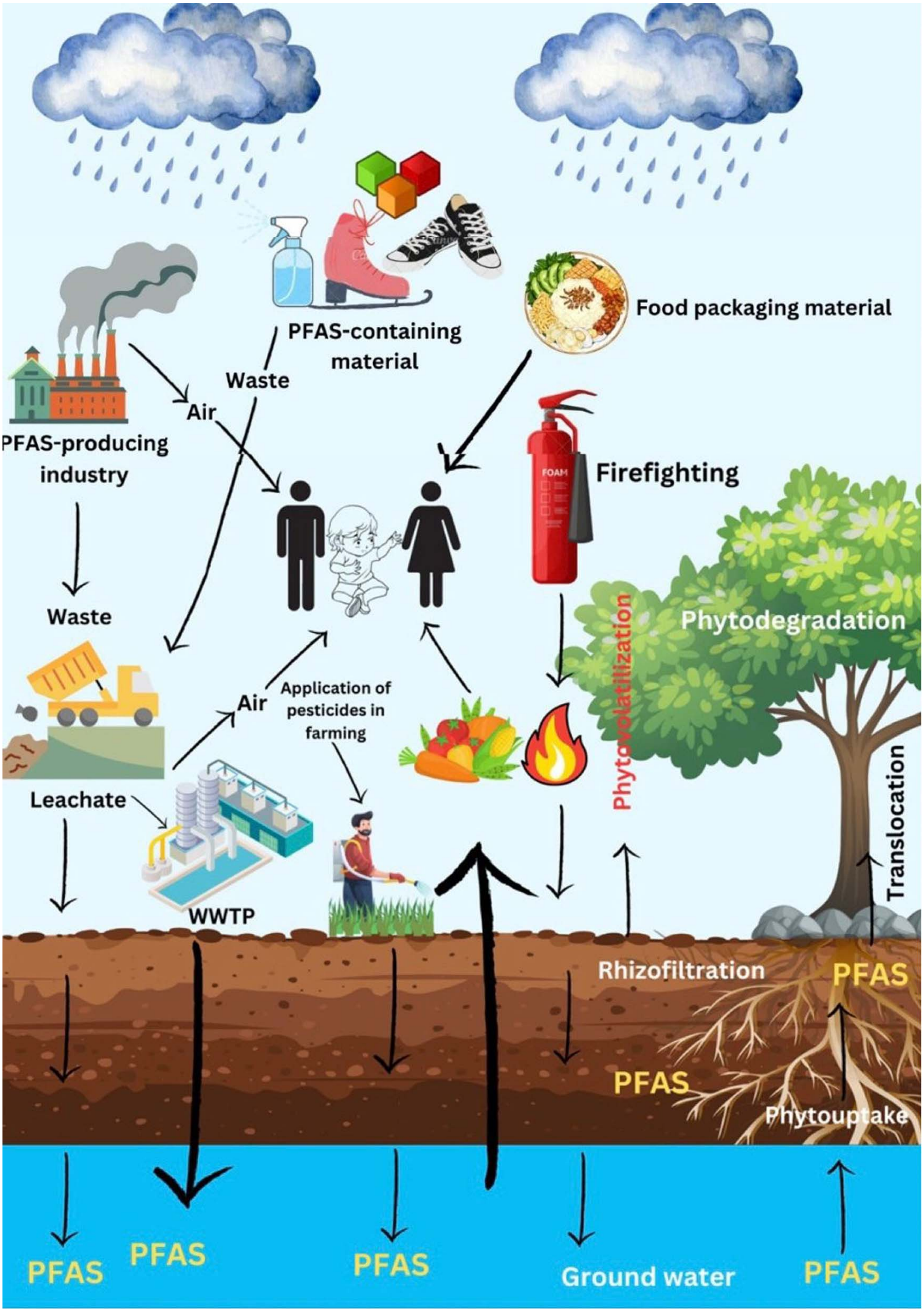


Fig. 1 Sources and routes of PFAS in the environment.

PFOS were the primary subjects of this investigation. According to the International Agency for Research on Cancer (IARC), PFOA may be carcinogenic to humans. Male employees at PFOA-producing plants had a 3.3-fold increase in prostate cancer mortality during the past ten years.[62] A follow-up investigation using a group for occupational exposure failed to find any evidence of a significant correlation between cancer mortality and ammonium perfluorooctanoate exposure; among exposed workers, no cases of kidney, prostate, or breast cancer were noted.[63] Two studies that focused on towns in the Mid-Ohio Valley and West Virginia located near chemical plants examined PFOA-contaminated drinking water, and found a link between PFOA exposure and cancer.[57,58] Two studies that focused on communities that had access to PFOA-contaminated drinking water due to their proximity to chemical facilities in the Mid-Ohio Valley and West Virginia reported a link between PFOA exposure and cancer. The research examines PFOA serum levels and medical history in a cohort of adult C8 Health Project participants, revealing a strong correlation between kidney and testicular cancer and serum PFOA concentrations.[57,58] To assess the cancer risk associated with exposure to PFOA and PFOS, a case study including the general Danish population was conducted.[64] The plasma concentrations of participants of both chemicals were evaluated, and the report revealed no correlation between the plasma concentrations and the risk of hepatic, pancreatic, bladder, or prostate cancer. Strong evidence of the carcinogenic effects of exposure to PFOA and PFOS is currently lacking and weak, despite research on the link between PFAS and cancer.

4.2 Immune system disorders

PFAS, in particular PFOS and PFOA, have been the subject of significant research on the immunotoxic effects on animals. It has been observed that these compounds modify both innate and adaptive immunity while also causing variations in inflammatory responses and cytokine production. Previous research on animals has demonstrated the immunotoxic effects of PFOA and PFOS,[65] prompting subsequent studies to investigate their impact on immunity-related disorders in humans.[66] Reports show the relationship between immunosuppression and PFAS exposure; the results are evaluated at the molecular and organ/system levels.[67] A two-fold increase in PFAS concentration was linked to a −49% (95% confidence interval [CI], −67% to −23%) decrease in serum antibody concentration for tetanus and diphtheria during the study of the impact of PFAS exposure on immunity in children.[68] The extent to which PFAS exposure affected the humoral immune response of the participating children may have varied depending on exposure levels, vaccine strains, and the time elapsed since immunization. Grandjean *et al.*[68] conducted a study on adolescents aged 7 to 13 years, which revealed a decrease in diphtheria antibody concentration in response to increased serum concentration of PFAS. The results indicated that doubling PFAS exposure at 7 years was linked to a decrease in diphtheria antibody level at 13 years.

4.3 Metabolic disorders

Studies on PFAS exposure and its effects on metabolism were conducted on a range of population groups, including adults, adolescents, children, pregnant women, and newborns. Clinical trials involving workers and individuals with obesity and diabetes have also been supported. Gilliland and Mandel[62] conducted one of the earliest studies in this regard, examining the relationship between PFOA serum levels and hepatic enzymes, lipoproteins, and cholesterol in workers exposed in the workplace. The predicted PFOA levels did not appear to be related to any major liver damage, even though the investigators measured the exposure using total serum fluorine. Studies on PFAS exposure and its effects on metabolism have been conducted on a range of population groups, including adults, adolescents, children, pregnant women, and newborns. Serum PFAS were positively correlated with glucose homeostasis in a cohort of adults and adolescents participating in the National Health and Nutrition Examination Survey (NHANES), indicating metabolic syndrome. The concentration of serum perfluorononanoic acid (PFNA) is primarily correlated with serum HDL-C and hyperglycemia (odds ratio 3.16). PFAS exposure has also been associated with increased blood insulin levels and β-cell activity, suggesting effects on glucose metabolism.[69] It was believed that the continuous exposure to the PFAS lead to the variation in the impact spectrum. The effects of PFOS and PFOA on humans have been extensively studied, and the literature reports the most persistent or long-term effects. In a community-based study of children in the C8 health project, increases in serum PFOA and PFOS between the first and fifth quintiles were linked to increases in total cholesterol of 4.6 mg dL^{-1} and 8.5 mg dL^{-1} and low-density lipoprotein cholesterol of 3.8 mg dL^{-1} and 5.8 mg dL^{-1}.[70] Similarly, among patients from NHANES 2003–2004, total cholesterol levels 9.8 mg dL^{-1} and 13.4 mg dL^{-1} were linked to an increase in PFOA and PFOS levels in serum from the lowest to highest quartiles.[71] Prenatal exposure is found to have effects on both mother and child later in life, according to studies conducted among a small cohort of pregnant women in the US.[72]

The chemical properties of different PFAS influence their biological effects, such as long half-lives in humans and other species, and their environmental persistence, which can be effectively irreversible. Adipose tissue could not support the bioaccumulation of PFAS.[73] Certain PFAS have structures that are similar to those of natural fatty acids.[74] According to Pérez *et al.*,[73] PFAS mostly accumulate in organs such as the brain, lungs, liver, serum, and kidney. The long-term exposure to human beings would increase the risk of cancer and has long-term impacts on immunological, endocrine, metabolic, andreproductive systems.[75] Research is being carried out to determine the significance of the data supporting all these interaction modifications through both outcomes and specific PFAS.[76]

PFAS exposure in pregnant women may lead to reproductive or pregnancy-related complications, such as birth of underweight infants,[77–80] high blood pressure,[81] preeclampsia,[82] increased time to pregnancy,[83,84] and gestational diabetes.[85]

According to a thorough assessment of the literature, 69 epidemiologic studies examined the connection between PFAS exposure and a variety of metabolic problems, including obesity, cardiovascular disease, and lipid homeostasis. According to Sunderland *et al.*[67] there is a positive relationship between blood lipids, such as triglycerides and total cholesterol, and PFAS exposure. The adverse effects of PFAS exposure on hypertension, stroke, diabetes, vascular disease, and insulin resistance were also examined by the authors through an analysis of epidemiologic research. Fig. 2 shows the negative health effects caused by PFAS ingestion *via* drinking water.

According to Boas *et al.*[86] PFAS impacts the thyroid through a variety of biological pathways related to thyroid homeostasis. These mechanisms include transport, metabolism, interference with thyroid receptors in target tissues, and thyroid hormone production. Possible mechanisms for thyroid disorder caused by exposure to perfluorocarbons (PFAS) include decreased circulating levels of thyroxine (T4) owing to the competitive binding of thyroid hormone transporter (THT) proteins,[87] elevated thyroid levels and hepatic T4 metabolism decreasing the thyroid fabrication of T4,[88] and decreased thyroid peroxidase (TPO) activity.[89] It was believed that the kidney primary affected organ by exposure of PFAS. This view was backed by epidemiologic data from an increasing number of individuals, animal studies, and *in vitro* models. Long-term kidney effects include kidney cancer, chronic kidney disease, and reduced kidney function over time.[90,91]

5. Determination techniques for PFAS in water

5.1 Ion mobility spectrometry (IMS)

IMS is a newly developed separation method that makes ion distinctions in the gas phase according to their size, shape, and charge state.[92] Whereas IMS separates ions based on variations in gas phase electrophoretic mobility, gas chromatography and liquid chromatography separate analytes based on changes in boiling point and polarity.[93] After ionization, inert gas (also known as the buffer gas) is introduced into a designated mobility separation area, where IMS separations take place.[94] An applied electric field causes ions to move through this region, and the length of time it takes for them to migrate through the gas (known as the drift time) is directly related to the size of the ions. The ion's collision cross section (CCS, usually expressed in units of Å^2) is a standard term used to characterize the measured size. Smaller ions migrate more quickly and encounter fewer collisions with the buffer gas in the IMS studies than bigger ions, whose migration is inhibited.[95] Because IMS separations take place in the gas phase, mobility analysis happens quickly, typically in less than 100 milliseconds per spectrum. Because of this, IMS may be easily integrated with current untargeted LC-MS/MS suspect screening techniques,[96–98] which allow for the addition of a molecular descriptor and additional separation mechanism for analyte characterization without requiring more time for analysis.[99] Furthermore, across a range of molecular classes, including carbohydrates, lipids, peptides, carbohydrates, and environmental pollutants like polyaromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs), prior IMS research has reported intrinsic connections between CCS and mass.[100–102] Based on the variations in gas phase packing efficiency (compactness) of the various chemical classes, distinct trendlines linking CCS to *m*/*z* have been found in all of the

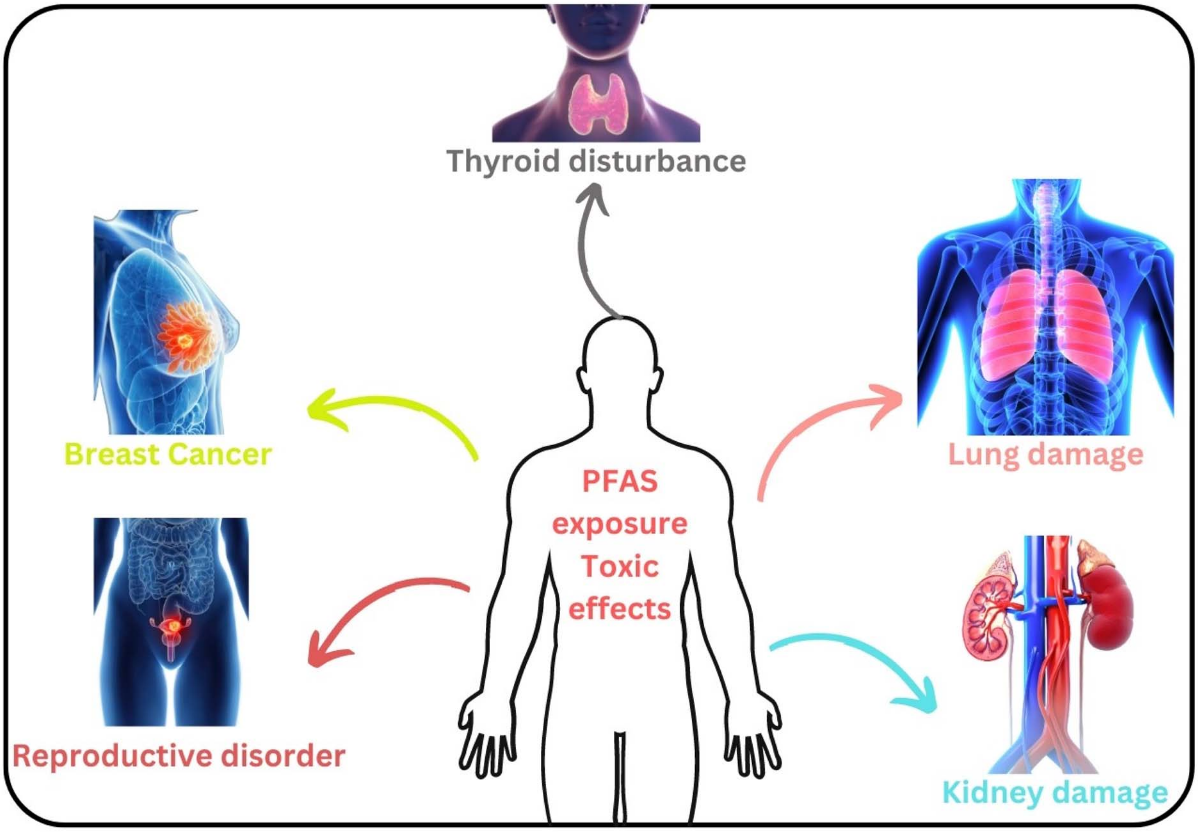


Fig. 2 Toxic effects caused by contamination by PFAS in aqueous solutions.

aforementioned situations. Hines *et al.*[103] reported that subclasses of vitamins, antibiotics, and other tiny biomolecules have unique mass-CCS trendlines. This information is helpful for untargeted research or natural product discovery, as it offers extra molecular details for unannotated properties.[104] IMS methodologies are increasing in popularity and are being used by several manufacturers for integrating IMS separations into untargeted MS processes owing to their precise selectivity and rapid actions.[105–109]

Yukioka *et al.*[110] carried out a study to determine an association between fragment ions and their molecule ions using the drift time obtained by ion mobility spectrometry. Standard solutions containing PFAS were examined using liquid chromatography/ion mobility quadrupole time-of-flight mass spectrometry (LC/IM-QTOF-MS) in order to validate the procedure. Even though co-eluting compounds were observed at similar retention times, all fragmentation flags for the PFAS were detected within a specified drift time range, and the molecular ion matched that of the PFAS in the spiked standard solution. Nine molecular ions were discovered when this technique was used on a domestic fire extinguisher liquid. As a result, the new linking technique used LC/IM-QTOF-MS to quickly search for potential molecular ions.

Luo *et al.*[111] investigated the application of coupled liquid chromatography, ion mobility spectrometry, and mass spectrometry (LC-IMS-MS) for the quick characterization of targeted as well as non-targeted PFAS in AFFF. Both the negative and positive ion modes of LC-IMS-MS were employed for the analysis of 10 formulations of AFFF from seven brands. Untargeted analysis of the formulations was followed by identification of PFAS-like features utilizing database matching, mass defect and homologous series evaluation, and MS/MS. The findings of this study revealed that 33 homologous series were identified in the tested AFFF formulation.

In contrast to data-independent acquisition, mass spectrometry has the potential to analyse target as well as non-target species. Nevertheless, it is taught to correlate the retention time with fragment ions of target precursor ions, due to the presence of co-eluting species in the environmental samples. As the conventional DIA approach is not sufficient to separate the ions, this technique, associated with ion mobility mass spectrometry, makes it easy to separate the target ion from the co-eluting ions by drift time. Initially, 32–96 PFASs were detected per sample during suspect screening using an internal database in groundwater samples impacted by firefighting foam ($n = 8$). It was observed that of all the pairs of respective precursor ions and fragment ions of the PFAS suspects, 5–19% (4–9 PFAS) of them were associated without considering the drift time of IMS, while 37–49% (15–43 PFAS) of them were associated with considering the drift time. The consideration of the drift time increased the association ratios in all samples.[112]

Mu *et al.*[113] also investigated the coupling ability of ion mobility spectrometry (IMS) with HRMS and machine learning techniques to accomplish the rapid and precise target and non-target screening of PFAS in wastewater. It was seen that only a few interfering peaks and a clearer spectrum were observed compared to traditional HRMS. Approximately 63% of misleadingly optimistic results were eliminated through the application of the collision cross section (CCS) technique in PFAS homologous series.

5.2 Liquid and gas chromatography

The analysis and determination of PFAS is frequently complicated due to inadequate information and a heterogeneous array of PFAS in the environment. Ultra-high performance liquid chromatography-quadrupole Orbitrap high-resolution mass spectrometry (UHPLC-Q-Orbitrap HRMS) was used to develop and validate a highly precise, accurate, and selective procedure for the qualitative and quantitative assessment of PFAS (24 perfluorocarbons and 30 precursors).[114] PFAS detection limits range from 0.6 to 5.4 ng L^{-1} (Shimadzu LC-MS-8060), with 84.13% recovery and a calibration range of 5–200 ng L^{-1} attained for 94% of the compounds examined, which includes all those specified in ASTM D7979.[115] With a total concentration ranging from 0.3 to 32.9 ng L^{-1}, 20 PFA, mostly PFBS, PFBA, PFOA, and PFPeA were successfully detected using the optimized approach in groundwater samples taken in China. Using 3-bromo-acetyl-7-methoxycoumarin (BrAMC), a fluorescence-based compound, HPLC with a UV detector was used to separate the long and short-chain PFAS in rodent liver samples.[116] The application of LC-MS/MS can help to check the presence or absence of PFOA, PFOS, and PFHxS in water.[117] Since these studies, a great deal of progress has been accomplished, including the introduction of various sample preparation methods, the application of various inner isotope guidelines (IS) for quantitative analysis, and tactics to increase accuracy while maintaining the best possible explanatory outcome. Although the collected samples are solid, they should all be dried, sieved, homogenized, extracted, and concentrated before being subjected to HPLC-MS analysis. It is necessary to employ a variety of pretreatment techniques to remove foundation interference and increase recovery. Bao *et al.* (2010)[118] found eight PFAS in river sediments using HPLC-MS/MS with negative electrospray ionization and a 1 : 5 ratio of sodium carbonate (Na_2CO_3) to tetrabutylammonium hydrogen sulfate for sample extraction.

In a separate study, Naile *et al.*[119] employed a method for recognizing multiple PFAS, and twelve PFAS were discovered in samples of soil and sediment collected from various sources. The material was homogenized, sonicated, and then HPLC-MS/MS analysis was carried out. However, because the solvents in some of the compounds were hydrophobic, the recoveries were only 20% or less. The detection of PFOS and PFOA in sewage sediment was carried out using a high sampling volume of methanol:acetic acid (9 : 1), followed by cleanup to extract the chemicals and analyze them using HPLC-MS/MS.[13] The samples were taken from landfills using passive sampling methods with sorbent-impregnated polyurethane foam (SIP) discs.[120]

According to Nakayama *et al.*[121] GC-MS is primarily used to identify PFAS that are present in the air as volatile, semivolatile, neutral, and nonionic compounds. These compounds include perfluoroalkane sulfonamides, fluorotelomer alcohols (FTOHs), and perfluoroalkane sulfonamide ethanols.[122] However, compared to liquid chromatography, this technique's restricted selection for PFAS measurement stems from its reliance on the type of analytes, which limits its application. Martin *et al.*[123] used GC with chemical ionization mass spectrometry (GC/CI-MS) for the analytical isolation and identification of PFOS precursors and fluorotelomer alcohols (FTOHs) present in ambient air. An analytical approach involving PFAS enrichment on glass-fiber filters (GFFs), polyurethane foams (PUFs), and XAD-2, followed by chromatographic measurement, was devised, optimized, and confirmed. Gas-chromatography electron impact mass spectrometry (GC-EIMS) has been used to analyze the concentration of perfluoroalkyl sulphonamides, such as *N*-methylperfluorooctanesulfonamidoethanol (MeFOSE), among others, to ascertain their sources, degree of occurrence, and partitioning in the air.[124] Alzaga *et al.*[125] established an analytical method for measuring perfluoro carboxylic acids (also known as PFC7-10A) in harbor sediments. Along with GC-MS-based determination, the process incorporated headspace solid-phase micro-extraction (SPME) and pressurized fluid extraction (PFE). PFCAs were found in the harbor sediments at low ppb quantities of 8 to 11 ng g^{-1} after the extraction parameters were optimized to produce recoveries of more than 95%, a limit of detection of 0.5–0.8 ng g^{-1}, and relative standard deviations from 15.5% to 16.8%. Fluorotelomer olefins (FTOs), fluorotelomer acrylates (FTAs), fluorotelomer alcohols (FTOHs), sulfonamides, and sulfonamidoethanol were among the perfluorinated compounds that were found to be most common in a spatial survey conducted throughout Asia as part of another study that used GC-MS analysis. Solvent extraction coupled with gas chromatography-positive chemical ionization mass spectrometry (SE-GC-PCIMS), a technique, was utilized to extract and analyze three neutral hydrophobic metabolic precursors of PFOS found in fish, fast food, and liver samples from Arctic marine mammals. Over 90% recoveries were achieved using this strategy. For the assessment and characterization of PFAS, a brand-new and effective analytical technique based on headspace solid-phase micro-extraction (HS-SPME) in conjunction with gas chromatography-atmospheric pressure photoionization high-resolution mass spectrometry (GC-APPI-HRMS) was presented. According to Ayala-Cabrera *et al.*[126] the approach demonstrated efficiency even at low concentration levels of PFAS, with low detection limits (0.02–15.2 ng L^{-1}), strong repeatability (RSD% < 11), and trueness (RE% < 12).

5.3 Electrochemical sensors

To date, several types of electrochemical sensors have been used to measure impedance (Z) over a range of frequencies (Hz). An ion-selective membrane measures the potential (mV), voltammetric sensors measure the change in current (pA), impedimetric sensors measure changes in the impedance (Z) over a range of frequencies (Hz), and conductometric sensors measure changes in the conductance (G, Ω^{-1}), which take place due to electrochemical reaction originating from an applied voltage (mV). Usually, electrochemical sensors such as voltammetric as well as potentiometric sensors are used for analysing PFAS. However, before the application of these electrodes, the surface should be functionalized so that they directly interrelate to the target analyte either by complexation or ion exchange. Molecularly imprinted polymers may be applied to surface functionalization, offering a polymeric matrix over the surface of the electrode with voids or detection sites, which correspond to the functional groups of the target analyte, shape, and size.[127]

Chen *et al.*[128] investigated potentiometric detection of PFO^- and PFOS in water, even for low concentrations ($\sim$0.07 μg L^{-1}), using ion-selective electrodes (ISEs), which were equipped with fluorous anion liquid exchange membranes. It was observed that the selectivity of ion-selective electrodes was affected by the occurrence of other perfluorinated compounds, which are only different in the number of carbon atoms. Karimian *et al.*[129] applied ferrocenecarboxylic acid (FcCOOH) to overcome the drawback (difficult detection of PFOS with poor electrical conductivity) and ferrocenecarboxylic acid (FcCOOH) works as an electroactive correspondent molecule contending with PFOS (non-electroactive) for molecularly imprinted polymers. The voltametric signal is reduced, and the association between the concentration of PFOS and the signal is developed if PFOS is in water. The findings indicate that in this case, the voltametric signal of the electroactive correspondent molecule (FcCOOH) is inversely proportional to the level of PFOS in the aqueous solution.

5.4 Biosensors

Currently, biosensors are predominantly utilized for the detection of PFAS in aqueous solutions. In this context, Zhang *et al.*[130] utilized electrochemical biosensors for the analysis of PFO that depends upon the inhibition of the biocatalysis process in enzymatic biofuel cells by the PFOS. Cennamo *et al.*[131] introduced a new method for biosensors by developing a configuration that involved a platform functionalized with a bio-receptor. This biosensor is characterized by a surface plasmon resonance (SPR) platform based on plastic optical fibers (POFs), together with a bio-receptor for the detection of PFOA and PFOS.

6. Remediation techniques

6.1 Chemical redox reactions

A DFT-based kinetic model was applied by Blotevogel *et al.*[132] to investigate the reductive defluorination of perfluorooctanoic acid through nanoscale zero-valent iron (nZVI) and zinc, and it was observed that the first step reductive PFOA defluorination has half-lives of about 8 years (Zn^0) and for Fe^0 about 500 000 years at metal-to-water ratios usually employed in permeable reactive barriers. These authors summarized that for the removal of PFOA from the water, the utilization of dehalogenation by zero-valent metals is not an optimal technique if appropriate catalysts are recognized to increase the rate of defluorination. Yin *et al.*[133] applied activated persulfate under acidic conditions and achieved PFOA degradation at approximately 89.9%. The findings indicate that the production of $SO_4^{-\bullet}$ played a prime role in the degradation of PFOA. Eqn (1)–(4) represent the prime mechanism behind the degradation of the PFOA. Both processes of HF removal as well as decarboxylation continue until the PFOA is changed into CO_2 and fluoride.

A specific type of CHP (catalyzed hydrogen peroxide propagation) called modified Fenton's reagent (MFR) has a long history of being used for ISCO of polluted soil and groundwater.[134] To start the propagation processes, MFR/CHP uses hydrogen peroxide and initiators like divalent minerals, soluble iron(II) or iron(III) chelates. A variety of reactive oxygen species and free radicals, such as the hydroxyl radical ($OH^{\bullet}$), perhydroxyl radical ($HO_2^{\bullet}$), superoxide radical anion ($O_2^{\bullet-}$), and hydroperoxide anion (HO_2^{-}), are produced by these catalyzed reactions in circumneutral pH systems. $O_2^{\bullet-}$ and HO^{2-} are both reductants and weak and strong nucleophiles, respectively:

$$\mathrm{Fe}^{3+} + \mathrm{H_2O_2} \rightarrow \mathrm{Fe}^{2+} + \mathrm{HO}_2^{\bullet} + \mathrm{H}^+ \tag{1}$$

$$\mathrm{OH}^{\bullet} + \mathrm{H_2O_2} \rightarrow \mathrm{HO}_2^{\bullet} + \mathrm{H_2O} \tag{2}$$

$$\mathrm{HO}_2^{\bullet} \leftrightarrow \mathrm{O_2}^{\bullet-} + \mathrm{H}^+ \quad (\mathrm{p}K_\mathrm{a} = 4.8) \tag{3}$$

$$\mathrm{HO}_2^{\bullet} + \mathrm{Fe}^{2+} \rightarrow \mathrm{Fe}^{3+} + \mathrm{HO_2}^{-} \tag{4}$$

In CHP systems, significant concentrations of superoxide radical anion are only seen at pH ranges close to 4.8 and higher (pK_a in reaction (2)). To investigate the oxidative treatment of PFOA, Mitchell *et al.*[135] used CHP reactors with neutral pH and hydrogen peroxide concentrations ranging from 0.25 to 2 moles per liter (M). In comparison to PFOA loss of 15% in a control sample, PFOA concentrations (baseline 100 μg L^{-1}) decreased to 68, 85, and 89% after 150 minutes at starting H_2O_2 concentrations of 0.25, 0.5, and 1 M, respectively. These experiments showed that MFR/CHP can quickly decrease PFOA concentrations. Heat-activated persulfate oxidation of per- and polyfluoroalkyl (PFOA) was further explored for groundwater cleanup by Park *et al.*[136] and Bruton and Sedlak.[137] For example, the kinetics of PFOA degradation increased five times at 50 °C for every five times increase in $S_2O_8^{2-}$.

6.2 Membrane filtration

The developed membrane technology can be easily customized to specific needs, which makes it a good candidate for the treatment of aqueous solutions. This method is typically applied for the separation and determination of the level of PFAS.[138] This is a superior method for the remediation of PFAS (with very high removal efficiency <99%) that can discard highly loaded PFAS when the concentration of the PFAS is very high 1000 mg L^{-1}.[139–145] Size exclusion is the predominant mechanism responsible for the elimination of the PFAS *via* the membrane process. Furthermore, electrostatic repulsion, diffusion, and cake layer filtration are the other mechanisms responsible for the removal of PFAS. Nanofiltration and reverse osmosis membranes are types of membranes that perform well in the rejection of PFAS. Nanofiltration is economical and more suitable for the remediation of PFAS as compared to reverse osmosis. NF270 nanofiltration can eliminate up to 99% of PFAS; however, some PFAS compounds, such as short-chain PFAS, have molecular masses below 270 Da and may not be fully removed.[141] NF270 might eliminate 96.6–99.4% PFAS for specific compounds like PFHxA.[140] The concentration of the PFHxA in the retentate was up to 870 mg L^{-1}. The shortcoming of the membrane method is that the concentration has to deal with a destructive method, such as AOP (advanced oxidation process).[140] In this situation, electro-oxidation is used and decreases PFHxA to 8–21 mg L^{-1}. Appleman *et al.*[42] found in their investigation that NF270 might be applied for the treatment of highly saline groundwater. Furthermore, Boonya-chart *et al.*[146] applied nanofiltration for eliminating PFOA from groundwater and showed 99.49 to 99.54% rejection of PFOA. The membrane can eliminate both neutral soluble and charged organic compounds having a molecular weight less than 200 Da. Therefore, nanofiltration could eliminate PFOA with a molecular weight of 414 g mol^{-1}. The membrane approach is an efficient method for the remediation of PFAS from aqueous solutions, although membrane fouling and low water levels would eventually reduce the work of the membrane. This will happen due to the presence of inorganic ions as well as humic acid in the groundwater, which leads to a decrease in the rejection efficiency of the membrane.[147] The PFAS rejection efficiency of the membrane can be enhanced by high-pressure PFOA concentration, but this will increase the operational cost.[146,148,149]

The membrane approach is highly developed and easily customizable, expanding its applicability and versatility across a range of applications, including wastewater treatment and desalination.[150,151] Boonya-atichart *et al.*[146] found a 99.49 to 99.54% removal efficiency of PFOA in groundwater using a nanofiltration (NF) membrane. The membrane can discard soluble neutral and charged organic molecules with molecular weights greater than 200 Da because of the pore size constraint. Therefore, PFOA with a molecular weight of 414 g mol^{-1} can be eliminated by NF. Furthermore, a study by Tang *et al.*[152] found that NF membranes may reject up to 90–99% PFOS. Fig. 3 shows an illustration of efficient techniques used for the remediation of PFAS from aqueous solutions.

6.3 Adsorption

6.3.1. Activated carbon. Activated carbon is emerging as an effective adsorbent for the elimination of pollutants from aqueous solutions due to its high sorption capacity as well as low cost.[153–157] Geometrical (for variation in size and shape of pores) and chemical heterogeneity (for different functional groups) are used for the characterization of activated carbon.[158] Several studies reported the use of granular activated carbon for the remediation of the PFAS from aqueous solutions *via* adsorption processes, including applications at large scales. Due to a large size, volume, and surface area, activated carbon has shown higher adsorption capacity.[159–162] Granular activated carbon successfully eliminated long-chain PFAS through hydrophobic interactions, although it did not eliminate short-chain PFAS successfully (achieving equilibrium in a shorter time). Powder-activated carbon is effectively used for the

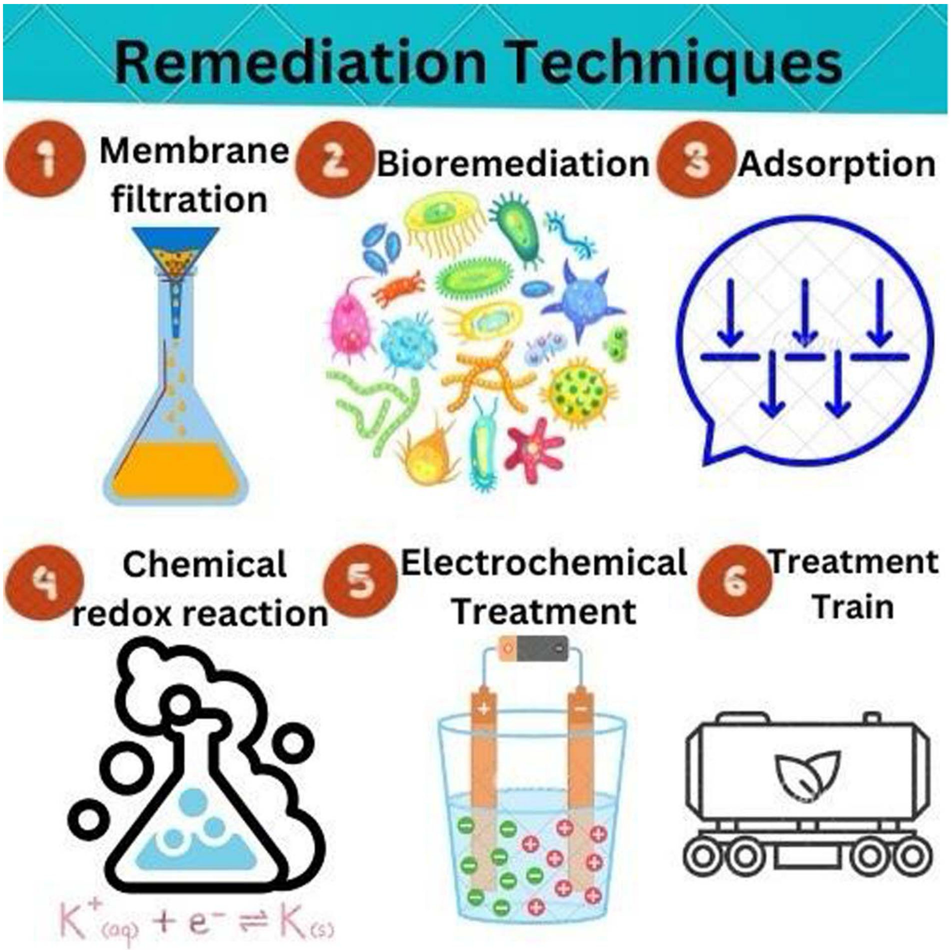


Fig. 3 Techniques used for the remediation of PFAS from aqueous solutions.

elimination of short-chain PFAS owing to its small size in comparison with granular activated carbon.[160]

Powdered activated carbon shows a comparatively higher adsorption capacity for PFOA as well as PFOS than granular activated carbon.[160] Du *et al.*[161] investigated the adsorption behavior of bamboo-derived activated carbon for PFOA, and the findings indicate that an equilibrium was reached in 34 h (a long time). Table 2 shows the efficiency of various adsorbents used for the removal of PFAS from water.

6.3.2. **Biochar.** Biochar is a porous, stable, carbonaceous, and environmentally benign substance. The main raw material used to make biochar is the abundant, cost-effective, and easily available biowastes from the food and agriculture sectors.[168,169] Biochar possesses various functional groups, primarily heterocyclic and aromatic, such as hydroxyl and carbonyl, which enhance PFAS adsorption.[170,171] PFAS removal with biochar is mostly achieved through the effects of hydrophobicity and electrostatic interactions.[164] It has been demonstrated that biochar is useful for eliminating heavy metals and pharmaceuticals from water.[172,173] Furthermore, its remarkable physical characteristics, such as pore size and specific surface area, can be tuned to meet the unique requirements for the removal of short-chain PFAS.

Patel *et al.*[164] fabricated an affordable biochar for the removal of PFAS from wastewater. The biochar was obtained from the anaerobic digestion of organic pollutants in the wastewater treatment process. Anaerobic digestion of organic pollutants produces biogas, which is subsequently used to sequester carbon *via* catalytic breakdown to produce hydrogen and carbon nanomaterials (CNMs), and then the biochar is loaded with carbon nanomaterials, and recognized as CNM-laden biochar. CNM-laden biochar showed a superior removal efficiency for PFAS (79%) as compared to biochar modified with ilmenite (54%). In order to remove PFAS, biochar was prepared by pyrolyzing hardwood and softwood shavings at different temperatures of 300, 400, and 600 °C. These biochars were post-modified *via* thermal oxidation for 30 minutes at 400 °C, which increased the pore size and surface area. After PFAS were adsorbed on biochar, heat activation at 500 °C with nitrogen or air destroys them. After heat activation, the biochar exhibits improved PFAS elimination efficiency in subsequent cycles.[174] In addition, biochar was obtained from the hardwood and softwood shavings at various pyrolysis temperatures from 300 to 600 °C. The material undergoes hot air oxidation after pyrolysis, which increases the material's surface area and pore size, leading to improvement in its absorption capacity. Compared to untreated biochar, this procedure triples the reactivity of

Table 2 Summary of the adsorption efficiency of various adsorbents for the removal of PFAS

Adsorbent	Type of PFAS	Adsorbent dosage	Initial concentration	Removal efficiency	References
DUT-5-2	PFOA	5 mg	30 mg L^{-1}	60.9	Hu *et al.*[163]
Carbon nanomaterial-coated biochar (CNM-biochar)	PFOS	1 g	310 μg L^{-1}	95	Patel *et al.*[164]
Sewage sludge-activated carbon ($ZnCl_2$-impregnated)	PFBA	5 mg	50 μg L^{-1}	87	Mohamed *et al.*[165]
Biochar-alginate composite beads	PFOS	1.5 g L^{-1}	—	99	Militao *et al.*[166]
Classical powder-activated carbon	PFOS	0.004 g	2 ppm	93	Pala *et al.*[167]
MIL-101 (Cr)@AC MOF	PFOS	100 ppm	2 ppm	80	

biochar derived from softwood for certain PFAS. The materials were subjected to a solution comprising PFCAs, PFSAs, and GenX. They were then allowed to dry, and the PFAS were removed by heating the dried materials for 30 min at 500 °C in air. Following treatment, additional sorption tests were conducted on the biochars and granular activated carbon (GAC), using a mixture of PFBA, PFPeA, GenX, PFHpA, and PFOA.

Krahn *et al.*[175] prepared a biochar from sewage sludge and wood chips by pyrolyzing at 700 °C for the elimination of long-chain PFAS-like PFOA, PFNA, and PFDA, and short-chain PFAS-like PFPeA, PFHxA, and PFHpA. All of the biochar's adsorption onto sewage sludge and wood chip biochar, except PFPeA and PFHxA, followed a strong log-linear relationship ($r^2 > 0.9$). These poor fits may be explained by the fact that ionic interactions are more significant for the adsorption of these short-chain PFCAs compared to hydrophobic ones for the adsorption of the longer-chain PFCAs.

In other studies, leftover coffee grounds were used to prepare biochar *via* two-step pyrolysis, followed by post-treatment to enhance its properties. In order to functionalize the biochar surface with nitrogen-containing groups such that radical-initiated polymerization may occur, two distinct methods were looked into. The first method was electrophilic aromatic substitution, which involved reducing the nitro group to $–NH_x$ with acetic acid, NH_4OH, and $Na_2S_2O_4$, then combining precisely measured amounts of pristine biochar with HNO_3 and H_2SO_4. In the second method, the biochar–water mixture was combined with melamine (BC–M) or ammonium chloride (BC–N), and the nitrogen modification was initiated by heating the mixture. Using thermally accelerated radical-initiated polymerization, a layer of molecularly imprinted polymers (MIP) was applied to the surface of nitrogen-modified biochar to increase the adsorbent's selectivity for PFAS. In comparison to unmodified biochar, the BC–M–MIP showed higher K-selectivity for PFBS (4.52) and PFOA (3.76) at 0.043 and 0.039 mg PFAA/g × g m^{-2}, respectively.[176] In a different experiment, different PFAS chain lengths (4 to 11 fluorinated carbons) were eliminated using eucalyptus charcoal and tree bark. The fact that the sorption increased with the increase in chain length indicates that hydrophobic interactions play a key role in the adsorption process.[177] To create biochar, Wu *et al.*[178] employed a variety of feedstocks, including switchgrass (SG), water oak leaves (WO), and biosolids (BS), and altered them by adding carbon nanotubes (CNTs) and $FeCl_3$ to increase their porosity and surface area. The composite made of oak leaves and biochar rapidly attained the adsorption equilibrium. In comparison to the other two biochars, the modified biosolid–biochars showed increased PFOA adsorption. BS–Fe demonstrated the highest adsorption capacity for PFOA (469.65 μmol g^{-1}), while WO–CNT demonstrated the lowest adsorption potential of 39.54 μmol g^{-1}.

In a different study, biochar was made from Douglas fir by gasification at 900–1000 °C, with syngas as a byproduct. Iron(III) chloride (18 g) and iron(II) sulfate (36.6 g) were added to a solution (2 L) containing biochar (50 g) to change the biochar. Fe_3O_4 particles were then created and deposited onto biochar following a 24-hour period during which the pH was maintained at 10, and a gentle introduction of 10 M NaOH was carried out. The Fe_2O_3-modified biochar was ready following the steps of vacuum filtration, three ethanol washes, and 50 °C drying. In comparison to PFOS (82%), the Fe_2O_3-modified biochar eliminated 75–85% of PFBS, GenX, and perfluorooctane sulfonamide (PFOS).[179] When compared to pristine biochar, modified biochar showed enhanced adsorption potentials for both short- and long-chain PFAS. The main processes of biochar-mediated PFAS elimination include hydrophobic and electrostatic attraction. Since short-chain PFAS are not often removed, more research is required to find sustainable removal strategies that target short-chain PFAS in particular during wastewater treatment.

6.3.3. Miscellaneous adsorbents. Current research focuses on exploring innovative adsorbents with lower cost and higher adsorption capacity. Some novel materials, such as cross-linked chitosan beads, effectively removed PFOS from aqueous solution at pH 3.[180] At pH 3, amino groups (99.4%) on the chitosan beads were protonated, and consequently, PFOS, carrying a negative charge, is adsorbed easily, which leads to enhanced adsorption capacity. At an equilibrium concentration of 0.33 mmol L^{-1}, the maximum adsorption capacity for chitosan biosorbent reached 5.5 mmol g^{-1} for PFOS, and it was greater than for traditional absorbents. Chen *et al.*[181] reported other cost-effective adsorbents including maize straw-origin ash, straw- and willow-derived chars, and carbon nanotubes. Single-walled carbon nanotubes and maize ash showed greater adsorption capacity for PFOS at ~700 mg g^{-1}. This may be due to the formation of hemimicelles and hydrophobic interactions preferring the binding of PFOS. Carbon nanotubes can serve as effective adsorbents for the adsorption of PFAS because of the greater adsorption capacity as well as equilibrium reached in a short time; nevertheless, ash attained equilibrium within 48 h along with an adsorption capacity similar to carbon nanotubes.

Furthermore, the cost of the ash is relatively low, and it is obtained easily *via* incineration of biomass; therefore, its utilization is a favorable option for other biomaterials, along with high recovery and reuse of agro-waste.[182,183] The regeneration of a cross-linked adsorbent was found to be quick and was achieved in merely five minutes without any significant decrease in the adsorption capacity for PFOS in the presence of other organic pollutants.

Adsorption is a well-developed method for the remediation of PFAS from an aqueous solution, but it suffers from some limitations. Generally, adsorption studies are focused on the remediation of specific PFAS, whereas actual aqueous solutions contain a mixture of PFAS with varying chain lengths and functional groups.[184,185] Also, further research is necessary to understand the interference of inorganic ions and organic substances. This inexorably restricts the adsorption capacity, which is important for the regeneration of the adsorbents after adsorption saturation; regenerated adsorbents demonstrate lower reutilization capacity when the concentration of PFAS in the aqueous solution is relatively low.[186]

6.4 Electrochemical approach

Certain electrochemical approaches, such as electrosorption, electrochemical oxidation, and electrocoagulation, are used in various applications.[187–191] The electrocoagulation approach is comparatively attractive due to its low capital expenditure and simple operation. The results obtained from Niu *et al.*[187] revealed the use of zinc as an anode and stainless steel as a cathode for the remediation of a PFOA aqueous solution at an initial concentration of 200 mg L^{-1}. The maximum removal efficiency reached 99.7%. The surface of the electrode becomes passive if a single electrode is used for a prolonged time. For the remediation of PFOA, Liu *et al.*[192] applied periodically reversing electrocoagulation (PREC) with an Al–Zn electrode. The mechanism behind the sorption was the hydrophobic interaction between PFOA and Al–Zn hydroxide flocs. The direct oxidation of the anode and the indirect oxidation of active radicals are the main mechanisms behind the electrochemical oxidation approach for the treatment of PFAS. Oxygen evolution potential (OEP), the electron transfer ability of the anode material, and the potential to produce $^{\bullet}$OH are the factors affecting the removal efficiency.[193,194] Schaefer *et al.*[188] observed that PFOA and PFOS are efficiently oxidized *via* boron-doped diamond anodes (electrochemical method), and the dominant mechanism is direct anodic oxidation. Although various studies suggested that the generation of $^{\bullet}$OH through the anode could not break the PFOA, the rate-limiting step for PFOA breakdown was the electron transfer at the anode surface.[195,196]

6.5 Treatment-train techniques

The goal of SERDP Project ER-2423 was to provide an economical *in situ* technique for treating groundwater that contains per- and polyfluoroalkyl compounds (PFAS). Horizontal reactive media wells were utilised in the project, also known as "*in situ* chemical oxidation of sorbed contaminants (ISCO-SC)", to remediate contaminated groundwater on-site. This technique is particularly helpful for locations affected by aqueous film-forming foam (AFFF). In treatment-train techniques, there is a combination of *in situ* and *ex situ* remediation techniques, which results in efficient remediation techniques as compared to any single method. Crimi,[197] in SERDP project 2423, integrates two potential remediation technologies: sorption *via* GAC and PFAS destruction using activated persulfate.[192] It was believed that the technology gives information on the regeneration of carbon, the effectiveness of persulfate treatment, and degradation products. Further SERDP ER-2426 technology was used for the investigation of reductive or oxidative methods applying [0] valent metals and bimetals (Pd/Fe, Mg, Pd/Mg) and synthesis using clay and vitamin B12-assisted co-solvents.[198,199]

6.6 Bioremediation

For many years, it was suggested that biotechnology is not an efficient technique for the elimination of PFAS. It was difficult for the microorganisms to utilize PFAS as their sole carbon source for cell growth. The cleavage of the C–F bond requires more energy, which is greater than the capacity of microorganisms. Currently, some bacterial strains under anammox and anaerobic conditions have appeared to be feasible to transform as well as degrade PFAS.[196,200,201] Anammox is a type of reaction in which ammonium gets oxidized in the absence of oxygen into dinitrogen gas using nitrite as the electron acceptor.[202,203] Early research indicated that the bioremediation of PFAS is not feasible because of the strong C–F bonds; however, a recent study[202] has shown that the biodegradation of PFAS is possible. Fluorotelomeric structures having CH_2 groups on the C–F backbone have shown sequential microbial defluorination instead of perfluorinated substances. Fluorotelomeric alcohols consisting of carboxylic and sulfonic acids were moderately biotransformed into perfluoroalkyl acids under aerobic as well as anaerobic environments.

The breakdown of fluorinated alkyl substances needs a minimum of one H atom in the alkyl chain for the place of the prime attack.[2] The main challenge in the oxidative replacement of fluorine atoms is the potential formation of a highly hydrophobic layer near the carbon–carbon bonds, which hinders the oxidative degradation. Due to this feature, the fluorine-saturated carbon chain element restricted the oxidation *via* the employment of microorganisms as sources of carbon as well as energy.[204] Only a few studies have been carried out on the microbial degradation of PFAS.[205,206] There are several contradicting reports, and every report needs further research work to understand the biotic transformations of these substances.[207] Various strains of bacteria were applied for the breakdown of the PFAS in an aerobic environment. Sulfonates containing a terminal hydrogen, such as H-PFOS and 2,2,2-trifluoroethane, can be partially degraded by the Pseudomonas strain D2 through defluorination under aerobic, sulfur-limited conditions.[208] Six products formed during the defluorination reaction carried only oxygen and fluorine, without sulfur. *Pseudomonas butanova* was applied for the breakdown of a 6–2 fluorotelomer alcohol precursor and this strain is not effective for the degradation of 8–2 FTOH.[209] Additionally, studies on the breakdown of PFAS using fungal species are ongoing,[210] because of a broad spectrum of substrate reduction catalyzed by extracellular ligninolytic enzymes. Under aerobic conditions, white-rot fungus *Phanerochaete chrysosporium* can reduce the 45% concentration of 6 : 2 FTOH within 35 days, along with the generation of various shorter-chain metabolites, for example, perfluorobutyric acid (PFBA) and perfluoropentanoic acid (PFPeA).[197,211] There are various approaches that have been applied in the bioremediation of PFAS from aqueous solutions, as shown in Fig. 4.

6.6.1. Phytoremediation. Phytoremediation is the most promising approach for the extraction of toxic pollutants from the environment by employing chosen plants. In the current scenario, the utilization of plants for the removal of pollutants from water becomes the optimized alternative. Phytoremediation comprises different types, as depicted in Fig. 4.[212]

The absorption of the pollutant through the root is known as phytoextraction. Phytostabilization is defined as the incorporation of the organic or inorganic substance into the lignin. In phytovolatilization, the hazardous substance is converted into the least toxic form by absorption of the pollutants through the roots. In phytodegradation, the breakdown of toxic substances is feasible by means of specific enzyme activity. In the rhizofiltration process, terrestrial plants are used for the absorption of different types of pollutants from water or soil through their roots.[213]

There are typically four steps involved in the elimination of PFAS from the environment, which are phyto uptake, translocation, bioaccumulation and biodegradation. Biodegradation in plants was previously recognized through the "plant detoxification mechanisms" of transformation, conjugation, and compartmentalization, which were also recognized as the "green liver model".[214] It has also been observed that the elimination of PFAS from wetlands was possible due to three major mechanisms: phytouptake, bioaccumulation, and translocation.[215,216] The phytouptake of PFAS in plants occurs through two pathways; one is passive and the second is active,[217] as shown in Fig. 5.

Anthracene-9-carboxylic acid, 4,4′-diisothiocyanostibene-2,2′-disolfonate, and 5-nitro 2-(3-phenylpropylamine) benzoic acid are examples of anion channel blockers, and metabolic inhibitors (NaN_3 and Na_3VO_4), and aquaporin competitors ($AgNO_3$ and glycerol) are examples of inhibitors that can be used to measure the active and passive transport of PFAS.[219,220] In maize (*Zea mays* L. cv. TY2), metabolic pathways contributed 43–83% and zero, respectively, to the transfer of PFOA and PFOS.[219] As a result, although PFOS uptake may be somewhat passive, PFOA uptake may be partially active. Nonetheless, wheat (*Triticum aestivum* L.) reduced its uptake of PFOA and PFOS by 58–87% and 64–94%, respectively, suggesting that both PFOA and PFOS are mostly transported in wheat.[220] If all factors are considered, then the transport mechanism of PFOS in plants may vary by species. Furthermore, PFAS transport in plants may be facilitated by aquaporin and anion channels.[219]

The mechanism behind the biodegradation of PFAS by plants as well as microorganisms is still not clear. There are various parameters, such as physicochemical properties, ionic strength, host plant species, exposure time, pH, chain length, and initial concentration, which affect the extraction process of PFAS from the environment.[215,221]

Phytofiltration is the main procedure by which aquatic plants eliminate PFAS.[222] The phytofiltration of PFAS into the aqueous phase mostly occurs in plant roots and submerged organs by the adsorption and precipitation mechanisms.[223] Based on diverse mechanisms of uptake, aquatic plants (free-

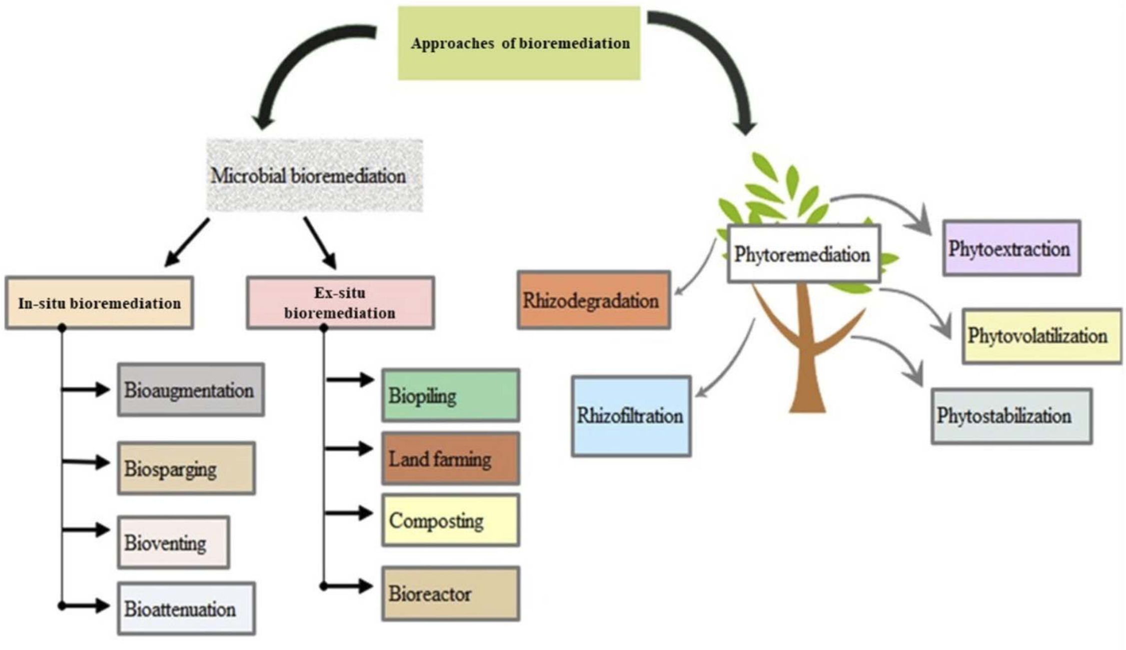


Fig. 4 Different approaches applied for the bioremediation of PFAS.

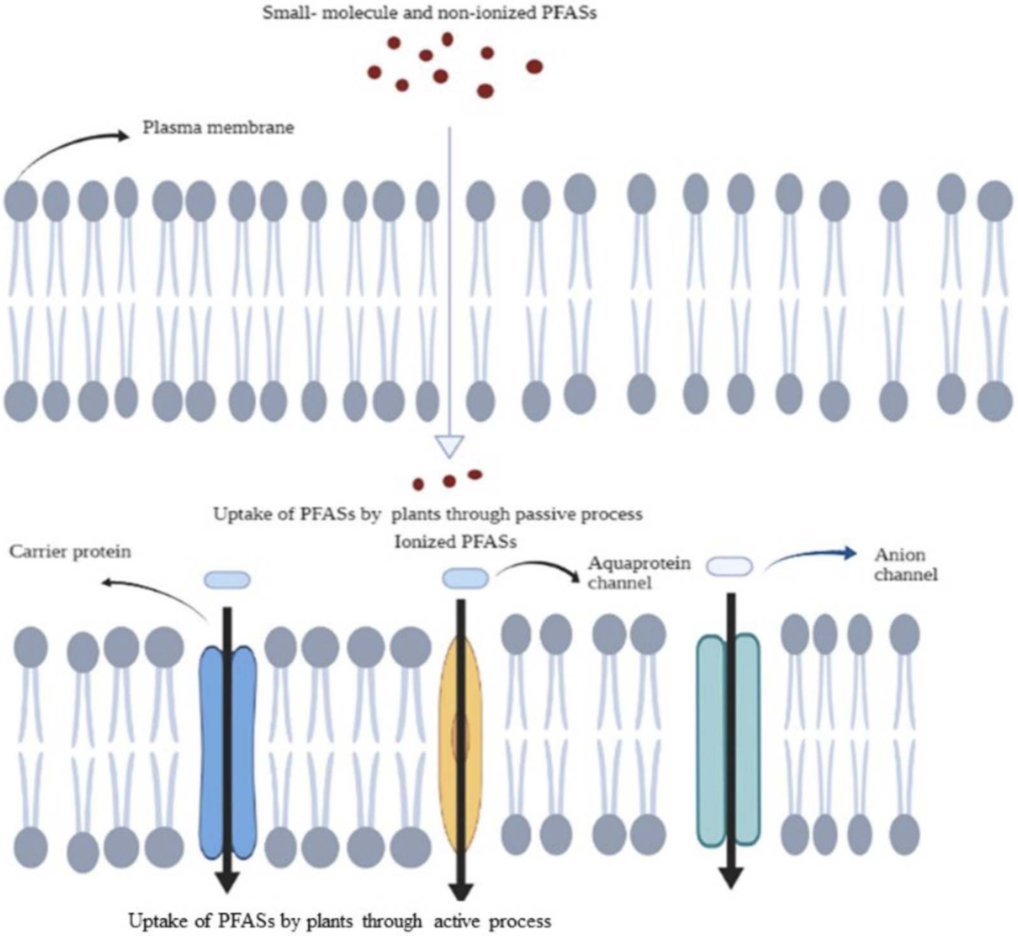


Fig. 5 Uptake of PFAS *via* plants through (a) a passive process and (b) an active process [modified from Wang *et al.*[218] with permission from Elsevier (ref. 218) (license number: 6157540752757 dated 28th November 2025)].

floating plants as well as submersed species) would be able to attract pollutants through the means of precipitation as well as adsorption. On the other hand, various terrestrial plants can eliminate pollutants *via* root uptake and bioaccumulation. Previous research suggests that submerged wetland plants with higher growth of root systems could attain higher uptake of PFOs.[224] The results of a study on *Echinodorus horemanii* (submerged plants) and *Eichhornia crassipes* (free-floating plants) found that the *E. horemanii* exhibits faster uptake of PFAS compared to *E. crassipes*, which may be due to its greater biosorption as well as leaf-water exchange capacities.[225] Consequently, the findings suggest that submerged plants exhibit greater uptake of PFAS compared to free-floating plants owing to their well-developed root systems as well as leaf-water exchange capacity in an aqueous medium.[225,226]

The prime mechanisms behind the elimination of organic contaminants are phytodegradation, phytofiltration, and phytovolatilization.[227–229] Moreover, it is important to note that PFAS do not undergo biodegradation, chemical disintegration or photolysis.[217] As the biodegradation of PFAS is restricted, the best alternative is plant uptake at the remediation site of the PFAS.[198] Past researchers indicate that the uptake mechanism of PFAS from the soil or water typically occurs *via* the plant's roots by means of passive and active processes, as shown in Fig. 5.[218,230] The main passive transportation depends on the diffusion of small-molecular and non-ionized PFAS as well as the driving force of the transpiration stream.[231] Once the PFAS goes into the root apoplast through diffusion, it is translocated to the part of the plant that is above the ground, for example, fruits, stems, leaves, and shoots, by the transpiration stream.[232] Meanwhile, active transportation largely involves selective adsorption through particular transporters such as ion channels and aquaporins for the transportation of ionized PFAS into plant cells.[219] However, airborne PFAS and analogues in the vapor phase and particulates could be adsorbed in plants through aerial parts, for example, foliage and bark.[233]

Yin *et al.*[234] investigated the removal efficiency for constructed wetlands of PFAS. In this study, the authors used eleven PFAS, out of which seven PFCAs were perfluoroalkyl carboxylic acids and perfluoroalkane sulfonates along with 7 PFAA precursors. The level of the PFAS changes from 1269 to 7661 mg L^{-1}. Throughout the year (sampling time), the composition of the PFAS remains the same in comparison with the PFCAs. It was believed that the CW treatment could eliminate approximately 61% of the total PFAS, along with 50–96% for individual PFAS. The reed bed shows a high efficiency from 42 to 49%.

Zhang and Liang[221] conducted an experiment to determine the efficiency of duckweed for the removal of PFAS from an aqueous solution. It was observed that removal of perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorobutanesulfonic acid (PFBS), and perfluorohexanoic acid (PFHxA) was marginal and much lower than those of perfluoroheptanoic acid (PFHpA), perfluorohexanesulfonic acid (PFHxS), perfluorooctanoic acid (PFOA), and perfluorooctanesulfonic acid (PFOS). The duckweed showed about 80% removal efficiency for PFOA and PFOS at pH 2.3 at an initial concentration of 0.200 mg L^{-1}.

7. Challenges for the remediation of PFAS from aqueous solutions and conclusion

PFAS remediation from water is also very challenging, and the advantages and disadvantages of the methods are discussed in the following. The techniques used for the remediation of PFAS from aqueous solutions are not efficient due to the special molecular structures and physicochemical properties of PFAS.[197,220] Furthermore, the bioremediation of PFAS requires further research to be successfully applied for effective PFAS degradation. Additionally, the choice of an appropriate adsorbent for the adsorption of PFAS is also a big challenge. The removal of short-chain PFAS is more challenging compared to that of long-chain PFAS. Innovative remediation approaches need specific conditions, more chemicals, materials, and high energy, and are expensive.[141,235,236]

Poly- and perfluoroalkyl substances are large groups of synthetic chemicals that are persistent and environmentally stable. These chemicals have a wide range of industrial applications, and with the rapid escalation in PFAS usage, their concentration in aqueous solutions has concurrently increased. These chemical substances are toxic and can seriously threaten human and animal health. This review discusses the sources of PFAS in aqueous solutions and their negative impacts on humans. Furthermore, various remediation techniques such as electrochemical treatment, adsorption, bioremediation, and membrane technology have been discussed critically, along with a special focus on the phytoremediation of PFAS. However, the phytoremediation of PFAS still needs to be extensively explored, because the survival rate of the plants is relatively low. Future research should focus on developing advanced, cost-effective, and eco-friendly remediation technologies for PFAS removal from aqueous media, with improved efficiency.

Conflicts of interest

The authors declare that they do not have any personal or financial conflicts of interest.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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