

REVIEW & ANALYSIS

Special Section: PFAS in Agroecosystems—Sources, Impacts, and Opportunities for Mitigating Risks to Human and Ecological Health

Current trends and patterns of PFAS in agroecosystems and environment: A review

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Abstract

Per- and polyfluoroalkyl substances (PFASs) are one of the more well-known highly persistent organic pollutants with potential risks to agroecological systems. These compounds are of global concern due to their persistence and mobility, and they often lead to serious impacts on environmental, agricultural, and human health. In the past 20 years, the number of science publications on PFAS has risen; despite this, certain fundamental questions about PFAS occurrence, sources, mechanism of transport, and impacts on agroecosystems and the societies dependent on them are still open and evolving. There is a lack of systematic and comprehensive analysis of these concerns in agroecosystems. Therefore, we reviewed the current literature on PFAS with a focus on agroecosystems; our review suggests that PFASs are nearly ubiquitous in agricultural systems. We found the current research has limitations in analyzing PFAS in complex matrices because of their small size, distribution, and persistence within various environmental systems. There is consistency in the properties and composition of PFAS in and around agroecosystems, suggesting evidence of shared sources and similar components within different tropic levels. The introduction of new and varied sources of PFAS appear to be growing, adding to their residual accumulation in environmental matrices and leading to possible new types of chemical compounds that are difficult to assess accurately. This review determines existing research trends, understands mechanisms and incidence of PFAS within agroecosystems and their impact on human health, and thereby recommends further studies to remedy research gaps.

Plain Language Summary

This is a review designed to provide a basis in understanding of the science and context surrounding PFAS within agricultural systems. In addition to providing a background, our review also expands on gaps in the research and holds discussion

Abbreviations: AFFF, aqueous film forming foam; BCF, bioconcentration factor; EK, electrokinetic; FTOH, fluorotelomer alcohol; HFPO-DA, hexafluoropropylene oxide dimer acid; PFAS, per- and polyfluoroalkyl substance; PFBS, perfluorobutanesulfonic acid; PFCA, perfluorocarboxylic acid; PFHxA, perfluorohexanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid; VOC, volatile organic compounds.

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about some of the pitfalls preventing remediation from happening at a large scale. We touch briefly on the health effects of PFAS and point to governmental efforts around the globe to prevent further infiltration of PFAS into agricultural ecosystems.

1 | INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) are a group of manufactured chemicals, which have gained considerable attention in recent years in both scientific spaces and in news media around the world. The diverse applications of these chemicals contributed both to their industrial success and widespread usage. PFAS can be characterized by two main attributes: their high thermal stability, and their remarkable chemical stability—both of which are due to carbon–fluorine bonding throughout the molecule. Carbon–fluorine bonds are extremely resilient due to their high bond energy. This property is the result of the extraordinary stability of the carbon–fluorine bond (485 kJ/mol), which, for example, is significantly higher than the bond energy of the carbon–oxygen single bond (358 kJ/mol) (Brunn et al., 2023). The $[-CF_3(CF_2)_n]$ chain is the defining feature of PFAS. The C–F bond is relatively short (1.35 Å compared to ~1.8 for the C–Cl bond), meaning it has low polarizability. The combination of both low polarizability and multiple opposing dipoles ensures that perfluorinated carbon chains participate in few, weak intermolecular interactions which in turn creates compounds that are water, oil, and dirt repellant. GenX shown in Figure 1 and other short-chain PFAS are of particular concern with respect to agroecosystems because of their extreme mobility and their tendency to concentrate in the edible parts of plants (Brendel et al., 2018). While not exclusive to short-chain PFAS, the tendency to concentrate in areas of concern combined with their extreme mobility points to a need for a better toxicological understanding of the molecules. A 2020 paper claimed that PFASs are ubiquitously distributed across all sections of industry and consumer products (Glüge et al., 2020). Thousands of structural variants of PFASs have been in use since the 1940s and a number of these chemicals are often accompanied by a host of health impacts including cancers, reproductive defects, developmental delays in children, increased risks of immunodeficiencies, and hormonal disruptions (Oulhote et al., 2016; US EPA, 2020). Many contributing factors lead to PFAS becoming the danger they are today, chief among them are their durability and persistence, propensity for movement in and throughout the environment, and wide-ranging distribution. As our understanding of the dangers of PFAS increases, one particular area of concern with regards to contamination is within agroecosystems.

Agroecosystems, in this paper's context, can be defined as any ecosystem built or utilized for agriculture and for growing crops with addition of supplemental nutrients in the form

of fertilizers or other soil amendments. Agroecosystems have high potential to act as sinks for some PFASs via bioaccumulation (Death et al., 2021). Examples of agroecosystems include, but are not limited to, polyculture systems, permaculture systems, and agroforestry-typical conventional farming systems. Understanding the pathways and dynamics of the movement of PFAS into and within agroecosystems is critical as, at present, dietary intake is understood to be the most significant source of human exposure to these chemicals (Death et al., 2021). The potential impact that PFAS may have on human health via dietary intake is more pressing when the persistence and longevity of these chemicals within agroecosystems is taken into account.

The recalcitrance of PFAS within ecosystems results in widespread distribution of the chemicals via many pathways. Due to their unique stability and mobility, volatile PFAS reaching up to 14 times the US EPA's lifetime advisory limits have been documented in rainwater in remote regions of Tibet (Chen et al., 2021). Recent studies have shown elevated levels of PFAS in both background environmental measurements as well as in agroecosystems across the world for a number of reasons: introduction of PFAS into agroecosystems through the application of biosolids (Caniglia et al., 2022) via atmospheric deposition (Schroeder et al., 2021), and, most importantly, through sources of contaminated water (Costello & Lee, 2020). The distribution of PFAS on a global scale helps to shed light on how pervasive the chemicals have become via source water.

For two to three decades, research has shown measurable amounts of PFAS virtually worldwide (Figure 2), with even some of the earliest data registering open ocean samples in the range of tens of picograms per liter in the central Pacific Ocean (Yamashita et al., 2005). Certain regions of the world have higher background concentrations of PFAS than others, but measurable quantities of PFASs have been detected on every continent and in many regions that have never had any large-scale manufacturing of the chemicals (Kurwadkar et al., 2022). While there have been trends and recent developments about limiting the production and transport of PFAS-containing products in certain regions, the resilience and mobility of these chemicals ensure that PFAS will remain in the environment virtually indefinitely without purposeful remediation efforts (Evich et al., 2022). Specific to the United States, recent data point toward the Eastern regions of the country as having higher concentrations of perfluorooctanoic acid (PFOA) and PFAS compared with the Western regions (Kurwadkar et al., 2022). Even with seemingly lesser

concentrations depending on location in the United States, the risk that PFAS poses to agroecosystems is not decreased in western regions of the United States: research still points to the presence of elevated concentrations of the chemicals in essential components of agroecosystems and agricultural production. Groundwater and surface water are essential sources of irrigation for agriculture, and regions of the United States that are traditionally thought of as “agricultural hot spots” (the American Midwest, the Central Valley in California, and the Great Plains) are some of the regions wherein source waters have elevated concentrations of PFAS and their precursor compounds (Kurwadkar et al., 2022; Pfothner et al., 2022). Regardless of levels of PFAS within specific regions, several studies have pointed to measurable amounts of PFAS in many sources of drinking water in the United States (Andrews & Naidenko, 2020; Kurwadkar et al., 2022; Pfothner et al., 2022). Seemingly low background concentrations of PFAS should still warrant concern through an agroecological perspective: The nature of uptake and bioaccumulation of PFAS seen throughout agricultural systems allows for low concentrations of the chemicals within soil and irrigation water to result in elevated concentrations within crops (Ghisi et al., 2019).

In this review, we provide an overview and understanding of the current science surrounding PFAS within the context of agroecosystems. The review approaches this subject by following the pathways of PFAS into and through agroecosystems starting with their introduction into the environment, moving toward their effects on the human body, and ending with remediation techniques. As well as providing a general overview of the mechanics of the chemicals, this paper examines a few case studies that support or exemplify some of the greater trends noticed throughout the literature available at the time of review. Through a multifaceted analysis of PFAS dynamics within agroecosystems, this review seeks to contribute valuable insights, identify gaps in the research, and promote a better understanding of the intricate relationship between these persistent chemicals and the ecosystems our societies rely on.

This review was conducted by systematic analysis of published research and review papers over the last 20 years on July 2023, using the Web of Science search engine.

2 | MAIN SOURCES OF PFAS IN AGROECOSYSTEMS AND THE ENVIRONMENT

Since their inception in the early 20th century, PFASs have been a part of many industrial and consumer products worldwide. Though these compounds may not intentionally be applied to or used in agroecosystems, they may enter these environments through various pathways. Sources of PFAS

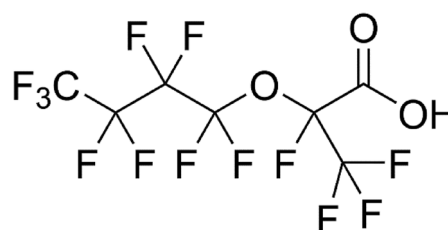
Core Ideas

- The most significant sources of per- and polyfluoroalkyl substance (PFAS) in agroecosystems are biosolids and contaminated irrigation water.
- The current methods for PFAS remediation are prohibitive for wide-scale usage in agricultural settings.
- There is a lack of substantial data on PFAS directly related to agriculture and agroecosystems.

in agroecosystems, either direct or indirect, are listed in Table 1. Two common point-source releases of PFAS into agroecosystems are aqueous film-forming foam (AFFF) contamination and land applications of wastewater byproducts such as biosolids; PFAS contamination can occur through the accidental release of AFFF at or near agroecosystems or into the components used within those ecosystems. Releases of AFFF can result in contaminated runoff and infiltration into soil and water bodies, both of which are essential components of agroecosystems. Improper treatment of influent at wastewater treatment plants (WWTPs) can release standard effluents, which can make their way into soil, rivers, lakes, and ecosystems. The release of such effluent can contribute to overall concentrations of PFAS in the receiving waters and mediums at, and near, any points of discharge. The literature has identified that the most prevalent sources of PFAS in agroecosystems are the direct land application of biosolids, fertilizers, and contaminated irrigation water.

2.1 | Biosolids or sewage sludge as a nonpoint source of PFAS

Recently, the use of biosolids as organic amendments to enhance soil health in agriculture and residential landscapes



Heptfluoropropylene oxide dimer
HFPO-DA or GenX

FIGURE 1 A chemical structure graphic of GenX.

TABLE 1 Major point and non-point sources of PFAS released into agroecosystems globally and specific to United States based on research studies in the last two decades

Origin/source/industry	Uses	Chemical compounds	Study location	References
Aqueous film forming foams (AFF)	Fire training facilities, airports, and military bases	PFOS, PFOA, PFBA, PFBS, PFHxA, PFHxS, PTFE, PVDF, PFCA	Alabama, IA (USA), China	(Pepper et al., 2021; Prevedouros et al., 2006; Wei et al., 2017)
Commercial/industrial products/food packaging industry/paper industry	Paper and packaging, pharmaceutical and personal care products (cosmetics, paperboards) wires, rubber coating and insulation, anti-graffiti paints	PFOA, PFOS, PFBA, PFAS	New York (USA), China	(Curtzwiler et al., 2021; Fenton et al., 2021; Grandjean & Clapp, 2015; Jha et al., 2021; Kim & Kannan, 2007; Prevedouros et al., 2006; Scheerder et al., 2005; Wei et al., 2017)
Commercial household products	Nonstick cookware, wetting agents in floor polishes, personal care products (cosmetics, toothpaste)	PFOA, PFOS, PFBA and other PFAS, PASE, FFFPs, AFFF	Germany, Canada, USA, Russia, Norway	(Jha et al., 2021; Organisation for Economic Co-operation and Development, & United Nations Environment Programme, 2013; Rankin et al., 2016)
Fertilizer companies and wastewater treatment plant (WWTP)	Fertilizers and biosolids made from WWTP	PFOA, PFOS, PFBA, PFCAs, PFASs, PFHpA, PFNA, PFDA, PFUnA, NMeFOSAA (precursor), PFDoA, PFBS, PFHxA and others	Alabama, Arizona (USA), Antarctica, Uganda, Vehendi Estonia, Southern Cross Australia	(Caniglia et al., 2022; Clara et al., 2008; Jha et al., 2021; Levine et al., 2023; Pepper et al., 2021; Sepulvado et al., 2011)
Irrigation water	Recycled wastewater for irrigation as a perennial source of certain PFAS	PFAS, PFOS, PFOAS, PFCAs, PFSA	USA, France, Netherlands, India, Canada, Japan, Mexico, Norway, Tibet, Pacific Ocean, Burkina Faso, Chile, Ivory Coast	(Chen et al., 2021; Costello & Lee, 2020; Kurwadkar et al., 2022; Oliaei et al., 2013; Pepper et al., 2021; Sharma et al., 2016; Wei et al., 2017; Yamashita et al., 2005)
Pesticide industries	Pesticides such as sulfluramid	PFOs, PFHxS, FOSAA, FOSA, PFOA	USA, China, Brazil	(Nascimento et al., 2018; Organisation for Economic Co-operation and Development, & United Nations Environment Programme, 2013; Pepper et al., 2021; Wei et al., 2017)

Abbreviations: FFFPs, film forming fluoropolymers; FOSA, perfluorooctanesulfonamide; FOSAA, perfluorooctanesulfonamide; PFBA, perfluorobutanoic acid; PFBS, perfluorobutanoic acid; PFDA, perfluorodecanoic acid; PFHxA, perfluorohexanoic acid; PFHxS, perfluorohexanesulphonic acid; PFNA, perfluorononanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonic acid; PFSAs, perfluoroalkyl sulfonic acids; PTFE, polytetrafluoroethylene; PFUnA, perfluoroundecanoic acid; PRB, permeable reactive barrier; PVDF, polyvinylidene fluoride.

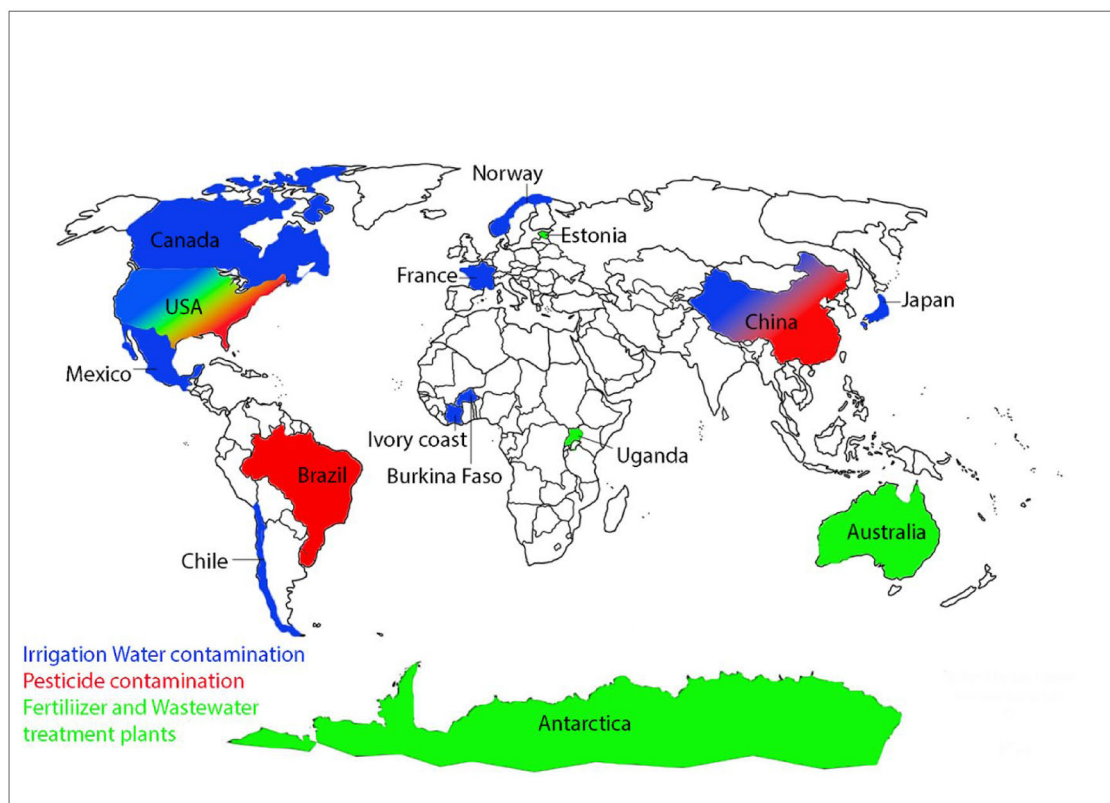


FIGURE 2 Global presence of per- and polyfluoroalkyl substances (PFASs) at sampling locations due to the application of biosolids, irrigation water, and pesticides by accumulation and leaching in the agroecosystems published till September 2023 (based on 22 studies). For further details on PFAS sources, uses, chemical compounds, and location of studies refer to Table 1.

has been on the rise. The US EPA implemented the National Biosolids project in 41 states in the United States and reported that 53% of biosolids produced nationally were recycled into soil (NBDA, 2020). Biosolids are repurposed wastewater byproducts from WWTPs that can be utilized as an organic amendment to boost overall soil sustainability. The practice not only converts the waste materials into useful organic products but also increases soil organic carbon content. However, because of their origin from WWTPs and the chemicals they may contain, biosolids-based feedstock can serve as a potential source of organic contaminants. Johnson (2022) examined agricultural soils that had been amended with Class B (mid-1990s–early 2000) and Class A (early 2000 to present) biosolids at depths of up to 9 m. He reported that concentrations greater than 1 (perfluorocarboxylic acids [PFCAs]) and 2 (perfluoroalkyl sulfonic acids [PFASs]) orders of magnitude were higher than global soil backgrounds, while PFASs were undetectable in an adjacent irrigation ditch. A statistically significant correlation between Σ PFAS soil content and historical biosolid loading rates was identified; this correlation was more significant for long-chain (more than seven carbons) PFAS, underscoring their persistence in soil. Perfluorooctane sulfonic acid (PFOS) and PFOA concentrations were found to decrease with soil depth, with little PFASs found below 5 m. A similar study detected 18 PFASs compounds in soil that had

been repeatedly treated with Class B biosolids for 35 years, while no PFASs were detected from samples collected from undisturbed sites with no history of agriculture, irrigation, or biosolids application. This study also revealed that total PFAS concentration declined with soil depth with an average attenuation of 73% at the top 183 cm of the soil surface (Pepper et al., 2021). Levine et al. (2023) conducted a soil column experiment to evaluate the potential sources of PFAS by application of various commercially available compost products to residential landscapes. The analysis found that perfluorobutanesulfonic acid (PFBS), PFOA, and perfluorohexanoic acid (PFHxA) leached from biosolids-based fertilizers, whereas no PFAS was detected from a manure-based compost. Organic amendments through biosolids may pose human and environmental health hazards when leached contaminants enter food crops grown in agricultural or home garden plots (Lazcano et al., 2020; Pozzebon & Seifert, 2023; Sivaram et al., 2022). A few toxicology studies have reported that biosolids, when land-applied directly to soils, are often well below concentrations known to be of environmental concern (Clarke & Smith, 2011). Despite this, there is an ongoing debate on the safety of land-applied biosolids in agricultural and residential landscapes due to limited data on contaminant levels and subsequent risks to human and environmental health (Pepper et al., 2021).

WWTPs with a high industrial influence tend to have higher levels of PFAS. Studies have found conventional WWTP to be not effective in removing synthetic organic compounds and later these pollutants accumulate in biosolids (Koumaki et al., 2021; Richardson & Ternes, 2018). A report by Statewide assessment of PFAS in Michigan found WWTP was not able to break down the C—F bond and thereby converted the perfluorinated precursors to perfluoroalkyl acids which are highly resistant to degradation (Helmer et al., 2022). Agroecosystems located near industrial operations raise concerns about the presence of heavy metals and organic chemicals in the soil and air. To date, only two states in the United States have adopted regulations on PFAS presence in biosolids. In 2021, Michigan State initiated prohibiting land application of industrially produced PFAS and recommended testing before application. In 2022, the Maine state House and Senate both passed a bill (LD 1911) that banned the use of biosolids that contained PFAS in land applications unless they were free from PFAS.

2.2 | Irrigation water as a nonpoint source of PFAS

Irrigation water can often be an important source of PFAS in the soil. A long-term study revealed that the application of class B biosolids and irrigation water for 35 years on agricultural land had very low non-detect levels of PFOS followed by PFOAs in irrigation water and soil samples (Pepper et al., 2021). In urban water bodies, concentrations of PFASs were observed in bulk air (sum of vapor and particulate phases) in the range of 8.28–16 pg/m³, in rainwater 0.91–13.2 ng/L, in snow 0.91–23.9 ng/L, in surface runoff water 1.11–81.8 ng/L and in lake water 9.49–35.9 ng/L, respectively (Kim & Kannan, 2007). In India, surface water from river Ganges, which is often contaminated in the form of spills, leachates and infiltrates, is utilized for irrigating agricultural crops. An analysis of 21 samples of irrigation water by Sharma et al. (2016) displayed the presence of approximately 15 different PFAS compounds; however, they were significantly lower suggesting it to be below safety thresholds.

In Minnesota, PFAS contamination has been caused by discharges and deposits from 3 M Company, the chemical manufacturer, at one of the largest per/polyfluorinated chemical (PFC) production facilities from 2002 to 2013. The facility's operations and disposal resulted in contamination of ground and surface waters including local drinking water wells. The fish in the river downstream of that plant were shown to have been contaminated with PFOS, and impacts on human health were documented due to the ingestion of contaminated drinking water (Oliaei et al., 2013).

Determining the safety thresholds of PFAS is difficult, as the current schema of grouping PFAS as one batch of

chemicals does not convey the intricacies between individual species. The EPA released the 2022 interim updated health advisory limits for PFOA, PFOS, GenX chemicals, and PFBS as anywhere from 0.004 parts per trillion, or 4 ng/L, to 2000 parts per trillion, or 2 µg/L; a difference of six magnitudes. Something to be considered is that this is defined by the EPA as the level at which these chemicals "... offer protection for people from adverse health effects resulting from exposure throughout their lives to these individual PFAS in drinking water" (US EPA, 2024b) and these reflect only the limits in finished drinking water. The current methods approved by the EPA for the detection of certain PFAS of concern (Method 537.1 Rev 2.0, and Method 533) cannot detect PFOA or PFOS below the recommended health advisory limits, meaning that if the analysis of a sample of drinking water has detected either of the chemicals, it is already past the recommended limit (US EPA, 2024a).

3 | FATE AND TRANSPORT OF PFAS

3.1 | PFAS in soil and water

PFAS movement through agroecological system components such as soil, water, and air can occur through several different mechanisms. PFAS can migrate through diffusion, advection, erosion, groundwater incorporation, volatilization, atmospheric transportation via fine particulate matter or droplets, and various other mechanisms. Listed below are a few of the most common or significant transport mechanisms of PFAS into/within the soils.

3.1.1 | Sorption

PFAS binds to the surface of soil particles primarily by sorption, which can be attributed to either hydrophobic or electrostatic interactions, or even both. The rate of sorption and the type and intensity depends upon a number of factors: PFCA and PFSA, the chemical properties of the PFAS species, soil biogeochemical characteristics such as soil pH, electrical conductivity, organic carbon content, cation-exchange-capacity, soil texture, soil micropores, and iron oxides; sorption is dependent on the interactions between these characteristics. Interactions between PFAS with their longest fluorocarbon tails and soil particles leads to the formation of strong, long-chain PFCA and PFSA sorption sites to soil. Soil organic matter (SOM) contains hydrophobic moieties which interact with the tails of PFCA and PFSA thus attaching to these sites. Hence, the higher the SOM, the more sorption of PFAS. Long-term application of biosolids containing long-chain PFAS has been shown to increase the probability of PFAS adsorption to soil. At present, several studies have identified that soils have the potential to serve

as a significant global reservoir of PFAS (Rankin et al., 2016; Strynar et al., 2012; Washington et al., 2019).

3.1.2 | Leaching

Many PFAS compounds are water soluble and may be transported via runoff or through groundwater movements and migrate through soil. PFAS can percolate through soil, moving with water through soil pore spaces. These particles' movement depends on soil hydraulic conductivity and flow rate within the soil pores. Sometimes, organic matter in the soil can act as a sorbent for PFAS, affecting their mobility and availability for leaching. PFAS leaching from composted biosolids can cause environmental contamination. Levine et al. (2023) demonstrated through an experimental test that the leachate concentrations of certain composts, specifically those "...that align with the US Composting Council Seal of Testing Assurance standards" have been found to result in low concentrations of PFAS when applied to agroecosystems.

3.1.3 | Surface runoff/soil erosion

When precipitation occurs and surface water flows over contaminated areas on the soil surface, PFASs are carried away through runoff. Surface water itself often carries many PFAS compounds through contamination by discharges either from industrial waste, wastewater treatment plant effluents, or soil leachate into aquatic systems. It has been noted that atmospheric deposition can lead to changes in surface water concentrations of PFAS, with the fact that relatively isolated surface waters may still have measurable concentrations of PFAS despite no clear point source or nonpoint source discharges. A study by Kim and Kannan (2007) measured the concentration of PFAS in different environmental matrices and found that the total PFAS concentration in surface runoff ranged between 1.11 and 81.8 ng/L. Surface runoff can account for significant contamination by PFAS in lakes.

3.1.4 | Persistence

One of the most significant qualities of PFAS is their persistence in the environment; they cannot easily break down and therefore remain in environmental mediums for extended periods of time. PFAS persistence in soil is either increased or decreased based on the ability of microorganisms to degrade these compounds which in turn impacts their potential for leaching in the soil. The hydrogeological properties of groundwater, such as its flow rate and flow direction, will also influence the leaching ability of PFAS and may further spread the contamination in the soil.

3.1.5 | Vapor intrusion

Large industrial facilities, landfills, and other contaminated sites where PFASs have been utilized or disposed of for extended periods can give rise to vapors that can contaminate soils and other environmental mediums. PFAS contaminated groundwater settling on shallow depths near the soil surface can release PFAS laden vapors, which migrate within agroecosystems. Mechanisms of PFAS transportation and their environmental fate is an area of ongoing research for scientists to better understand and manage PFAS contamination and remediation. Ammonium perfluorooctanoate, a kind of PFAS, was reported to be transported via air emissions from a fluoropolymer manufacturing site to a well field by depositing in soil and then later carried along precipitation to underground aquifers (Davis et al., 2007).

3.2 | PFAS in water

The current understanding of how PFAS enters into agroecosystems is, as previously stated, largely via contaminated source water—through the application of improperly or under-treated fertilizers, biosolids, and other soil conditioning additives. To understand how PFAS-contaminated source water enters into agroecosystems, we first have to understand how the chemicals enter water bodies. One of the most significant sources of contamination is direct release; one of the more common methods of direct release is via aqueous film forming foam (AFFF).

AFFFs are a type of fire retardant, which was first invented in the early 1960s via a joint effort of the United States Navy and the chemical manufacturer 3 M. AFFF is a remarkable and useful invention in terms of its firefighting capabilities largely because of the presence of a multitude of PFAS species, notably PFOS and PFOA, which allow for a dense and stable foam to form in high heat conditions, perfect for the intended use of the compound. AFFF is released into the environment predominantly at aviation and military sites and during other firefighter training and testing situations where an alcohol-resistant retardant is needed. Traditional AFFF has been in a phase-out process in the United States (excluding some exceptional circumstances) beginning at the federal level in late 2019. Nonetheless, explicit bans on either AFFF as a whole or on mixtures containing PFASs are slow to materialize due to the utility of the compounds (FEMA, 2022).

PFASs from AFFF enter the environment through emissions and travel via run off resulting in contamination of those systems. Once AFFF or other compounds reach soils, the molecules adsorb to particulate matter via electrostatic interaction (McGarr et al., 2023). Precipitation, surface flow, and groundwater interactions allow for PFAS to distribute broadly

throughout agroecosystems. Several recent papers discuss tide and wave associated movements due to their importance in the potential aerosolization of PFAS, contributing to atmospheric transport (Faust, 2023). Faust (2023) concluded that additional long-term monitoring of PFAS on particulate matter should be conducted in under-sampled areas to better understand the nature of PFAS transport. One conclusion that can be taken away from the paper is that global projections for PFAS might not be taking into account a potentially important component of atmospheric transport.

While surface water dynamics are important to the movement of PFAS, groundwater dynamics are another important area to consider within the confines of these chemicals and their movement in agroecosystems; groundwater is often used as source water for irrigation within agroecosystems, and if not used directly as source, water can still have impacts on surface water. While there are many individual sources of PFAS that may contribute to groundwater contamination (Hepburn et al., 2019; Kurwadkar et al., 2022; Wei et al., 2018), virtually all incidences of contamination of groundwater operate based on the same base mechanics: leaching and infiltration (B. Guo et al., 2022). Groundwater PFAS concentrations are impacted via surface water runoff, atmospheric deposition, and point source releases of AFFF or other PFAS-containing compounds, and once contaminated, act as a sink for large periods of time, producing contaminant “plumes.” Gefell et al. (2021) identified that groundwater plumes can last for extended periods of time without remediation if sufficient precursors are present. Complicating this extension and modeling of PFAS in groundwater is the recently discovered trend showing that concentrations of PFAS and their precursors at the surface-water/groundwater boundary are impacted by seasonal variations (Tokranov et al., 2021).

Two areas of water transport of PFAS that warrant further study are seasonal dependencies and the effects that tropical storms or storm events have on PFAS concentrations within waterbodies. A recent study has demonstrated that the concentration of short-chain PFAS seems to be significantly affected by seasonal and hydrological conditions, whereas long-chain species such as PFOS show little dependence on these conditions (Tokranov et al., 2021). However, further study is warranted before any trends can be identified across multiple systems. An increased understanding of the effects of seasonality on transport of the various classes of PFAS could contribute to the refinement of existing transport models. Understanding the effect that storm events have on PFAS mobility is important due to wave-action and the potential re-aerosolization of the chemicals following those storms. Few studies regarding the specifics of transport following a storm event have been published; however, one model based off of real-world data theorized that PFAS released during

storm conditions can travel up to 31 km in 2 days (Hodgkins et al., 2019). An aspect of large storms or hurricanes that was outside of the bounds of Hodgkins et al. (2019) is the resuspension of sediment often seen during large storm events. According to Ahmadiresket et al. (2021), total PFAS concentrations in sediment cores taken from the Pensacola Bay System watershed following Hurricane Sally were 47% lower on average compared with pre-hurricane sediment samples. Martinez et al. (2022) found a maximum of an 177% increase in mean PFOS levels in St. Augustine Intracoastal waterway following Hurricane Dorain—interestingly enough, the study also noted that “This study observed a considerable increase of PFOS during the 2019 Hurricane Dorian while other PFAS had little to no changes in concentration in comparison.” The results of these two studies point toward PFAS resuspension following hurricanes, but without further data and analysis, no firm conclusions can be drawn regarding typical interactions between large storm events and PFAS concentrations in coastal areas.

3.3 | PFAS in air

The movement of PFAS through the atmosphere is gradually becoming better understood, both in terms of the forces that drive its spread as well as the potential health risks. The dangers these chemicals pose to agroecosystems is not through direct atmospheric uptake on parts of the plants, but rather in the deposition of atmospheric contaminants onto soils and waters as components of those agroecosystems (Olatunbosun et al., 2020). Point-source emissions of PFAS via industrial processes or disposal practices can lead to widespread environmental contamination via sorption to soil and vapor particles (Dauchy, 2023), but it is important to note that the PFAS that spread via the atmospheric medium are largely neutral PFAS, with fewer of the ionic PFAS that typically accumulate in water, sediment, and soil (Kurwadkar et al., 2022). Neutral PFASs are still a meaningful source of hazard with concerns to agroecosystems; however, as neutral PFAS can be converted into ionic PFAS via photochemical and biological processes, both of which are abundant within agroecosystems (S. Wang et al., 2020b; Q. Wang, Ruan, et al., 2020). Atmospheric PFAS emissions and their precursor compounds are a difficult problem to manage: the majority of regulations toward PFAS either focus on drinking water remediation or are simply lacking with respect to thoroughness or a cradle-to-grave approach. Recent discoveries point to the potential for PFAS to be released into the atmosphere from the processing or disposal of the recycling of lithium-ion batteries (Rensmo et al., 2023). Rensmo et al. (2023) wrote that a trend in the electric vehicle manufacturing industry has the potential to be a source of PFAS due

to the processes that many recyclers have begun to adopt. Along the same pathway of emissions, landfill-dumping and other disposal methods of PFAS-containing products that do not eliminate the chemicals prior to disposal result in the release of PFAS from the surface of those products, resulting in atmospheric emissions (Capozzi et al., 2023; Hamid et al., 2018; Tian et al., 2018). Degradation of certain precursor compounds such as fluorotelomer alcohols (FTOHs) and per-fluoroalkylsulfonamides (e.g., NMe-FOSE, NEt-FOSE, and NMe-FOSA) was also noted, which later transform to PFCAs and PFSA in the environment (Kim & Kannan, 2007). In addition to general leaching from land sites, PFASs were noted in a review by Faust (2023) to have the capability to "...be transferred from the sea surface to sea spray aerosol particles through wave breaking and bubble bursting, and PFAS emitted to the atmosphere in the gas phase can sorb to particulate matter through gas-particle partitioning." This ability to aerosolize and move into the atmosphere from sea surface interactions needs to be carefully considered moving forward, as research has shown quantifiable PFAS concentrations in certain surface waters across the world (De Silva et al., 2011; Kurwadkar et al., 2022; S. Wang et al., 2020a). An increase in surface water PFAS concentration would likely result in an increase in wave-action dispersal and aerosolization of PFAS. An increase in the aerosolization of PFAS would result in an increase in atmospheric concentrations, which have been documented as sources of PFAS in soils via electrostatic interactions (Kurwadkar et al., 2022).

3.4 | PFAS in plants

Plants have been found to bioaccumulate certain PFASs such as PFCAs and PFSA (Ghisi et al., 2019; S. Wang et al., 2020b; W. Zhang & Liang, 2022; D. Zhang, Zhang, & Liang, 2019; W. Zhang, Zhang, Zagorevski, et al., 2019; Zhou et al., 2020). A new replacement class called Ether PFAS (fluoroalkyl ether compounds) is on the rise; the distribution of five ether PFAS chemicals (PFMOPrA, PFMOBA, GenX, ADONA, and F-53B) was investigated for their presence in plant-soil systems. Uptake of the chemicals was confirmed in the plant *Carex comosa* (longhair sedge) through plant roots and shoots, indicating their possible presence within agroecosystems. The uptake and accumulation of F-53B PFAS through plant roots in water spinach and its toxicity were reported with limited upward translocation (T. Tang et al., 2020). Food crops such as wheat, maize, soybean, and pumpkin were also found to contain F-53B toxicity at severe levels (Q. Lin et al., 2020; J. Zhou et al., 2020). It was reported that these PFAS could become non-extractable with the aging process because of their reduced mobility in soil and bioavailability (W. Zhang et al., 2021).

3.5 | PFAS in aquatic organisms

Bioaccumulation of PFAS has been documented in aquatic organisms such as fishes and aquatic plants. These compounds enter the organisms either through their food or water ingested and transfers up the food chain. Aquatic organisms impacted by PFAS can pose risks to environmental and human health. A study analyzed the concentration of PFAS in various fish species (smallmouth bass, bluegill, white bass, and freshwater drum) in a section of the Mississippi River located downstream of a 3 M plant and recorded abnormally high concentrations of PFOS in the tissues of the studied species (Minnesota Pollution Control Agency, 2013).

4 | PFAS COSTS, ECONOMICS, POLICIES, AND REGULATIONS

4.1 | Policies and regulations on PFAS

Since their inception in the 1940s, PFASs have seemingly been omnipresent across the entirety of Earth's surface, and understanding regional impacts has become a challenge. Despite these difficulties, regulatory agencies such as the US EPA made efforts to address the problem. Agencies have led or commissioned studies to collect background and baseline data for specific PFASs across different matrices.

Most regions are seeing some increase of PFAS even where legislation is put in place to limit the production and spread of the chemicals, meaningful reductions in background concentrations can take years, if not decades to be realized (Kurwadkar et al., 2022). Further complicating, background levels is the fact that long-range atmospheric transport has been confirmed to be a method by which PFASs are distributed (Chen et al., 2021), meaning that PFAS concentrations are subject to external influence irrespective of the extent that a country or region produces, remediates, or prevents the spread of the chemicals intra-nationally.

For years, many regions around the world have been working to address PFAS contamination in various environmental media. PFAS regulations differ across the globe, with some countries imposing outright bans, some implementing partial bans, and others setting quantitative limits. According to Antea Group (2023), certain types of PFASs such as PFOS and LC-PFCAs are banned in the European Union (EU) under the Stockholm Convention on POP; (Spyrakakis & Dragani, 2023). Denmark, Germany, the Netherlands, Norway, and Sweden proposed restrictions on perfluorinated carboxylic acids (C9-14 PFCAs) in January 2023. The ban is unlikely to take effect soon however, due to the speed at which these legislative actions typically progress.

On February 7, 2023, the European Chemicals Agency (ECHA, 2023) proposed banning an entire class of PFAS.

The ECHA's proposed ban has sparked debates on its potential economic, industrial, and environmental consequences. Although certain types of PFASs are known to be harmful, a complete ban may make the replacement of PFAS-based materials for environmental transition, medical devices, and everyday products difficult; finding safer alternative materials in the short term may be hard.

In recent years, there have been rising concerns in the United States about the adverse health effects of PFAS, which have prompted calls from both federal and state governments to impose limitations on ongoing PFAS use, as well as remediate contaminated water sources (Conder et al., 2008; Reade, 2018). The United States EPA's third Unregulated Contaminant Monitoring Rule (January 2017) estimated that 4% of water sources detected at least one PFAS compound above the minimum acceptable level (Crone et al., 2019).

Governance and research expenses for the establishment and determination of PFAS contamination hotspots are significant; in addition to technical expertise required to assess exposure, the costs associated with ongoing monitoring and enforcement are also significant. For example, North Carolina spent over \$5 million in creating its PFAS Testing Network (Cordner et al., 2021). State and local governments may also incur substantial legal costs. States such as New Hampshire and New Jersey have been sued by PFAS manufacturers for health-protective drinking water regulations, while others such as Minnesota, Alabama, and Michigan received compensation of around \$850 million, \$39 million, and \$168 million, respectively, from PFAS polluting firms (Gardella, 2021).

Despite these contaminations, there is no federal regulatory limit for PFOA and PFOS: the EPA adopted a lifetime health advisory level of 0.004 ppt for PFOA and 0.02 ppt for PFOS (US EPA, 2023a), but this is simply advisory and is non-enforceable. Some states have banned PFAS in food packaging and tightened PFAS in drinking water levels to 1 ppt or lower (Grandjean & Clapp, 2015; Hoylman, 2020). Additionally, the US EPA has started modifying the discharge permits for surface water governed by the National Pollutant Discharge Elimination System (NPDES) and Toxics Release Inventory (TRI) programs to report for PFAS, with the eventual goal of setting maximum contaminant levels (MCLs) for six PFAS compounds in drinking water. This is expected to be finalized by the end of 2023 (US EPA, 2023c), but as of the time this paper was written no such limits have been enacted. PFOA and PFOS concentrations have been proposed to be as low as 4 ng/L for PFOA and PFOS, and 1 ng/L for perfluorononanoic acid, PFHxS, PFBS, and hexafluoropropylene oxide dimer acid (HFPO-DA, commonly referred to as GenX Chemicals) (US EPA, 2023b). Several states passed a law or proposed legislation that outlines quantitative limits or ban PFAS in their entirety prior to federal regulations. One notable example is the state of Maine, which recently passed a first-

in-the-nation law banning products made using intentionally added PFAS. Although the law is planned to be introduced by 2030, companies in the state are required to report their PFAS use effective from January 1, 2023.

PFASs are poorly regulated in Asia and Latin America. Although few countries such as India, Sri Lanka, China, and Brazil have some restrictions on PFOS or PFOA in line with the Stockholm Convention, most of the PFAS chemicals are not regulated (International Pollutants Elimination Network, 2019). With increasing international awareness, the regulations on PFOA are likely to evolve over time.

4.2 | PFAS pollution control policy in the United States

One recent challenge is the management of solids, which may be contaminated with PFAS because of its discharge to sewer systems from private industries as well as people themselves and subsequent accumulation in biosolids produced from municipal wastewater treatment plant processes. This issue encompasses various considerations pertaining to testing, disposal, treatment, migration to groundwater and drinking water quality, and potential uptake to crops and animals if land applied. As there is no federal regulation or guidance for PFAS-contaminated biosolids, it is currently up to individual states to establish and set best practices for managing these contaminated wastewater residuals (Hughes, 2023).

There have been many efforts to better understand the challenges associated with PFAS in biosolids (see 2020 review of models by Arcadis, the National Council for Air and Stream Improvement, Water Environment Federation, American Forest & Paper Association, and NACWA; a 2022 Northeast Regional Sludge End-Use and Disposal Estimate from the New England Interstate Water Pollution Control Commission (NEIWPCC); and the biosolids fact sheet from ITRC, among other resources). Hughes (2023) also argues that the EPA in its PFAS Strategic Roadmap also committed to completing by December 2024 a risk assessment for PFAS in biosolids. Maine is currently the only state with a ban on the land application of biosolids, and very few states have either enacted or proposed legislation for managing PFAS in biosolids. However, many states have or are considering other regulatory actions. New Hampshire is the only state with requirements for facilities to monitor PFAS in biosolids. Massachusetts and Michigan do not have any legislation on monitoring; however, they both have related requirements. A survey of states revealed that no states had a ban on landfilling, with 27 states having not enacted or proposed legislation with impacts on PFAS in biosolids.

Exposure to PFAS through drinking water is also a global human-health risk. According to a recent US Geological Survey (2023) study, up to 45% of the nation's tap drinkable water

is estimated to have one or more types of PFAS. There are over 12,000 types of PFAS, most of which can be detected with current tests. The USGS findings are based on testing of the presence of 32 types. According to the US Environmental Protection Agency (EPA, 2024c) through the Bipartisan Infrastructure Law, the agency is making an unprecedented \$21 billion available to strengthen the nation's drinking water systems, including dealing with PFAS contamination. Of that, \$9 billion is set aside strictly for tackling PFAS and emerging contaminants. Moreover, the agency is setting enforceable Maximum Contaminant Levels at 4.0 parts per trillion for PFOA and PFOS, individually. This standard will reduce exposure from these PFASs in the nation's drinking water to the lowest levels feasible for effective implementation.

Although there are minimal guidelines, some states/regions have offered some action levels for produce, milk, beef, and so on. The Stoneridge and Songbird, in Maine, put the state at the forefront of a farm contamination crisis. Following these incidents, the state has gathered the most comprehensive data on PFAS in food, and as of 2021, its Department of Environmental Protection set action thresholds for PFOS in milk (210 parts per trillion) and beef (3.4 parts per billion), the first standards thresholds for food in the United States. So far, authorities in Maine have detected PFOS above the set standards for sale in at least one instance, which led them back to a farm with contaminated water. State authorities are working to develop standards for more foods, although testing and enforcement remains a challenge (Food Print, 2023).

4.3 | Societal costs and economics of PFAS

Direct healthcare expenditure associated with PFAS exposure in Europe is estimated to range between \$55 and \$89 billion per year (Goldenman et al., 2019). This translates to a population and exchange rate-adjusted annual health-related cost of \$37–\$59 billion in the United States; societal costs are marginal private costs typically borne by polluters plus pollution costs. Indirect social costs, although extensive, are difficult to estimate (Cordner et al., 2021). Such societal costs are plausible and have policy implications (Institute of Medicine [IOM], 1981); however, such indirect societal costs associated with health impact of PFAS in the United States are currently lacking (Cordner et al., 2021).

In 2022, the global market for PFAS was just over \$28 billion; this is a relatively small portion of the total market value of all chemicals, which stands at \$4.73 trillion. To help put this in perspective, PFAS production accounts for only 0.006% of the value of global chemical production. However, the PFAS market size reflects total revenue from the total net sales and not necessarily the industry generated profits (Chemsec, 2023). The societal cost of using toxic PFAS across the global economy is cited as approximately \$17.5

trillion per year, while the profit yielded globally for PFAS manufacturers is paltry, at approximately \$4 billion annually (Perkins, 2023). According to the PFAS industry, compounds used in consumer goods and industrial applications by the US fluoropolymer segment results in significant economic benefits of \$2 billion annually (Wood Environment & Infrastructure Solutions UK Limited, 2020). However, the industry fails to account for the associated costs of long-term, wide-ranging exposure to PFAS. To date, most PFAS-related costs are borne by third parties, such as society and government. The decision-making process on the regulation of PFAS contamination hotspots is time consuming, labor intensive, and expensive (Brennan et al., 2021; Cordner et al., 2021).

PFOA exposure accounted for approximately 4% of low birth weight (LBW) in the United States in 2003 and 2004, with related costs estimated at \$13.7 billion (Malits et al., 2018). In 2019, the Nordic Council of Ministers estimated the disease-related costs due to occupational PFAS exposure in Europe to be around \$55–\$89 billion (Goldenman et al., 2019). It is worth noting, however, that these are conservative estimates as they exclude health expenditure induced by PFAS exposure. The lower bound of PFAS-attributable disease costs in the United States is estimated to be approximately \$5.52 billion, with the higher bound estimated to be as high as \$62.6 billion (Obsekov et al., 2022). These findings should not be used to draw causal conclusions because of confounding factors, hence further research is required (Zabel & Dennis, 2012). This review of the economy surrounding PFAS shows that the societal costs of usage of the chemicals are astronomically high, often borne by third parties (i.e., governments and communities), and far exceed societal benefits.

5 | IMPACT OF PFAS ON HUMAN HEALTH AND ECOSYSTEM

Soil, water resources, and the environment are all essential sources for a healthy population. We depend on these ecosystems as we use them in numerous ways, from medium for plant growth and crop production to the provision of clean, potable drinking water. Unfortunately, both agricultural and nonagricultural activities have led to the fragmentation of these ecosystems' health, which are intricately linked to human health. Outside of climate change, the degradation or contamination of these resources from substances such as PFAS will no doubt cause serious and irreparable damage, resulting in significant threat both to the environment as well as to human health and activities. Further, the cost of ecosystems being degraded is unevenly distributed as vulnerable groups of the population as children, pregnant women, and lactating mothers are more likely to be at risk. From the chemistry of PFAS alone that has been extensively discussed in the current literature, we know PFAS can cause

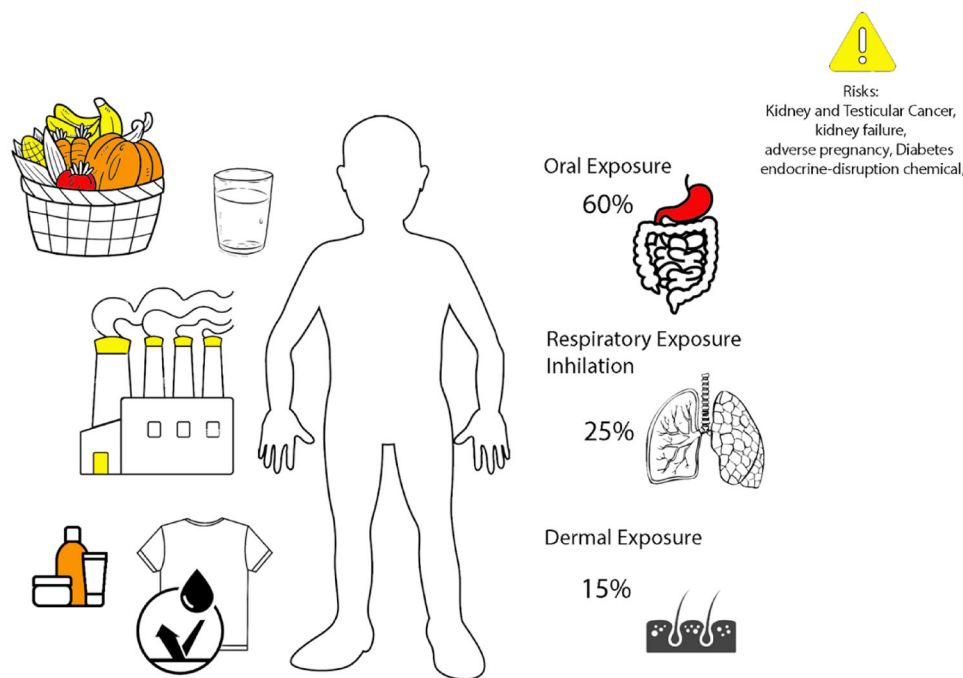


FIGURE 3 Major pathways of human exposure to per- and polyfluoroalkyl substance (PFAS) and their adverse effects on human health.

serious health effects. There are still limits, however, in our ability to fully understand PFAS repercussions on the environment and subsequently on human health (Martin et al., 2010; Panieri et al., 2022; Vestergren & Cousins, 2009). Due to advances in understanding the significance of PFAS on human health from numerous recent toxicological studies and reviews, there is great recognition of the adverse effects of PFAS on human health at a global scale (Dickman & Aga, 2022; Pelch et al., 2019; Schiavone & Portesi, 2023). These reviews and studies report that once PFAS enter the body, they can accumulate in a variety of tissues and organs resulting in toxic mechanisms which impair endocrine, behavioral, neurological, urogenital, hepatic, reproductive, digestive, and respiratory systems. Additionally, some are known to produce carcinogenic effects. Some of the most serious PFAS effects on human health effects are described below.

5.1 | Routes of PFAS exposure to humans

PFASs are among the most persistent toxic chemicals in the environment; this persistence, along with their association with an array of occupational, industrial and household applications, results in multiple routes of human exposure (Fenton et al., 2021; India-Aldana et al., 2023; Pelch et al., 2019; Sunderland et al., 2019). The most significant pathways that PFAS can enter the human body (Figure 3) include oral, dermal, and inhalation, while some lesser known routes include gestational pathways and parental care through breast milk (LaKind et al., 2023; Zheng et al., 2021). Occupational

exposures such as agricultural, chemical manufacturing, and firefighting with AFFF are also common routes (NIOSH, 2023). For the general public, the use of household consumer products, workplace products, as well as being in the general environment are also relevant routes of exposure (Domingo & Nadal, 2019; Haug et al., 2010; Trudel et al., 2008). Outside of the normal routes of exposure, the potential of PFAS to have either an acute, subacute, sub-chronic, or chronic adverse health effect depends on both endogenous and exogenous factors such as concentration, dosage, duration, age, gender, ethnicity, genetic, diet, and general health status (Fenton et al., 2021). While recent work is beginning to identify more definitive trends in relation to the effects of PFAS on human health, much is still unknown; research is still ongoing and our understanding on how these different routes of exposure affect human health increases day by day (Nadal & Domingo, 2014).

5.1.1 | Dermal exposure

While dermal exposure is one of the most common routes through which humans can be exposed to PFAS, it is also the least researched with very little information available (Ragnarsdóttir et al., 2022). Dermal exposure and absorption is usually caused by application of cosmetics or wearing of water repellent clothing (Schultes et al., 2018; van der Veen et al., 2020; Xia et al., 2022). In addition, absorption may occur occupationally during firefighter training, at fluorochemical manufacturing plants, and in professional ski

waxing (Dobraca et al., 2015; Lucas et al., 2023; Mazumder et al., 2023; Rosenfeld et al., 2023)

5.1.2 | Oral exposure

Ingestion has long been recognized as the primary route of PFAS exposure in humans. Oral exposure usually arises from consuming contaminated water or food; in support of this, there are many accounts of human ingestion via contaminated food or food products (Berger et al., 2009; Miranda et al., 2023; Piva et al., 2023; Schaidt et al., 2017; Scher et al., 2018). Results from animal bioassays are key in determining human health hazards. Rodents are commonly used in toxicological studies to predict human health toxicity outcomes. In one such study, laboratory investigation on the effects of mice from oral PFAS exposure showed that the liver was the primary target of toxicity (Heintz et al., 2023).

5.1.3 | Respiratory exposure

Due to the presence of airborne particulate matter of PFAS, and because they are distributed differently between indoor and outdoor environments, the potential for respiratory exposure should be considered significant (Hall et al., 2020; Shoeib et al., 2005). A large number of studies have reported on the presence of PFAS in indoor dust from homes, and various types of businesses (childcare centers, colleges), as well as outdoors such as agricultural lands (Borthakur et al., 2022; Fraser et al., 2013; Ismail et al., 2023; Juhasz et al., 2023; Schildroth et al., 2022; Winkens et al., 2018; Young et al., 2021).

Impacts of PFAS exposure

Cancer. There is interest in the link between PFAS and cancer in humans, due in part to studies showing an association between PFAS and certain types of cancer in rats and trout (IARC, 2017; Lau et al., 2007). Human study results, however, have been inconsistent. According to a meta-analysis of epidemiologic studies, Chang et al. (2014) found no clearly defined linkage in human exposure situations of PFOA or PFOS and cancer. In a population-based study of residents who lived in a PFAS exposure area in Australia, evidence was minimal in suggesting that exposure to PFAS either caused cancer or was a contributory factor to certain types of deaths (Law et al., 2023). Despite the association between PFAS and cancer being inconclusive in some instances, most studies suggest that exposure to certain PFAS increases the risk of cancer in humans. In a literature review based on three (cohort, case-control, and cross sectional) different types of epidemiologic study designs, Steenland and Winquist (2021) found that (while the results were inconsistent)

there is enough evidence to suggest that there is some association between PFAS and testicular and kidney cancer. Gender specific abnormalities such as testicular, prostate, ovarian, and other cancers, which are hormone dependent, have been reported after prolonged exposure to high concentration of PFAS (Bartell & Vieira, 2021; Bonefeld-Jørgensen et al., 2014; Boyd et al., 2022; Freire et al., 2023; Messmer et al., 2022; Steenland & Winquist, 2021). Both case-control as well as cohort and ecological studies have shown that testicular cancer, compared to all other cancers, provided the strongest association yet of the carcinogenicity of PFAS (Bartell & Vieira, 2021; Steenland & Winquist, 2021).

Endocrine function and metabolism. Certain chemicals or compounds have the ability to disrupt the normal functioning of hormones and the endocrine system which eventually manifest in reproductive, neurological, and metabolic abnormalities (Ahn & Jeung, 2023; Diamanti-Kandarakis et al., 2009). PFASs have been regarded as an endocrine-disruption chemical based on a wide range of studies. For example, several epidemiological and clinical findings have presented an association between PFAS exposure and symptoms of kidney failure (Stanifer et al., 2018). Additionally, exposure to PFAS during pregnancy was positively correlated with adverse effects on mothers as well as fetal abnormalities (X.-X. Gao et al., 2023; C.-Y. Hu et al., 2023). In other studies, PFAS exposure contributed to the exacerbation of adverse pregnancy outcomes such as preeclampsia, low birth weights, as well as an increased risk of preterm births (Blake & Fenton, 2020; X.-X. Gao et al., 2023; Kashino et al., 2020; Meng et al., 2018). Emerging scientific evidence also suggests that PFAS exposure increases the risk of developing diabetes. Based on observations from a nested case control study of adult diabetes mellitus among US women, Q. Sun et al. (2018) reported a potential relationship between higher plasma concentrations of PFAS and incidences of diabetes. (K. Gao et al., 2019; Kashino et al., 2020).

Ecological health risks. PFASs' chemical and physical properties such as their persistence and abilities to bioaccumulate and biomagnify often influence their ecotoxicity. Their persistence and transformation in both aquatic and terrestrial biomes for years, not only have a direct impact on the health of the local ecosystems but have indirect negative impacts when they are transferred and recycled to other levels and species throughout the food chains of diverse ecosystems (Dalal-meh et al., 2018; Ghisi et al., 2019; Miranda et al., 2023; Squadrone et al., 2015). PFAS mobility and transfer routes are however not only limited to movements within the food chain itself, organisms and animals can also be exposed to PFAS-contaminated soils from foraging for food or simply from burrowing behavior in and around their habitats (Brake et al., 2023).

Several studies have shown the ecological impact of PFAS on soil properties and structure. For example, PFAS affects

soil structures and soil ecosystems functions such as biogeochemical cycles, which then negatively affects plant growth and crop production (Y. Wang et al., 2023). PFAS concentrations in soils therefore pose an ecological risk to ecosystems, the environment, and humans. The persistence of PFAS in soils has a close relationship with absorption, sorption, and desorption properties (Bradl, 2004; Milinovic et al., 2015). Soils through these properties maintain large amounts of pollutants, act as a sink for PFAS then reemit the PFAS into the atmosphere and aquatic systems, which then plays an important role in ecotoxicity posing a risk to both aquatic and terrestrial habitats and their inhabitants. A study investigating global concentrations of PFAS in soils from diverse locations of the world including urban, rural, and industrial areas found that of over 100 contaminated soils, PFAS concentrations were as low as 0.4 to a high of 460,000 µg/kg (Brusseau et al., 2020). With such widespread concentrations and occurrences, both ecological and human health risks exist.

Several studies show that PFAS can cause injury to many different animal species including dairy cows, dogs, cats, bears, eagles, as well as rats (Guruge et al., 2008; Wu et al., 2020). For example, Brake et al. (2023) reported that exposure to PFAS concentrations caused changes to enzymes, cholesterol, and thyroid hormone levels in livestock. Furthermore, a large number of studies have reported on the negative effect of PFAS on seafood including both marine and freshwater fish species such as salmon, trout, flounder, and cod (EFSA, 2020; Martin et al., 2003; Penland et al., 2020; Schrenk et al., 2020). Equally, multiple organs failures, reproductive, respiratory, circulatory impairments along with endocrine, enzymes, and metabolism imbalances were reported in studies where mice were exposed to PFAS (Blake et al., 2020; Crebelli et al., 2019; Deng et al., 2020; Torres et al., 2021).

PFASs have also been shown to have negative ecological effects on microbial communities and plants interfering with processes such as photosynthesis, root and plant growth, cell structure, metabolic functions, and nutrients translocation and biomass production (Ebinezzer et al., 2022; C. Guo et al., 2023; J. Li et al., 2022; P. Li & Li, 2021; L. Zhou et al., 2016). These changes could have a negative effect on food production due to a reduction in plant productivity.

6 | PFAS REMEDIATION AND MANAGEMENT IN SOIL/SOLIDS

Three broad approaches have been undertaken for the remediation of PFAS in solids and soil: degrade, immobilize, or mobilize (Bolan, Sarkar, Yan, et al., 2021). Degradation refers to the destruction of PFAS without pre-extraction, immobilization involves the stabilization of PFAS by restricting their mobility in the environment, and mobilization refers to the removal of PFAS from solids and soils. While many mobiliza-

tion methods can partially degrade PFAS, these approaches typically require subsequent degradation of extracted PFAS. Many of these approaches are currently under investigation. In practice, excavation followed by incineration is mostly used for large-scale PFAS removal from soils or solids. This approach, while energy and cost-intensive, ensures near-complete mineralization of PFAS contaminants. There are few reports of soil remediation specifically for downstream agricultural applications. Despite this lack of information, PFAS remediation in soils and biosolids has been the subject of several recent reviews (Bolan, Sarkar, Vithanage, et al., 2021; Garg et al., 2023; Kang et al., 2023; Mahinroosta & Senevirathna, 2020; Ross et al., 2018).

6.1 | PFAS degradation

Incineration is the most common method for the destruction of PFAS in solids. Incineration of excavated soils can be conducted off site or on-site in portable rotary kilns. Temperatures over 1000°C are required for mineralization of organic matter; these temperatures can be attained by thermal or plasma processes. A significant concern related to incineration is the potential release of products of incomplete combustion such as hydrogen fluoride or freon causing some municipalities to ban the practice within their boundaries or requiring off-gas scrubbing (Aleksandrov et al., 2019; J. Wang et al., 2022). Plasma, which consists of partially ionized gas, can reach temperatures up to 5000°C resulting in complete mineralization of organic matter. However, the high energy and costs required for this approach may prove prohibitive to large scale application, such as at large agricultural plots.

Ball milling is a high-energy mechanochemical degradation technique that has been shown to decompose organic contaminants, including PFAS in soils through intense mechanical stress and heat (Cagnetta et al., 2016). The process may be made more efficient by the addition of co-milling reagents that promote nucleophilic substitution on the carbon backbone of PFAS (Ateia et al., 2021; Battye et al., 2022) or degradation through free radical pathways (Cagnetta et al., 2017; Y.-B. Hu et al., 2018; Hu et al., 2023; Turner et al., 2023; N. Wang et al., 2019; N. Yang et al., 2023).

While *chemical oxidation* of PFAS for water treatment has been widely studied (Ross et al., 2018), a limited number of laboratory-scale experiments using contaminated soil slurries have been reported. Dombrowski et al. (2018) concluded that the oxidation of PFAS in soil slurries using heated sodium persulfate was inefficient due to competing reactions of oxidant with soil organic matter and other radical scavengers. On the other hand, persulfate oxidation has been shown to alter the PFAS composition by increasing the fraction of more mobile PFCAs, suggesting an effective pretreatment for soil washing or soil flushing (Shojaei et al., 2021).

Sonolysis involves the use of high-frequency (>16 kHz) sound waves or ultrasound to promote the decomposition of solutes in solution (Sathishkumar et al., 2016). Sonolysis-induced cavitation causes localized increases in temperature (4000–10,000°C) and pressure (1000 bar) resulting in hydroxyl radical (HO●) formation. Solute is decomposed through pyrolysis in the vicinity of cavitation or by reaction with hydroxyl radical. Additives that readily form free radicals can enhance the degradation process (Hamdaoui & Merouani, 2017; Hu et al., 2018; J.-C. Lin et al., 2015). Sonolysis has proven effective for PFAS degradation in solution (Cao et al., 2020), but few studies using soil have been reported. Using dual-frequency ultrasound for the treatment of contaminated soil, Kewalramani et al. (2022) found that PFAS degradation was minimal, but enhanced desorption of PFAS from soil was observed. On the other hand, the inclusion of 1 ppt sodium persulfate contaminated soil slurry has been shown to enhance PFAS degradation (Lei et al., 2020).

In part due to its strength and scarcity in nature, the C–F bond is highly resistant to microbial degradation. Nonetheless, transformations by bacteria and fungi have been reported (Berhanu et al., 2023; Z. Zhang et al., 2022). Microbial transformations of the head group, such as the hydrolysis of sulfonamides (Mejia Avendaño & Liu, 2015) and the substitution of sulfonate with hydroxyl or carbon–carbon bond cleavage to yield shorter chain PFCAs, have been observed (Huang & Jaffe, 2019; Kwon et al., 2014). Polyfluoroalkyl compounds such as FTOHs may be converted to shorter-chain PFCAs and even to trifluoroacetic acid (M. Sun et al., 2020). Complete biodegradation of PFAS is rare, but recently, microbial defluorination of PFAS has been reported by a bacterial consortium (Huang & Jaffe, 2019) and by a pure microbial culture (Yu et al., 2020).

6.2 | PFAS immobilization/stabilization in soils

The immobilization or stabilization approach to remediating PFAS-contaminated soil involves the addition of sorbents to restrict PFAS mobility and bioavailability, thereby minimizing uptake by plants. Typical sorbents are activated carbon (AC): either granulated (GAC) or powdered, biochar, and clay minerals. Several proprietary adsorbents are available for soil remediation including RemBind and FLUORO-SORB (McDonough et al., 2021). Sorbents are typically left in place and PFAS may desorb and leach over time. Hence, continuous monitoring and repeated treatments may be required. Many sorbents can achieve a significant reduction of leaching of long-chain PFAS from soils; however, immobilization of shorter-chain PFAS remains a challenge (McDonough et al., 2021). Navarro and coworkers compared the effectiveness of AC and biochar over 4 years in laboratory studies

and determined that AC was more effective at retaining PFAS when compared to biochar. Further, biochars tend to lose their ability to retain PFAS over time (Navarro et al., 2023).

6.3 | PFAS mobilization

Thermal desorption (TD) involves the treatment of contaminated soils under vacuum at 300–600°C to stimulate desorption of volatile contaminants (Kang et al., 2023; C. Zhao et al., 2019). This method is well established for volatile organic contaminants (VOCs) such as polychlorinated biphenyls, polycyclic aromatic hydrocarbons, and Hg (Blanchard & Stamnes, 1997) but is currently under laboratory-scale investigation for PFAS (Crownover et al., 2019; Söregård et al., 2020). While the TD process is considered a mobilization technique, the elevated temperatures may lead to partial degradation by pyrolysis or oxidation. TD is conducted ex situ on excavated soil. In situ treatment could be performed using thermopiles or covered piles of excavated soil, which are heated under vacuum to temperatures of 500–600°C using heated rods inserted into the soil or a diesel/gas burner; contaminants are desorbed and volatilized with the collection of off-gasses (Kaštanek et al., 2020). The approach has been used to remove VOCs from Superfund and other sites (Cappuyns, 2013; US EPA, 2012) and has been considered for PFAS removal (Mahinroosta & Senevirathna, 2020; Ross et al., 2018) but at the time of this writing no field scale experiments have been reported.

Soil washing involves the on or off-site solvent extraction of contaminants from excavated soil. Water is typically the extraction solvent, but organic cosolvents may be included to increase solubility and facilitate desorption. The most common PFASs have relatively high-water solubility (560, 9500, and 1400 mg/L for PFOS, PFOA, and PFHxS, respectively) making soil washing a promising and environmentally friendly remediation method (Abou-Khalil et al., 2022). Recently, a field study of soil washing for PFAS contamination on size fractionated soil demonstrated that the removal efficiency is positively correlated to fraction size with gravel in the range of 95%–99% and the fine soil ranging from 24% to 64% (Quinnan et al., 2022). A recent study reported a significant enhancement of PFAS removal by employing foam fractionation during soil washing (Pang et al., 2022). Foam fractionation is used for the concentration and removal of PFAS from wastewater in which air or gas is bubbled into the solution because PFASs have surfactant properties, a foam that is highly concentrated in PFAS forms on the surface, which may be removed by aspiration or skimming off the surface. Soil flushing is similar to soil washing but involves pumping extraction solvent (again typically water) into contaminated soils in situ avoiding the need for soil excavation

and transport and contaminated wash water is collected for downstream processing (Atteia et al., 2013). A field study comparing soil washing and soil flushing reported comparable PFOS removal rates. Interestingly, the solvent/soil ratio was significantly less for soil flushing to attain similar results, but the predicted in situ treatment time is longer. The soil tested was described as medium fine sand, and the author points out that soil flushing is most effective for sandy to gravelly soil and may not be appropriate for all soil types (Høisæter et al., 2021).

Phytoremediation, the use of plants for the removal of contaminants from environmental media including soils, could represent a sustainable and environmentally friendly approach to PFAS remediation (Adu et al., 2023; K. H. D. Tang, 2023; Hwang et al., 2020). PFAS uptake by plants occurs either through passive diffusion or active uptake by ion channels or aquaporins and is dependent on the PFAS structures (Wang et al., 2020b). The efficiency of plant uptake is characterized by the bioconcentration factor (BCF) which is defined as the ratio of the PFAS concentration in the plant to that of the soil after extraction. BCF values >1 and >10 are indicative of accumulation and hyperaccumulation, respectively (Huff et al., 2020). BCF is negatively correlated with PFAS chain length, meaning that as PFAS chain length increases, the rate at which plants are able to take up those PFAS decrease—it should be understood that this does not mean that longer chain PFAS will not bioconcentrate within plants. BCF is not constant across all plant life; however, recent studies have identified several hyperaccumulating plants (Hilliard et al., 2023; Huff et al., 2020; Nassazzi et al., 2023; Qian et al., 2023; S. H. Yang et al., 2022). Gobelius et al. (2017) reported what may be the largest documented BCF at this time: a remarkable 5450 for the five carbon perfluoropentanoic acid by long beech fern. However, the BCF dropped significantly for PFAS of six or more carbons. Z. Zhang et al. (2022) reported no uptake of PFAS having more than eight carbons into the shoots of timothy grass. Nassazzi et al. (2023) estimated that 458, 466, and 420 90-day crop cycles would be required for 90% removal of PFOS from soil using sunflower, mustard, and hemp, respectively. With respect to biotransformation of PFAS in plants, Phase II conjugates of PFAS (lactic acid and glycosidic) have been reported in pac choi (Sunyer-Caldú et al., 2023), which may enhance the mobility of PFAS. The biodegradation of PFAS by plants is limited and is largely mediated by microbial activities as described above (S. H. Yang et al., 2022; S. Zhao et al., 2018). Complete degradation of PFAS in plants has not been demonstrated but microbial chain length reduction and head group transformations may facilitate plant uptake.

Electrokinetic (EK) mobilization involves the application of a low-intensity electric field across a contaminated area through the insertion of electrodes into the soil. Contaminants are mobilized by electromigration, electroosmosis, or

electrophoresis. Electromigration involves the movement of ions toward electrodes of opposite charge. Electroosmosis is the movement of water molecules toward the cathode, which can help mobilize and transport contaminants. Electrophoresis is the movement of charged colloidal particles toward the electrode of opposite charge. These processes may compete, making the movement of various PFAS species somewhat unpredictable. EK removal of PFAS from soil is negatively correlated to chain length with a sharp drop in efficiency between seven and eight carbons—as a reminder, PFOS, one of the EPA's designated PFAS of concern, is an eight-carbon molecule. Migration may be facilitated by the inclusion of solvents or surfactants and may include the incorporation of a permeable reactive barrier or anion exchange membrane perpendicular to the contaminant plume, which serves to trap the mobilized contaminants (Ganbat et al., 2022, 2023; Niarchos et al., 2022). EK has been found to be most effective for clay-like soils and is therefore complementary to soil washing or soil flushing (Abou-Khalil et al., 2023). This well-developed technology has been used commercially for the large-scale treatment of soils contaminated with heavy metals such as lead, chromium, or cadmium (Rosestolato et al., 2015) and chlorinated organics (Gomes et al., 2012) but is still in the laboratory scale development stage for PFAS.

6.4 | Management of PFAS in agricultural soils

PFAS mobilization and degradation technologies often have high energy requirements, making them a costly approach to remediation of low value or modestly contaminated soils. Furthermore, many of the remediation strategies have not advanced beyond the laboratory scale. Phytoremediation, while being less energy intensive, may be unrealistic due to the time requirements for multiple crop cycles to reduce PFAS concentrations to safe levels. Phytomanagement is the use of plants, soil conditioners, or modified agricultural practices on contaminated lands to minimize environmental and health risks while at the same time providing an economic return from the use of the land (Evangelou & Robinson, 2022; Searce et al., 2023). The phytomanagement strategy has been used on lands contaminated with toxic metals, radioisotopes, pesticides, or polycyclic aromatic hydrocarbons (Garbisu et al., 2020; Gerhardt et al., 2017; Rizwan et al., 2017; Saran et al., 2024). One approach to phytomanagement is to cultivate crops that are not intended for consumption by humans or animals. Examples include timber or biofuels. Alternatively, produce consumed in quantities insufficient to pose a health risk, such as herbs or honey, may be grown on lightly contaminated soil. When crops are grown for human consumption on PFAS contaminated soils, several factors must be considered. These factors

include the specific contaminating species of PFAS, the bio-accumulation factor, and the translocation factors (TFs) of candidate crops, for different classes of PFAS. TF is defined as the ratio of PFAS in the root/shoots. Long-chain PFASs (C6 or higher), which are less mobile, tend to accumulate in the roots, whereas the more mobile short-chain PFASs are translocated to shoots. The growth of root vegetables should be avoided in soil contaminated with long-chain PFAS, while leafy vegetables should not be grown in soil contaminated with short-chain PFAS. PFASs in general have shown less translocation to the edible portion of fruit crops. Both the bio-accumulation factor and TF of PFAS have been documented for numerous food crops (B. Xu et al., 2022). Finally, soil type and the selection of fertilizers should be considered. The organic carbon content of soil is the most significant factor influencing PFAS mobility in soil with high organic carbon soils effectively retaining long-chain PFAS. Organic fertilizers would be favored over inorganic nutrients under these circumstances. Indeed, soil amendments such as GAC or biochar can be used along with phytomanagement strategies to immobilize PFAS. Searce et al. (2023) proposed several factors to be considered for the use of PFAS contaminated agricultural soil along with a conceptual gradient of factors for management decision-making.

7 | RESEARCH GAPS AND FUTURE DIRECTIONS

Despite a growing body of research publications on PFAS in the last two decades, our understanding of these new and emerging contaminants is still extremely limited. We understand much of PFAS's chemical nature, persistence, transport, and other fundamental characteristics, but it has been a challenge to predict the properties of these new emerging by-products.

In our review, we found very few modeling experiments that predict the nature of PFAS contaminations; this is likely due to a lack of long-term studies with large-scale monitoring efforts to precisely identify the cause and effect of PFAS, especially in agroecosystems. Many studies have reported evidence of non-detectable PFAS concentrations in soil, air, water, and plants and are understood to be below harmful levels, but there are numerous studies that point to serious health effects due to their presence in our agroecosystems. However, these contradictory findings would depend upon various other factors such as the age, gender, diet, level of immunity, and overall health of those affected, as well as the concentration and duration of exposure in a particular study.

In the context of this review paper, we noticed that there is a lack of awareness about PFAS in agricultural products among people in general, but especially from agricultural producers, growers, and other agroecosystem affiliates. Producers need

to understand the nature of these inputs, such as biosolids, and the ramifications they pose before utilizing them for growing crops. There is a lack of understanding of PFAS as it relates to socioeconomic impacts, and therefore relevant methods to mitigate these impacts should be in place. Government and policymakers should be able to devise effective policies and regulations at the local, regional, and international levels to increase public awareness and protect their health.

8 | CONCLUSIONS

The primary purpose of this review was to present recent trends and updates regarding the current knowledge and research pertaining to PFAS in agroecosystems. We have reviewed the most common sources within agroecosystems, mechanisms of movement, persistence in various environmental matrices, impacts on human health, cost, policy regulations, and remediation measures to reduce and control contamination.

This review points out the fact that there is not sufficient research available on PFAS in agricultural systems with few studies pointing out the impacts of biosolids on soil and its impact on ecological systems. They are quickly becoming a focal point in the lens of concern for human health, and our knowledge of their impact on agroecosystems is not being generated as fast as their potential threats are. Also researching new emerging and transforming chemical compounds is a challenge for scientists. These new compounds can have unknown effects on our agroecosystems including indirect effects on human health.

Modern agricultural practices have their own pitfalls such as the use of biosolids or the irrigation of crops with contaminated water that have inadvertently contributed to the proliferation of PFAS in soils, surface water, groundwaters, and agroecosystems. These elevated concentrations have resulted in measurable harm to human health. The background concentrations of PFAS around the world have generally risen within the past couple of decades, and as these concentrations continue to rise, so do the impacts they impose on our agricultural systems. Unfortunately, there is not a perfect technique available so far for large-scale agriculture-contaminated soil remediation either because of the size and nature of these persistent organic pollutants or expensive and time-consuming method.

Many countries, states, and regions have begun to enable restrictions to keep away from these harmful chemicals. We believe that this is a great step in the right direction, but larger efforts and consensus both intra- and internationally are needed in the future. It is important that governments pass strict regulations banning their production and use, as well as generate large-scale remediation efforts of these forever chemicals.

With the increasing number of research articles published on PFAS in the last two decades and the advancement of technology, one can be assured that new PFAS soil remediation techniques will likely be developed to protect our agroecosystem. We believe that eradication of PFAS within agroecosystems should focus on the elimination of these substances within mediums, while simple reductions in their usage may still lead to lower concentrations in agricultural mediums; their presence in these systems will still result in long-term harm to environmental and human health. The average consumer lacks control over the reduction of PFAS before their entry into the environment, given that many potential sources of PFAS originate from industrial products, processes, and applications; for this reason, reductions in PFAS cannot fall onto the onus of the consumer. While decreasing PFAS usage marks a positive step, failing to prevent their release into the environment effectively shifts the burden of remediation onto future generations.

AUTHOR CONTRIBUTIONS

Jackson C. Verley: Conceptualization; data curation; formal analysis; investigation; methodology; software; supervision; validation; writing—original draft; writing—review and editing. **Everald McLennon:** Investigation; supervision; visualization; writing—original draft. **Kathleen S. Rein:** Data curation; investigation; visualization; writing—original draft; writing—review and editing. **Johanne Dikgang:** Writing—original draft. **Vanaja Kankarla:** Conceptualization; data curation; formal analysis; funding acquisition; investigation; methodology; project administration; resources; supervision; validation; visualization; writing—original draft; writing—review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

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