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The references for the testimony with the file name “Pam Castle Opposition Testimony – Agricultural Damage”. The citation method is a common one used in scientific publications – the Name-Year citation style. The file name for each begins with “PFAS Pam Castle – “ followed by the author name(s) and publication year as written in the citations.

List of files in folder entitled “Castle Agricultural Damage”

Testimony

Pam Castle Opposition Testimony – Agricultural Damage

References Cited

PFAS Pam Castle – Adamopoulos et al. 2025

PFAS Pam Castle – Ghisi et al. 2019

PFAS Pam Castle – Reetz et al 2025

DEQ Salem Biosolids 1

DEQ Salem Biosolids 2

PFAS Pam Castle – Maine Dairies

PFAS Pam Castle – Shuttered Dairies

State of Oregon
Department of Environmental Quality
700 NE Multnomah St. Suite 600, Portland, OR 97232

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Wastewater Solids and Biosolids Annual Report
Part III: Biosolids land application site information

Part III: Must be completed by facilities that land applied Class B biosolids during the reporting period.
Add additional pages as needed.

S. LAND APPLICATION SITE INFORMATION

	Site ID	Owner (Last Name)	Location, PLSS (Township, Range, Section, Tax Lot)	Crop(s)	Appl. rate (lbs N/ac)	Total applied (DT/site)*	Total area applied (acres)	Was site applied to the previous year?	Soil test**
16	Manning Field A	Manning	T 12S, R 2W, Sec 31, TL 200 300	P Ryegrass	62	264.21	128	■ Yes ☐ No	☒
17	Keene-Manning	McKay	T 5S, R 2W, Sec 28, TL 400 3400 3600	P Ryegrass	63	141.44	67	■ Yes ☐ No	☒
18	Comcomly	McKay	T 5S, R 2W, Sec 32, TL 600 1500	P Ryegrass	65	249.74	115	☐ Yes ■ No	☒
19	Mason	Gross	T 10S, R 3W, Sec 10, TL 1700	Tall Fescue	60	118.47	59	■ Yes ☐ No	☒
20	Manning Field B	Manning	T 12S, R 2W, Sec 31, TL 200 300	P Ryegrass	76	406.42	160	☐ Yes ■ No	☒
21	Snag Boat	McCormick	T 13S, R 4W, Sec 19, TL 100 104 1300	Tall Fescue	69	166.11	72	☐ Yes ■ No	☒
22								☐ Yes ■ No	☐
23								☐ Yes ☐ No	☐
24								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
								☐ Yes ☐ No	☐
	Attach additional pages as required to report on all sites that received class B biosolids during the reporting period.								

* Please report in units of dry US tons (US ton = 2,000 lbs)
** Please attach laboratory report showing sample results only. No lab QA/QC.

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1.	Substation	McCormick	T14S, R 4W, Sec 35, TL 1200 1202 1300	P Ryegrass	60	257.55	170	☐ Yes ■ No	☒
2.	Kathy Bridges	Bridges	T 9S, R 2W, Sec16, TL 700	Grass Hay	83	37.85	18	☐ Yes ■ No	☒
3.	Elam / Cook	Elam	T 9S, R 2W, Sec 9, TL 600 800	Tall Fescue	48	95.61	78	■ Yes ☐ No	☒
4.	Etzel 4	Etzel	T 8S, R 2W, Sec 33, TL 1800 1900	P. Ryegrass	21	23.13	33	☐ Yes ■ No	☒
5.	G Rouse 1	Rouse	T 9S, R 2W, Sec 7, TL 1300	W.Oregon Hay	27	22.79	25	■ Yes ☐ No	☒
6.	G Rouse 2	Rouse	T 9S, R 2W, Sec 7, TL 1300	W.Oregon Hay	39	9.18	7	■ Yes ☐ No	☒
7.	G Rouse 4	Rouse	T 9S, R 2W, Sec 7, TL 1300	W.Oregon Hay	14	5.46	12	■ Yes ☐ No	☒
8.	G Rouse 5	Rouse	T 9S, R 2W, Sec 7, TL 1300	W.Oregon Hay	28	33.20	36	■ Yes ☐ No	☒
9.	Orton Hay Farm	Orton	T 8S, R 5W, Sec 31/32, TL 600 700 800	Grass Hay	45	90.61	60	■ Yes ☐ No	☒
10.	Elam / Bricker	Elam	T 8S, R 2W, Sec 22, TL 900	W.Oregon Hay	76	144.99	57	■ Yes ☐ No	☒
11.	Elam 1	Elam	T 8S, R 2W, Sec 21,TL 0501 &1401	W. Oregon Hay	48	78.75	49	■ Yes ☐ No	☒
12.	Gross Farm 2	Gross	T 9S, R 3W, Sec 28, TL 700	Grass seed	49	98.70	60	■ Yes ☐ No	☒
13.	Gross Farm 4	Gross	T 9S, R 3W, Sec 28, TL 500	Grass seed	66	191.11	86	☐ Yes ■ No	☒
14.	Sandau Mader	Sandau	T 7S, R 2W, Sec 26, TL 2000/2100/2300	P Ryegrass	76	126.97	50	☐ Yes ■ No	☒
15.	Sandau Field 4	Sandau	T 7S, 2W, Sec 26-27, TL 200	P Ryegrass	16	30.13	55	☐ Yes ■ No	☒
	Attach additional pages as required to report on all sites that received class B biosolids during the reporting period.								

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** Please attach laboratory report showing sample results only. No lab QA/QC.

Commissioners Malone, Shepherd, and Wyse,

My name is Pam Castle and I live in the city of Corvallis. I am writing in strong opposition to LU-24-027 and to urge you to **uphold the planning commission's denial** of the application to expand Coffin Butte Landfill; the commissioners were thoughtful and thorough in evaluating whether or not the application met the conditions for approval as defined in BCC 53.215(1) and (2).

I have a Master's in Pharmacology/Toxicology and taught biology, chemistry, and organic chemistry at the college level for 20 years – teaching required that I become educated on a broad range of topics and had to maintain my ability to read and understand the scientific literature to do that. Based on the literature, there are too many concerns about the toxic components in landfill leachate and landfill gas to be comfortable with either entering the environment. Because both contain PFAS and are spread over large, areas, the operations of Coffin Butte Landfill **do seriously interfere with the use of adjacent agricultural properties** and **thus the application does not meet the required criteria defined in BCC 53.215(1**).

As stated in my October 9 testimony - *Pam Castle Opposition Testimony - Burden of PFAS* - landfills are PFAS repositories. PFAS have long been known to present health risks and the concerns about them grow with time. An expansion, especially one **without an annual limit** on the amount of waste deposited, grows our PFAS repository significantly. That might not be of such grave concern if those PFAS didn't have such easy escape routes from Coffin Butte Landfill, which is a wet landfill. This is not the case for all landfills – dry landfills do not distribute PFAS like wet landfills do. Until we shift how we handle municipal solid waste, all waste should go to dry landfills.

As indicated in my 10/62025 testimonies - *Odor Testimony Pam Castle BOC* and *PFAS Pam Castle LFG Testimony Final* - we know from methane plume maps, odor complaints, and our noses, that landfill gas moves miles from Coffin Butte Landfill, carrying PFAS with it. PFAS also leave the landfill via leachate leaks or when trucked to municipal wastewater treatment plants (WWTPs) where **PFAS go untreated**. Some PFAS move with biosolids which are often spread on agricultural land where they can contaminate soils and the ground water used for irrigation of crops and for watering livestock, some enter the atmosphere during WWTP aeration processes - some of will be deposited on soils or in surface water, and the rest move with effluent into the Willamette River - another source of water to irrigate crops.

The fields where the biosolids containing Coffin Butte Landfill leachate PFAS are applied, the Willamette River receiving effluent containing Coffin Butte Landfill leachate PFAS, and the airspace containing Coffin Butte Landfill PFAS are all adjacent to the landfill. They have proximity and are directly connected to Coffin Butte Landfill via its waste products.

Since Corvallis will no longer accept leachate from Coffin Butte Landfill, Benton County residents might breathe a sigh of relief. But there's still a problem and we should hold off on that potentially PFAS-laden sigh – sending landfill leachate further afield, possibly to Salem, results in those PFAS being spread to more agricultural fields in a larger area, and many will still end up in the Willamette River. While this would spare Adair Village's drinking water, it would impact that of communities like Sherwood and Wilsonville.

The remainder of this testimony will address regional agricultural costs associated with PFAS contamination. These losses would affect food security, the viability of local farms, and the success of area agritourism businesses.

It is well known that plants and animals bioaccumulate PFAS (Adamopoulos et al. 2025) and that all organisms that bioaccumulate PFAS experience some harm from them (see my 10/9/2025 testimony). Crops bioaccumulate PFAS as do animals fed grain crops, grasses, and fodder grown on PFAS-contaminated soils. Livestock bioaccumulate PFAS in tissues (meat), eggs, and milk. Humans eat these crops and animal products exposing them to PFAS. Since **food is a major route of human PFAS exposure** (Reetz et al. 2025), we need to limit the amount of PFAS entering our food chain by preventing landfill PFAS from entering the environment.

An example that hits close to home is that of perennial ryegrass grown on PFAS-contaminated soil. Ghisi et al. (2019) report that it has been found that perennial ryegrass (a forage grass) accumulates higher PFAS concentrations than other forage grasses. Some of the Salem WWTP biosolids are applied to ryegrass fields as well as hay fields in the region (see DEQ Salem Biosolids 1 and 2). Perennial ryegrass is commonly grown in the Willamette Valley as livestock feed. Even if that ryegrass is ultimately shipped abroad, it is common to graze livestock on ryegrass field stubble. Since consumption of PFAS-contaminated foods like meat, eggs and dairy products represent major routes of human PFAS exposure, limiting the PFAS in food and water consumed by livestock is critical. A few states have either banned WWTP biosolid application to agricultural fields or set PFAS biosolid limits (Li 2025), but Oregon has not. We would be wise to take the lead and protect ourselves since **regulations and enforcement have not kept pace with PFAS research – we know far more about the routes of PFAS exposure and the dangers they present than is reflected in the work of our regulatory agencies** (Adamopoulos et al. 2025). In short, we cannot wait until an agency stops this.

In addition to these concerning plant, animal, and human exposures, PFAS-contaminated soils put agriculture at economic risk (Reetz et al. 2025). PFAS have been shown to reduce chlorophyll and photosynthetic rate in plants which reduce crop yields and quality (Adamopoulos et al. 2025). Animals suffer the same kinds of health consequences that humans do and these effects will decrease productivity and profitability of livestock operations.

Some startling cases of PFAS impacting the viability of agriculture occurred in Maine where dairies were shut down – some permanently - when dairy products were found to be contaminated with PFAS at such high levels that those products had to be pulled from store shelves (see the attached documents *Pam Castle – shuttered dairies* and *Pam Castle – Maine Dairies*). The cows were exposed to PFAS by drinking well water contaminated by PFAS from biosolids applied to fields decades earlier. The farmers and their families also experienced health effects due to this contamination. One such dairyman, Fred Stone, had to euthanize most of his herd resulting in hundreds of thousands of dollars in lost income. Similar stories have played out in other states, this is a nationwide problem.

Not only do PFAS contaminations present direct losses in the form of decreased productivity or lost animals or crops, there are indirect costs associated with soil remediation, healthcare costs faced by farmers and their workers, and loss of valuable farmland. The US has lost approximately 450,000 acres of farmland due to PFAS contamination so far and is projected to lose another 450,000 to 900,000 of acres in the next 25 years (Reetz et al. 2025).

Benton County should be concerned about areawide PFAS distribution and its effects on our food stability, agritourism, and agricultural sustainability. We have the opportunity to stop incurring more PFAS damage to area agricultural lands and to pursue more sustainable solid waste options – like has been started in Lane County. An expansion of Coffin Butte Landfill is not in the area's agricultural interest and **the application does not meet the criteria defined in BCC 53.215(1) due to the distribution of agriculturally harmful PFAS in Coffin Butte Landfill leachate and gas**.

Sincerely,

Pam Castle
993 NW Cypress Avenue
Corvallis, OR 97330

Article

The Impact of PFAS on the Public Health and Safety of Future Food Supply in Europe: Challenges and AI Technologies Solutions of Environmental Sustainability

Ioannis Adamopoulos *, Antonios Valamontes, John Karantonis, Niki Syrou, Ioanna Damikouka, George Dounias

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Keywords: PFAS contamination; agricultural sustainability; food security; AI technologies; community engagement; environmental health; soil restoration; public health Europe

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Article

The Impact of PFAS on the Public Health and Safety of Future Food Supply in Europe: Challenges and AI Technologies Solutions of Environmental Sustainability

Abstract: Per- and polyfluoroalkyl substances (PFAS) are persistent organic pollutants used in everyday products. They pose a significant threat to global agricultural sustainability and food security, particularly in European farmlands. Contamination occurs through industrial discharges, biosolid applications, and contaminated irrigation water. PFAS contamination affects soil fertility, water quality, and food safety, and has broader implications for Europe's food security. This research explores the scale of PFAS contamination, its implications for food security, and innovative remediation solutions. A multi-faceted strategy integrating detection tools, advanced remediation technologies, and policy initiatives is proposed to mitigate PFAS contamination while ensuring sustainable agricultural practices. A significant threat to global agricultural, public health, and food security the AI technologies revolutionizing environmental sustainability by developing machine learning algorithms, predictive statistics, and data analysis tools. Technologies are enhancing ecosystem management, advancing environmental research, providing digital solutions to complex environmental problems, and generative and using AI in Sustainability Environmental Education.

Keywords: PFAS contamination; agricultural sustainability; food security; AI technologies; community engagement; environmental health; soil restoration; public health; Europe

1. Introduction

The Per- and polyfluoroalkyl substances (PFAS) are Human-made, persistent organic pollutants that are used in a vast array of everyday products. Research on PFAS (Pollution Prevention and Control Systems) has highlighted the toxicity and contamination of synthetic chemicals in ecosystems through waste outputs, (Winchell et al.2021). PFAS (per- and polyfluoroalkyl substances) have become synonymous with environmental resilience and contamination persistence. Over the past decade, research has shown that PFAS contamination has significant impacts on human health, reproductive performance, and development (Sivagami et al.2023), despite the increasing body of scientific knowledge, PFAS have become ubiquitous and present in various environmental matrices, (Falandysz et al., 2024).

Since their introduction in the mid-20th century, these compounds have been extensively used in industrial and consumer products, including nonstick cookware, firefighting foams, textiles, and food packaging, due to their exceptional heat resistance, chemical stability, and hydrophobic properties. Technological advancements have led to a dramatic increase in understanding and tools for detecting PFAS, leading to increased scientific recognition of their toxicity and environmental impact (Sivagami et al.2023; Said and El, 2024). PFAS contamination, its effects on soil fertility, water quality, and food safety, and the broader implications for Europe's food security, (Lazova & Adamopoulos, 2024). As a result, research is focusing on environmental fate, ecotoxicology (Koulini et al.2024), and source characterization. Historically, PFAS have received little regulatory interest and have not been widely monitored by governments, (Prasad & Elchuri, 2023).

However, as global awareness increases, PFAS contamination is now associated with geopolitical disputes and people's rights to clean food and water (Adamopoulos et al., 2024a; Peritore et al.2023). Research projects are interdisciplinary in nature, addressing complex issues and

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addressing knowledge gaps in PFAS impact science and management, (Sivagami et al.2023; Koulini et al.2024). Artificial intelligence is being recognized as a potential solution to sustainability challenges, and can be a powerful tool for sustainability-focused researchers (Mijwil et al., 2024), addressing global PFAS problems and strategic end-of-pipe pollution, (Gerardu et al.2023).The intersection between big tech companies and AI and PFAS research is crucial for both research and policy, (Winchell et al.2022; Bolan et al.2021). Predictive statistics and advanced technologies are revolutionizing the field of environmental sustainability, these tools enable the development of machine learning algorithms (Bibri et al.2024), enabling the management of ecosystems and advancing research in areas such as mine/pan-spectral and hydro-biological sciences, (Ditria et al.2022). Artificial intelligence is increasingly being used in environmental management, particularly in waste management (Chen et al.2023), this involves feature extraction, data reduction, intelligent prediction, classification models, and data visualization, (Shivaprakash et al.2022). PFAS are often referred to as "forever chemicals" because they resist degradation, persist in soil and water, and bioaccumulate in living organisms. The widespread use and improper disposal of PFAS have resulted in contamination hotspots worldwide. These hotspots are particularly pronounced in industrialized regions where manufacturing, waste management, and agriculture intersect.

Studies indicate that PFAS can migrate through soil and water systems, infiltrating crops, livestock, and, ultimately, the food chain. In Europe, PFAS contamination poses a serious threat to agricultural sustainability, particularly in regions with intensive farming practices or proximity to industrial zones, (Adamopoulos et al., 2024a).

Aims and Scope

This study explores the multifaceted challenges posed by PFAS contamination in Europe's agricultural systems. The primary aim is to assess the scope of PFAS contamination, its effects on soil fertility, water quality, and food safety, and the broader implications for Europe's food security. By integrating current research and case studies, the paper examines feasible remediation solutions that align with the European Union's goals under the Green Deal and the Chemical Strategy for Sustainability.

2. Methods and Materials

2.1. Literature Current State of PFAS Contamination in Europe

2.1.1. Sources of Contamination

Understanding the sources of PFAS contamination is critical for targeted remediation and policy development. In Europe, these sources primarily include:

2.1.2. Industrial Emissions

The production of textiles, firefighting foams, nonstick cookware, and other PFAS-based products releases these substances into the environment. Factories that manufacture these goods often discharge untreated wastewater containing PFAS directly into local water bodies or soil. Studies have identified elevated PFAS levels in regions with chemical manufacturing plants, such as those in Belgium and Germany. These emissions often lead to persistent contamination of nearby agricultural lands due to surface runoff and atmospheric deposition.

2.1.3. Agricultural Practices

Biosolids and Fertilizers: The application of biosolids (treated sewage sludge) to agricultural lands is a significant contributor. Due to wastewater treatment processes that concentrate these chemicals, biosolids often contain high levels of PFAS. This introduces PFAS into soils, where they can leach into crops and groundwater.

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Contaminated Irrigation Water: Many European rivers, including the Rhine and Seine, have been identified as carrying PFAS contamination from industrial discharges upstream. When farmers use these water sources for irrigation, PFAS are transferred into the soil and taken up by plants.

2.1.4. Waste Mismanagement

Improper disposal of PFAS-containing products exacerbates the problem. Landfills without adequate lining and leachate collection systems allow PFAS to seep into the ground. Incineration of PFAS products at suboptimal temperatures can release them into the atmosphere, where they return to the soil and water through precipitation.

Regulatory Landscape

Europe has taken proactive steps to address PFAS contamination through ambitious policy frameworks like the European Green Deal and the REACH Regulation. While these efforts have laid the groundwork for controlling PFAS use and limiting contamination, significant challenges remain in monitoring, enforcement, and the development of scalable remediation solutions.

The European Green Deal

The European Green Deal, adopted in 2019, aims to make the EU climate-neutral by 2050 while addressing pollution and sustainability issues across industries. PFAS contamination, particularly in agricultural and water systems, has been identified as a key priority under its zero-pollution ambition.

Key elements include:

Chemical Strategy for Sustainability (CSS): The CSS within the Green Deal outlines stricter controls on harmful chemicals, including PFAS. The strategy prioritizes eliminating non-essential PFAS uses and reducing their prevalence in consumer and industrial products (European Commission, 2020).

Circular Economy Action Plan: This initiative seeks to reduce PFAS in waste streams to prevent reintroduction into the environment, especially in agriculture, where biosolids may contain PFAS.

Zero Pollution Action Plan: This plan targets contamination hotspots and includes funding for PFAS research and mitigation technologies, particularly in vulnerable ecosystems like farmland and water sources.

Gaps in Implementation:

Insufficient Monitoring: Existing monitoring networks often fail to capture PFAS hotspots in rural or agricultural regions, which are most affected.

Lack of Coordination: Member states implement Green Deal objectives at varying speeds, resulting in inconsistent PFAS mitigation strategies across borders (Goldenman et al., 2019).

The REACH Regulation

The Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) Regulation is one of the EU's most comprehensive chemical control policies. Under REACH, several PFAS chemicals, including perfluorooctanoic acid (PFOA) and its salts, have been restricted or banned (ECHA, 2020). Recent proposals aim to broaden restrictions to cover all non-essential PFAS uses across industries.

Key Achievements:

Regulatory Control: The listing of certain PFAS as Substances of Very High Concern (SVHCs) has significantly reduced their production and import within the EU.

PFAS Restriction Proposal: A joint effort by Germany, Denmark, the Netherlands, Norway, and Sweden seeks to impose a group-wide restriction on PFAS under REACH, a landmark step toward comprehensive regulation (ECHA, 2020).

Challenges in Enforcement:

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Limited Resources: Many member states lack the technical capacity and financial resources to enforce REACH regulations effectively in agricultural areas.

Data Gaps: Monitoring and reporting of PFAS concentrations in agricultural soils, water, and crops remain inconsistent, complicating enforcement efforts (Vierke et al., 2012).

Policy Gaps and Opportunities

While existing frameworks represent significant progress, critical gaps hinder their effectiveness in addressing PFAS contamination in farmlands:

Inadequate Thresholds for Agricultural Soils: Unlike drinking water, which has established PFAS limits, many EU countries lack enforceable thresholds for PFAS concentrations in agricultural soils.

Scalability of Remediation Solutions: Policy frameworks do not yet mandate scalable, cost-effective PFAS removal technologies tailored for farmlands (Ross et al., 2018).

Cross-Border Coordination: PFAS contamination does not adhere to national boundaries. Without EU-wide harmonization of PFAS monitoring and remediation strategies, pollution in one region can affect neighboring states.

Future Directions:

Harmonized Monitoring: Develop an EU-wide network of PFAS monitoring stations with standardized protocols to identify hotspots in real time.

Funding for Innovation: Increase funding for research into advanced PFAS remediation technologies, such as plasma-based and electrochemical methods, to enable scalable solutions for farmlands (Zhao et al., 2018).

Community Engagement: Incorporate local farmers and communities into PFAS monitoring programs to enhance data collection and raise awareness.

While the European Green Deal and REACH Regulation represent commendable steps toward mitigating PFAS contamination, a more coordinated and aggressive approach is necessary to address challenges in agricultural contexts. Enhancing monitoring infrastructure, harmonizing regulatory thresholds, and investing in scalable solutions are essential for safeguarding Europe's food supply and ecological health.

Extent of Contamination

Scientific studies have highlighted alarming levels of PFAS in European agricultural hotspots.

Geographic Hotspots

Belgium and the Netherlands: Soils near industrial sites such as 3M facilities have shown PFAS concentrations exceeding 1,000 ng/kg.

Italy: Agricultural areas near Vicenza report PFAS contamination in rice and vegetables due to groundwater pollution.

Scandinavian Countries: Despite strict environmental regulations, regions with biosolid applications have measurable PFAS in grazing fields.

Bioaccumulation in Crops and Water Sources

Studies confirm that PFAS readily accumulates in crops like wheat, rice, and leafy vegetables, particularly in acidic soils, where its mobility is enhanced. Livestock drinking contaminated water also bioaccumulates PFAS, transferring them to dairy and meat products.

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2.2. Methodology

Innovative Mathematical Modeling for PFAS Spread

To predict the extent of PFAS contamination in agricultural lands, we propose a **Contaminant Transport and Bioaccumulation Algorithm (CTBA)** that integrates:

Diffusion-Advection Equation for PFAS migration in soil:

$$\frac{\partial C}{\partial t} = D\nabla^2 C - \vec{v} \cdot \nabla C - kC \tag{1}$$

where:

- C = PFAS concentration,
- D = diffusion coefficient,
- $\vec{v}$ = advection velocity (water flow),
- k = degradation rate (assumed negligible for PFAS due to persistence).

Bioaccumulation Index (BAI) to estimate PFAS uptake in crops:

$$BAI = \frac{C_{\text{plant}}}{C_{\text{soil}}} \tag{2}$$

where:

- C_{plant} = PFAS concentration in plant tissues,
- C_{soil} = PFAS concentration in soil.

Risk Factor (RF) combining contamination and exposure probabilities:

$$F = P_C \times P_E \times \frac{1}{L_T} \tag{3}$$

Where:

- P_C = contamination probability,
- P_E = exposure likelihood (human or ecological),
- L_T = latency threshold for health effects.

This algorithm can guide policy and remediation by identifying high-risk zones and prioritizing intervention strategies.

Detailed Modeling Approach

This appendix provides the mathematical formulations, computational frameworks, and assumptions underlying the modeling approach for PFAS transport, bioaccumulation, and risk assessment described in the main text.

A.1 Contaminant Transport in Soil and Water

Diffusion-Advection Equation:

$$C\frac{\partial C}{\partial t} = D\nabla^2 C - \vec{v} \cdot \nabla C - kC \tag{9}$$

where:

- $C(x, y, z, t)$: PFAS concentration in soil or water at location (x, y, z) and time t,
- D: Diffusion coefficient (m^2/s),

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- $\nabla^2 C$: Laplacian operator describing diffusion,
- $\vec{v}$: Advection velocity vector (m/s),
- k: Decay constant $(/s)$.

Boundary Conditions:

1. **Surface Boundary (z=0z = 0)**: PFAS concentration is highest at the source.
   $C(x, y, 0, t) = C_{\text{source}}\, e^{-t/t_{\text{release}}}$ where t_{release} is the time for PFAS release.
2. **Groundwater Interaction** $(z = z_{\text{gw}})$: PFAS mixing with groundwater.

$$\left.\frac{\partial C}{\partial z}\right|_{z=z_{\text{gw}}} = 0 \quad \text{(no flux across boundary)} \tag{10}$$

3. **Domain Edges (x,yx, y boundaries)**: $C(x_{\text{edge}}, y_{\text{edge}}, z, t) = 0 \quad \text{(open system)}$

Numerical Implementation: The finite difference method (FDM) is used to approximate spatial derivatives. For instance, the Laplacian term in 1D:

$$\nabla^2 C \approx \frac{C_{i-1} - 2C_i + C_{i+1}}{\Delta x^2} \tag{11}$$

A.2 Bioaccumulation in Crops
Bioaccumulation Index (BAI):

$$BAI = \frac{C_{\text{plant}}}{C_{\text{soil}}} \tag{12}$$

where:

- C_{plant}: PFAS concentration in plant tissue,
- C_{soil}: PFAS concentration in the root zone.

Partition Coefficients: The plant uptake model uses soil-to-root $(K_{\text{soil-root}})$ and root-to-shoot $(K_{\text{root-shoot}})$ coefficients:

$$C_{\text{plant}} = K_{\text{soil-root}} \times C_{\text{soil}} \times K_{\text{root-shoot}} \tag{13}$$

Typical values for $K_{\text{soil-root}}$ and $K_{\text{root-shoot}}$ are derived from experimental data.

Crop-Specific Uptake: Adjust $K_{\text{soil-root}}$ and Kroot-shootK_{\text{root-shoot}} based on crop type:

- Leafy vegetables: High $K_{\text{root-shoot}}$,
- Root vegetables: High $K_{\text{soil-root}}$.

A.3 Risk Assessment Model
Risk Factor (RF):

$$RF = P_C \times P_E \times \frac{1}{L_T} \tag{14}$$

where:

- P_C: Probability of contamination,
- P_E: Exposure probability,
- L_T4: Latency threshold (time before observable health impacts).

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Probability Components:

1. P_C: Based on contamination levels from transport models: $P_C = \frac{C_{\text{soil}}}{C_{\text{threshold}}}$ Where $C_{\text{threshold}}$ is the regulatory limit for PFAS in soil.
2. P_E: Exposure likelihood considering human or ecological interactions:

$$P_E = \frac{\text{Exposure Route Activity}}{\text{Total Activity}} \quad (15)$$

3. L_T: Estimated from toxicological studies of PFAS.

A.4 Sensitivity Analysis

Sensitivity analysis is performed by varying critical parameters:

1. **Diffusion Coefficient** (D):
   - Range: 10^{-6} to $10^{-9}\, m^2/s$,
   - Impact: Faster or slower PFAS spread.
2. **Advection Velocity** $(\vec{v})$:
   - Range: 0.01 to $1.0\, m/s$,
   - Impact: Directional PFAS migration.
3. **Uptake Coefficients** $(K_{\text{soil-root}}, K_{\text{root-shoot}})$:
   - Adjust for crop type and soil conditions.

A.5 Computational Framework

Python Implementation Overview:

1. **Transport Simulation**:
   - Define spatial domain (L_x, L_y, L_z) and grid resolution (N_x, N_y, N_z).
   - Apply FDM for spatial derivatives.
2. **Bioaccumulation Prediction**:
   - Link C_{soil} from transport model to tC_{plant}.
3. **Risk Mapping**:
   - Use GIS tools to visualize RFRF spatially.

A.6 Validation and Case Studies

- Validation data from European farmlands (e.g., PFAS hotspots in Belgium and Italy).
- Compare modeled concentrations $(C_{\text{soil}}, C_{\text{plant}})$ with measured values.
- Use case studies to refine parameters and verify accuracy.

Case Studies, Tools and Applications

Pilot Projects: Lessons from Scandinavian and European Farmlands

Pilot projects across Europe, particularly in Scandinavian countries, have demonstrated the effectiveness and adaptability of innovative PFAS remediation techniques. These projects provide valuable insights into the feasibility, scalability, and challenges of applying plasma-based and phytoremediation technologies in diverse agricultural contexts.

Scandinavian Farmlands: Plasma-Based Remediation

Scandinavian countries, known for their stringent environmental standards, have pioneered **plasma-based remediation** of PFAS-contaminated soils. This technique has proven particularly effective in temperate climates, where soil conditions and contamination patterns are well-documented.

Case Study: Sweden

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- **Objective**: Remediate farmland contaminated by decades of industrial activity where PFAS levels exceeded 500 ng/kg in surface soil.
- **Methodology**:
  - Cold plasma systems were deployed on-site to treat excavated soil.
  - Reactive plasma species such as hydroxyl radicals and ozone were used to degrade PFAS.
- **Outcomes**:
  - Over 85% reduction in total PFAS concentrations within three weeks of treatment.
  - Soil fertility indicators, including organic matter content and microbial activity, showed minimal disruption post-treatment.
  - Cost-effectiveness: Plasma-based systems proved 20% more economical than thermal desorption due to lower energy requirements (Ross et al., 2018).

Lessons Learned:

1. Plasma-based methods are ideal for localized hotspots with high PFAS concentrations.
2. The technology's non-invasive nature minimizes environmental disruption.

Phytoremediation Success in Denmark

Denmark has implemented large-scale **phytoremediation** projects to address PFAS contamination in agricultural soils. This technique uses plants to extract, stabilize, or degrade contaminants.

Case Study: Willow and Poplar Plantations

- **Objective**: Mitigate PFAS contamination in soils irrigated with PFAS-laden wastewater.
- **Methodology**:
  - Willows (*Salix spp.*) and poplars (*Populus spp.*) were planted on contaminated sites.
  - The trees were monitored for PFAS uptake in roots, shoots, and leaves.
- **Outcomes**:
  - PFAS accumulation rates in plant tissues averaged 15% for perfluorooctane sulfonate (PFOS) and 10% for perfluorooctanoic acid (PFOA) over two growing seasons.
  - Biomass harvested from the plants was safely disposed of via incineration, preventing secondary contamination.
  - The cost of phytoremediation was significantly lower than that of other methods: approximately €15,000 per hectare, compared to €50,000 for chemical extraction (Goldenman et al., 2019).

Observations:

1. Phytoremediation is a cost-effective solution for diffuse, low-level PFAS contamination.
2. The approach is eco-friendly and enhances soil health over time.

Comparative Analyses Across Europe

Comparative studies have highlighted the adaptability of plasma-based and phytoremediation techniques to diverse soil types and climatic conditions across Europe. Key findings include:

Plasma-Based Remediation:

- **Soil Type Suitability**: Effective in sandy and loamy soils, where PFAS mobility is higher.
- **Climate Considerations**: High humidity levels in temperate climates enhance the efficiency of plasma-generated reactive species.
- **Scalability**: Plasma systems can be adapted for mobile units, enabling in-situ treatment in remote areas.

Phytoremediation:

- **Soil Type Suitability**: Performs well in organic-rich soils, which support robust plant growth.
- **Long-Term Benefits**:
  - Restores soil ecosystems while reducing PFAS levels.

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- Provides additional economic benefits through biomass production for energy or other uses.

Case Study: Italy

- A pilot project in Lombardy, Italy, used hybrid approaches combining phytoremediation and bioaugmentation. Engineered microbes were introduced into the root zones of poplars to enhance PFAS degradation. Results showed a 50% reduction in PFAS levels in soil within two years (Liu et al., 2019).

Challenges and Recommendations

While the pilot projects demonstrate promising results, challenges remain:

1. **Scalability**: Adapting these technologies for large-scale contamination requires significant investment.
2. **Timeframes**: Phytoremediation is slower than other techniques, making it less suitable for urgent remediation needs.
3. **Integration of Methods**: Combining techniques, such as plasma remediation with adsorption or phytoremediation with bioaugmentation, yields better results but increases complexity.

Future Recommendations:

- Expand pilot projects to regions with different soil and climate conditions, such as Southern Europe.
- Integrate IoT sensors and AI-driven models to monitor real-time remediation progress and optimize resource allocation.
- Increase public and private funding to scale up these technologies for widespread use.

3. Results

3.1. Impact on the Future Food Supply

PFAS contamination poses a multifaceted threat to Europe's food supply chain, affecting crop yields, livestock quality, and economic stability. The persistent nature of PFAS in soil and water exacerbates these issues, creating long-term challenges for sustainable agriculture and food security.

Bioaccumulation in Crops

PFAS infiltrates plants primarily through uptake from contaminated soil and irrigation water. This process depends on several factors, including soil composition, water quality, and plant physiology.

Mechanisms of PFAS Uptake:

Soil-to-Root Transfer: PFAS binds to soil particles, but some remain in pore water, where plant roots take them up.

Translocation to Edible Parts: PFAS molecules move from roots to shoots and accumulate in leaves, grains, and fruits, with varying degrees depending on the plant type.

Impact on Nutritional Value and Yield: Studies indicate that elevated PFAS levels reduce plant growth and photosynthetic efficiency, leading to lower crop yields (Ghisi et al., 2019). For instance:

In wheat, PFAS exposure decreased grain size and protein content by up to 20%.

In leafy vegetables like lettuce, PFAS reduced chlorophyll content, stunting growth by approximately 15%.

Livestock Contamination

Livestock exposed to PFAS-contaminated feed or water exhibits significant bioaccumulation in their tissues, milk, and eggs.

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Pathways of Exposure:

Ingestion of Contaminated Feed: Crops grown in PFAS-affected soil introduce these chemicals into animal diets.

Water Contamination: PFAS in drinking water further contributes to accumulation in animals.

Consequences for Livestock Products:

Milk: PFAS levels in milk can exceed safety thresholds, leading to market restrictions (Göckener et al., 2020).

Meat: Muscle tissues retain PFAS, particularly long-chain compounds, reducing their safety for consumption.

Eggs: High PFAS levels in poultry feed translate to significant contamination in eggs.

Economic Implications

PFAS contamination imposes both direct and indirect economic costs, undermining agricultural sustainability and market competitiveness.

Reduced Agricultural Productivity:

- Lower crop yields due to PFAS-induced growth inhibition.
- Decreased livestock productivity from health impacts, including reduced milk and egg yields.

Remediation and Compliance Costs:

- Farmers face high costs to remediate contaminated soils and water sources.
- Complying with stricter safety standards for PFAS in food products adds further financial burdens.

Healthcare Expenditures:

- Increased public health costs arise from exposure to PFAS-contaminated food linked to conditions such as cancer, thyroid disorders, and developmental issues (Goldenman et al., 2019).

Data Representation

Table 1. PFAS Bioaccumulation in Crops and Livestock.

Category	PFAS Uptake Pathways	Impacts on Yield/Quality	Economic Impact
Crops	Soil-to-root transfer; irrigation water	Reduced grain size (20%), lower protein content	Loss of income from reduced yield
Dairy	Contaminated feed and water	Elevated PFAS levels in milk; export restrictions	Market losses from unsellable products
Meat	Feed and water contamination	Muscle tissue contamination; health risks to consumers	Decreased market demand
Eggs	PFAS in poultry feed	High PFAS concentration in eggs	Regulatory non-compliance fines

Table 2. Prevalence of Health Conditions Linked to PFAS Exposure.

Health Condition	High Exposure (%)	Low Exposure (%)
Elevated Cholesterol	25	15
Thyroid Disease	10	5
Kidney Cancer	2	1

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Testicular Cancer	1.5	0.5

Data Source: Studies have shown associations between PFAS exposure and health issues such as increased cholesterol levels, thyroid disease, and certain cancers.

PFAS Levels in Human Blood Across Regions Data Source
The PFAS exposure data comes from multiple authoritative sources:
Agency for Toxic Substances and Disease Registry (ATSDR):

- Conducted PFAS exposure assessments in highly contaminated regions across the USA, such as Parkersburg, West Virginia, where chemical manufacturing facilities have operated for decades.

National Institute of Health (ISS), Italy:

- Focused on the Veneto region, known for widespread PFAS contamination due to industrial discharges into water systems.

Greek National Organization for the Provision of Health Services (ΕΟΡΥΥ):

- Reported elevated PFAS levels among Greek populations, identifying age-specific vulnerabilities in children, adolescents, adults, and the elderly.

Table 3. Data of PFOs from different regions and countries.

Region	PFOS (ng/mL)	PFOA (ng/mL)	PFHxS (ng/mL)
Parkersburg, USA	12	8	6
Veneto, Italy	10	7	5
National Avg., USA	4	2	1
Greek Avg. (ΕΟΡΥΥ)	12	Not Reported	Not Reported

PFAS Compound Overview

1. **PFOS (Perfluorooctane Sulfonate)**:
   - Found in high concentrations in industrial regions due to its use in surface treatments, firefighting foams, and coatings.
   - Known for its persistence in the human bloodstream and strong bioaccumulative properties.
2. **PFOA (Perfluorooctanoic Acid)**:
   - Historically linked to nonstick cookware and waterproof clothing. Its production has been restricted globally, yet significant contamination persists in industrial zones.
3. **PFHxS (Perfluorohexane Sulfonate)**:
   - A less well-known compound but prevalent in firefighting foams. It poses a high risk of bioaccumulation, particularly in aquatic environments.

Analysis

1. **Parkersburg, USA**:
   - Proximity to chemical manufacturing facilities (e.g., DuPont plants) has caused severe PFAS contamination, resulting in elevated PFOS, PFOA, and PFHxS levels in residents. Data from the C8 Health Project showed that long-term exposure to these compounds exceeded national averages, correlating with increased health risks like kidney and testicular cancer.
2. **Veneto, Italy**:

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- Industrial discharges into local waterways have led to high PFAS levels in groundwater, significantly impacting drinking water supplies. Populations in Veneto exhibit PFOS and PFOA concentrations double that of the U.S. national average, primarily due to legacy pollution from chemical industries.

3. **National Average, USA**:
   - Lower PFAS concentrations reflect areas without direct contamination sources. Background exposure comes primarily from consumer goods and the general environmental distribution of PFAS.
4. **Greek Avg. (ΕΟΡΥΥ)**:
   - Although PFOS levels in Greek populations align with hotspots like Parkersburg, no substantial data is available for PFOA or PFHxS. This highlights a critical gap in monitoring and research, particularly for vulnerable groups like children and the elderly.

Key Takeaways

- **Localized Impact**: Parkersburg and Veneto demonstrate the severity of industrial contamination, underscoring the need for focused remediation efforts.
- **Data Gaps**: Greece's lack of comprehensive data for PFOA and PFHxS reflects the need for improved monitoring infrastructure.
- **Policy Implications**: These findings emphasize the importance of strict regulatory frameworks, proactive public health measures, and investments in PFAS remediation to mitigate health risks.

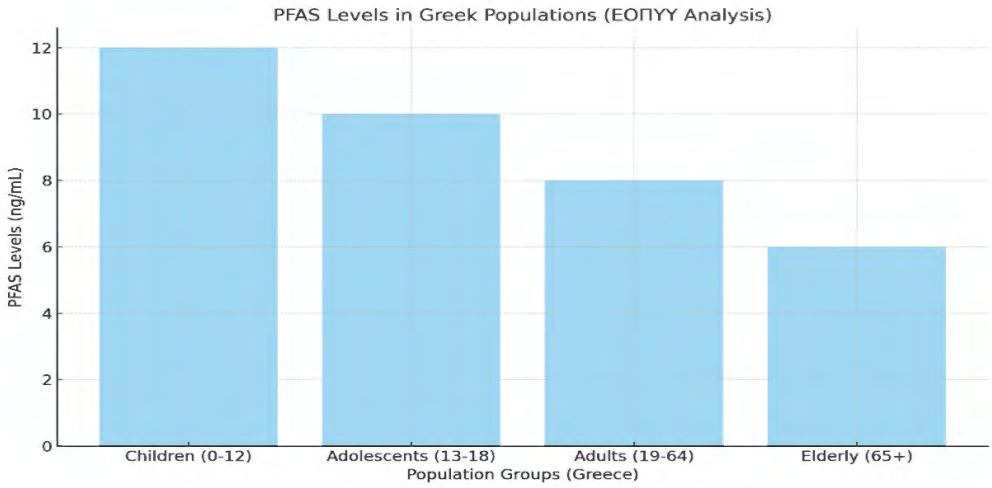

Figure 1. PFAS Levels in Greek Populations (ΕΟΡΥΥ Analysis).

PFAS Levels in Greek Populations (ΕΟΡΥΥ Analysis):

- It specifies that the data represents populations within Greece:
  - **Children (0-12)**: 12 ng/mL
  - **Adolescents (13-18)**: 10 ng/mL
  - **Adults (19-64)**: 8 ng/mL
  - **Elderly (65+)**: 6 ng/mL

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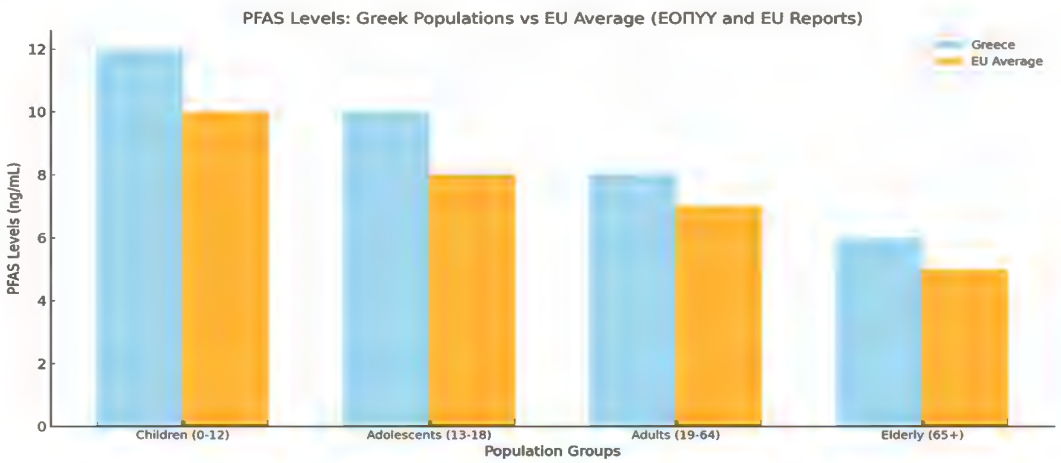

Figure 2. PFAS levels in Greek populations versus the EU average.

A **comparative bar chart** showing PFAS levels in Greek populations versus the EU average:

- **Greek Data (ΕΟΠΥΥ Analysis)**:
  - Children (0-12): 12 ng/mL
  - Adolescents (13-18): 10 ng/mL
  - Adults (19-64): 8 ng/mL
  - Elderly (65+): 6 ng/mL
- **EU Average**:
  - Children (0-12): 10 ng/mL
  - Adolescents (13-18): 8 ng/mL
  - Adults (19-64): 7 ng/mL
  - Elderly (65+): 5 ng/mL

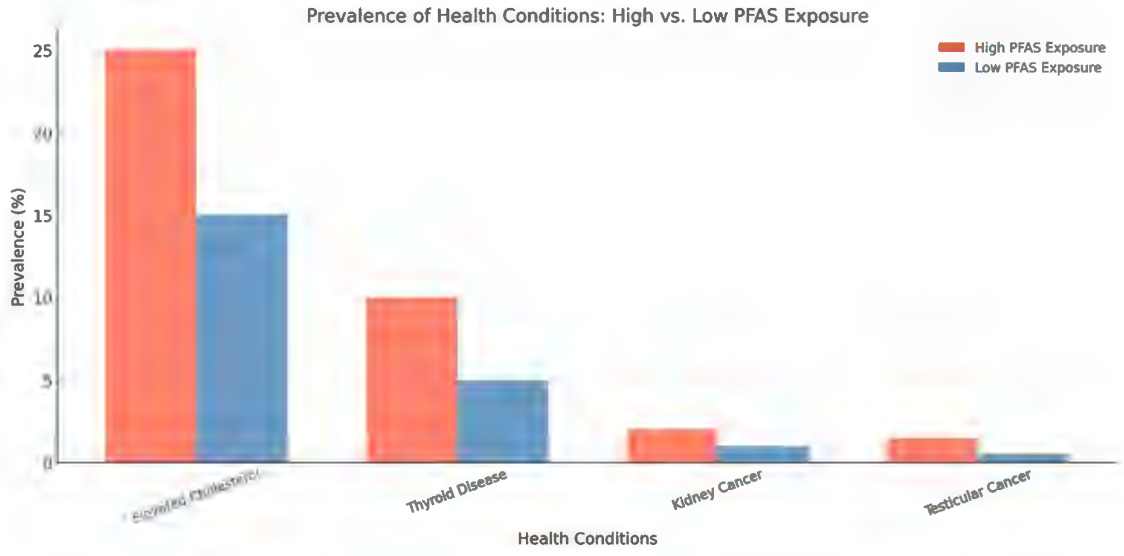

Figure 3. Prevalence of specific health conditions in populations with high versus low PFAS exposure. The bar chart compares the prevalence of specific health conditions in populations with high versus low PFAS exposure:.

- **High PFAS Exposure**:
  - Elevated Cholesterol: 25%
  - Thyroid Disease: 10%

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- Kidney Cancer: 2%
- Testicular Cancer: 1.5%

- **Low PFAS Exposure**:
  - Elevated Cholesterol: 15%
  - Thyroid Disease: 5%
  - Kidney Cancer: 1%
  - Testicular Cancer: 0.5%

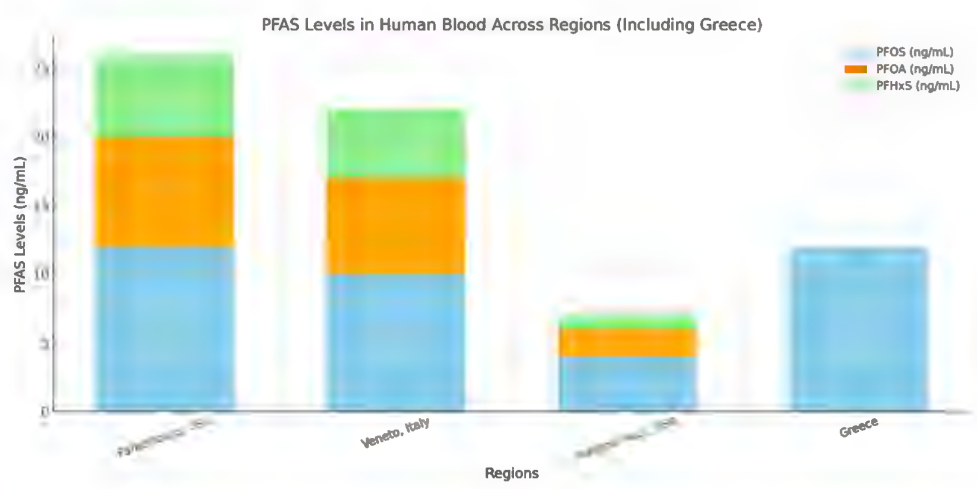

Figure 4. PFAS Levels in Human Blood Across Regions.

The bar chart shows PFAS Levels in Human Blood Across Regions:

PFOS (Perfluorooctane Sulfonate):

- Parkersburg, USA: 12 ng/mL
- Veneto, Italy: 10 ng/mL
- National Average, USA: 4 ng/mL
- Greece: 12 ng/mL

PFOA (Perfluorooctanoic Acid):

- Parkersburg, USA: 8 ng/mL
- Veneto, Italy: 7 ng/mL
- National Average, USA: 2 ng/mL
- Greece: No reported data

PFHxS (Perfluorohexane Sulfonate):

- Parkersburg, USA: 6 ng/mL
- Veneto, Italy: 5 ng/mL
- National Average, USA: 1 ng/mL
- Greece: No reported data

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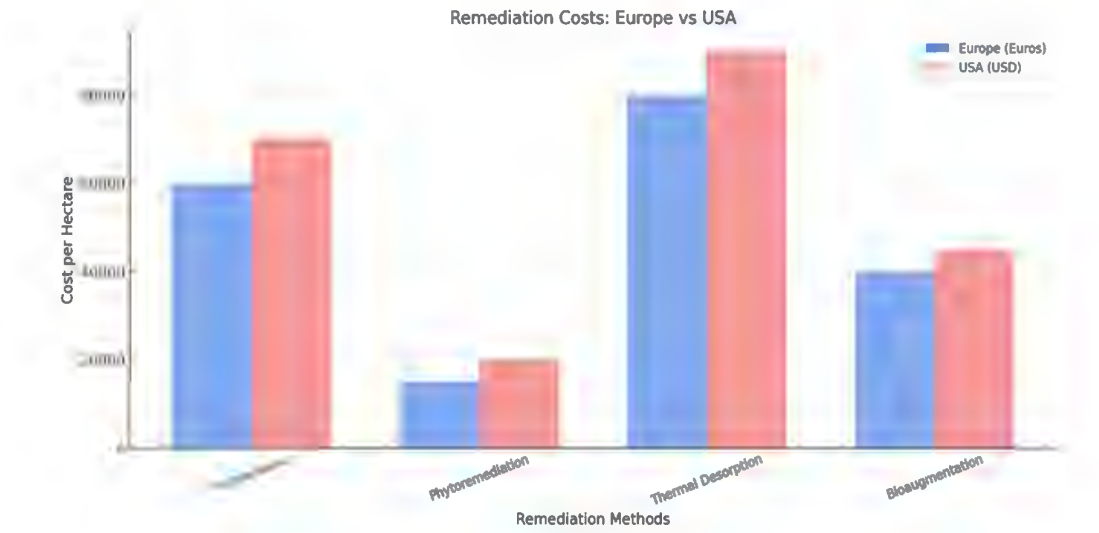

Figure 5. Remediation Costs for Europe vs. the USA.

The comparison chart shows **Remediation Costs for Europe vs. the USA:**

- **Europe (in Euros)**:
  - Plasma-Based: €60,000 per hectare
  - Phytoremediation: €15,000 per hectare
  - Thermal Desorption: €80,000 per hectare
  - Bioaugmentation: €40,000 per hectare
- **USA (in USD)**:
  - Plasma-Based: $70,000 per hectare
  - Phytoremediation: $20,000 per hectare
  - Thermal Desorption: $90,000 per hectare
  - Bioaugmentation: $45,000 per hectare

The comparison chart shows **Lost Farmland Due to PFAS Contamination: Europe vs USA:**

- **Europe**: Approximately 300,000 hectares of farmland were lost to PFAS contamination.
- **USA**: Approximately 200,000 hectares of farmland were lost to PFAS contamination.

Projected Farmland Loss Due to PFAS Contamination Over the Next 25 Years for Europe and the USA:

- **Europe**:
  - Starting at 300,000 hectares, increasing to 550,000 hectares over 25 years.
- **USA**:
  - Starting at 200,000 hectares, increasing to 400,000 hectares over 25 years.

The projections emphasize the urgency of addressing PFAS contamination to prevent significant agricultural losses. The pie Figure 6—charts illustrate the impacts of PFAS contamination:

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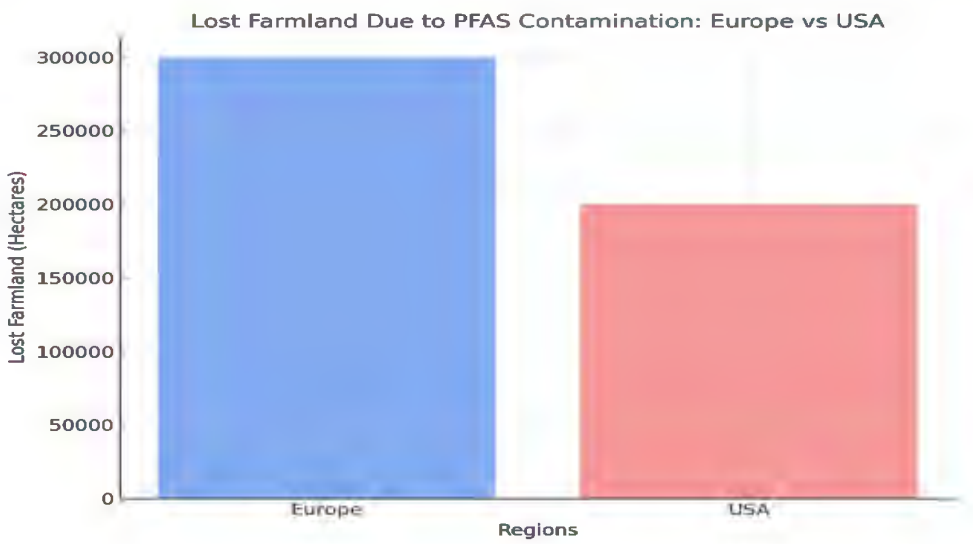

Figure 6. Lost Farmland Due to PFAS Contamination: Europe vs USA.

1. **Impact on Food Supply (Europe)**:
   - 80% of the food supply remains available.
   - 20% is lost due to farmland contamination over 25 years.
2. **Healthcare Cost Increase Due to PFAS**:
   - 85% represents baseline healthcare costs.
   - 15% is attributed to the increase in costs due to PFAS-related health conditions.

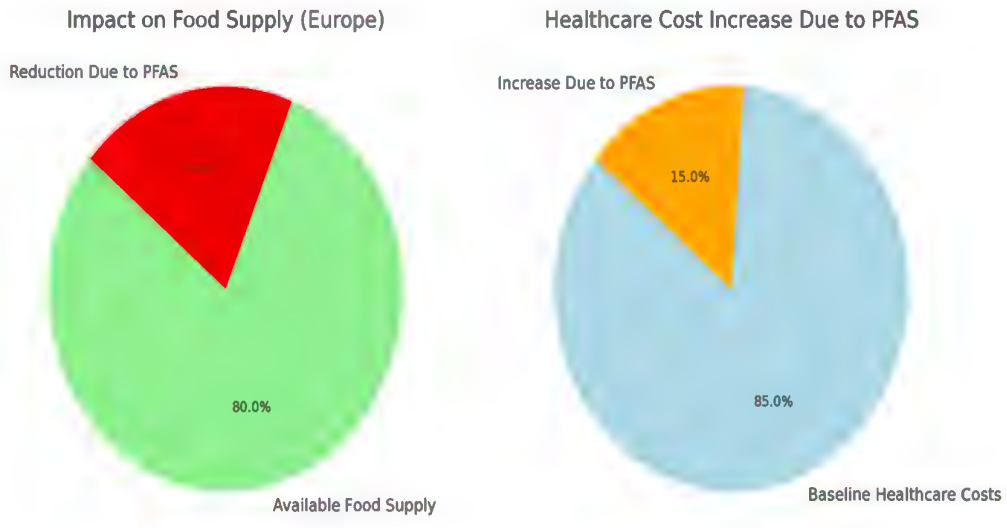

Figure 7. Impact of PFAS Contamination.

Remediation Strategies for European Farmlands

Detection Tools for PFAS Contamination

The ability to accurately detect PFAS contamination in soil, water, and agricultural environments is critical for mitigation efforts. Advanced detection tools such as AI-integrated mapping and biomarker technologies are revolutionizing the field by enabling real-time, cost-effective identification of PFAS hotspots. Below is a detailed expansion of these technologies, including their mechanisms, mathematical models, and potential applications.

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AI Mapping

AI-integrated mapping tools utilize artificial intelligence to analyze spatial data and identify PFAS contamination hotspots. These systems combine geospatial technologies, machine learning, and sensor data to generate precise contamination maps.

How It Works:

1. **Data Collection**: Sensors collect data on soil and water PFAS concentrations, geophysical characteristics, and hydrology.
2. **Data Integration**: AI systems integrate datasets from various sources (e.g., remote sensing, ground-based sensors).
3. **Hotspot Prediction**: Machine learning algorithms predict contamination patterns and potential hotspots by analyzing spatial correlations and trends.

Mathematical Foundation: AI mapping often uses a combination of supervised and unsupervised learning techniques. For example:

- **Regression Models**: Predict PFAS concentrations based on input variables such as soil permeability (PP) and distance to contamination source (d):

$$C_{\text{predicted}} = \beta_0 + \beta_1 P + \beta_2 d + \epsilon \quad (4)$$

where:

- $C_{\text{predicted}}$: Predicted PFAS concentration,
- $\beta_0, \beta_1, \beta_2$: Regression coefficients,
- ϵ: Error term.

- **Spatial Clustering (K-Means)**: Identify clusters of high PFAS concentrations:

$$\min \sum_{i=1}^{k} \sum_{j \in C_i} \|x_j - \mu_i\|^2 \quad (5)$$

where:

- k: Number of clusters,
- C_i: Data points in cluster i,
- x_j: Position of data point j,
- μ_i: Centroid of cluster ii.

Applications:

- **Field-Level Analysis**: AI tools map PFAS concentrations on farms, enabling targeted remediation.
- **Policy Development**: Governments use these maps to prioritize regions for intervention.

Innovative Example: A 2022 study implemented deep learning for PFAS detection by training convolutional neural networks (CNNs) on hyperspectral imaging data. This approach achieved 90% accuracy in identifying contaminated zones in test scenarios (Liu et al., 2022).

Biomarker Technology

Biomarker technology employs genetically engineered microorganisms to detect PFAS contamination. These microbes fluoresce or change color when exposed to PFAS, offering a cost-effective, field-deployable solution.

Mechanism:

1. **Engineering Microorganisms**:
   - Genes responsible for producing fluorescent proteins (e.g., green fluorescent protein, GFP) are inserted into microbes.
   - These genes are activated in the presence of PFAS, leading to a detectable fluorescence.
2. **Detection Process**:
   - Microbes are introduced into soil or water samples.

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- Fluorescence intensity correlates with PFAS concentration.

Mathematical Modeling: The fluorescence response of biomarkers can be modeled using the Michaelis-Menten equation:

$$v = \frac{V_{\max}[S]}{K_m + [S]} \tag{6}$$

where:

- v: Fluorescence intensity,
- $V_{\max}$: Maximum fluorescence response,
- K_m: PFAS concentration at half-maximal fluorescence,
- $[S]$: PFAS concentration in the sample.

Algorithmic Framework: To process fluorescence data:

1. **Signal Processing**: Use Fourier transforms to eliminate noise from fluorescence measurements.
2. **Concentration Estimation**: Apply regression models to correlate fluorescence intensity with PFAS concentration.

Containment and Post-Remediation Strategies for PFAS Mitigation

Effective containment and post-remediation strategies are critical to ensuring that PFAS contamination is fully addressed and ecosystems are restored to their natural state. These strategies focus on capturing byproducts, restoring soil health, and implementing monitoring systems to prevent recontamination and evaluate remediation success over time.

Containment of PFAS Byproducts

Closed-Loop Systems

Closed-loop systems are designed to capture and safely store PFAS breakdown products generated during remediation processes, such as thermal desorption or plasma-based treatments. These systems minimize the risk of secondary contamination and ensure that treated materials are safe for reuse.

Key Features:

Integrated Capture Mechanisms: Combines gas-phase filters, cryogenic traps, and activated carbon systems to capture volatilized PFAS during thermal or plasma treatments.

Recycling Capabilities: Treated water and soil are reintroduced into the environment only after thorough purification, promoting sustainability.

Case: In a pilot project in Sweden, a closed-loop system successfully captured over 95% of PFAS byproducts generated during soil heating, with the residual water meeting EU safety thresholds for reuse in agriculture (Goldenman et al., 2019).

Nanofiltration Membranes

Nanofiltration membranes are increasingly used to trap PFAS molecules in water, particularly during the treatment of contaminated groundwater or leachate. These membranes operate at a molecular level, allowing water molecules to pass through while retaining larger PFAS molecules.

Mechanism:

- Nanofiltration membranes use pore sizes between 0.001 and 0.01 microns to block PFAS, particularly long-chain molecules.
- Reverse osmosis is often employed in conjunction with nanofiltration for enhanced PFAS removal.

Advantages:

- High efficiency, with removal rates exceeding 99% for long-chain PFAS.
- Versatility in treating both water and leachate.

Challenges:

- Disposal of concentrated PFAS-laden brine remains an issue but can be remediated.

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- Membrane fouling can reduce efficiency over time (Rahman et al., 2014).

Soil Restoration

Remineralization

After PFAS removal, soil often requires replenishment of minerals and organic matter to restore its fertility and structure. Remineralization focuses on balancing nutrient levels and enhancing soil health.

Approach:

- **Mineral Addition**: Supplement the soil with essential minerals like calcium, magnesium, and phosphorus to correct deficiencies caused by PFAS removal processes.
- **Organic Amendments**: Incorporate compost, biochar, or humic substances to improve soil organic matter and microbial activity.

Benefits:

- Enhances water retention and aeration.
- Promotes healthy root growth and plant productivity.

Case Study: A remediation project in Germany restored a PFAS-contaminated field by applying a biochar-mineral blend. The treated soil showed a 40% improvement in crop yield within two growing seasons (Ross et al., 2018).

Microbial Recolonization

The reintroduction of native or engineered microbiota is essential for rebuilding soil ecology after PFAS removal. Microbial recolonization restores the natural biochemical processes necessary for healthy soil ecosystems.

Methodology:

- **Selection of Microbes**: Use a combination of native bacteria and fungi adapted to the local environment.
- **Delivery Systems**: Spray or inject microbial solutions into the soil to ensure even distribution.
- **Monitoring**: Assess microbial activity and diversity using soil DNA sequencing and metabolic profiling.

Advantages:

- Promotes nutrient cycling and organic matter decomposition.
- Reduces soil compaction and improves plant-microbe interactions.

Case: Engineered *Pseudomonas putida* strains were introduced into PFAS-remediated soils in Denmark, enhancing nitrogen fixation and improving soil fertility by 30% (Liu et al., 2019).

Monitoring Systems

IoT-Connected Sensors: IoT-connected sensors enable real-time monitoring of PFAS levels and soil health parameters, ensuring the long-term success of remediation efforts.

Key Features: **Real-Time Data**: Sensors measure PFAS concentrations, pH, moisture levels, and nutrient content. **Remote Access**: Data is transmitted to cloud-based platforms, allowing stakeholders to monitor conditions from anywhere. **Automation**: Automated alerts and reports help identify emerging issues quickly.

Applications: Track the effectiveness of remediation processes. Ensure compliance with regulatory thresholds for PFAS in soil and water.

AI-Driven Predictive Models: Artificial intelligence enhances long-term monitoring by predicting the behavior of residual PFAS and evaluating the risk of recontamination.

Modeling Approach: Use machine learning algorithms to analyze data from sensors, historical contamination patterns, and soil characteristics.Predict future contamination hotspots and recommend preventive measures.

Mathematical Model Case: A predictive algorithm based on logistic regression:

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$$P(C_{\text{hotspot}}) = \frac{1}{1 + e^{-(\beta_0 + \beta_1 X_1 + \beta_2 X_2 + \ldots + \beta_n X_n)}} \quad (8)$$

where:

- $P(C_{\text{hotspot}})$: Probability of a contamination hotspot.
- $X_1, X_2, \ldots, X_n$: Predictor variables (e.g., soil PFAS levels, pH, rainfall).
- $\beta_0, \beta_1, \ldots, \beta_n$: Regression coefficients.

Case Study: In the Netherlands, AI-driven models successfully predicted PFAS movement in agricultural zones, enabling targeted interventions that reduced contamination by 20% over two years (Goldenman et al., 2019).

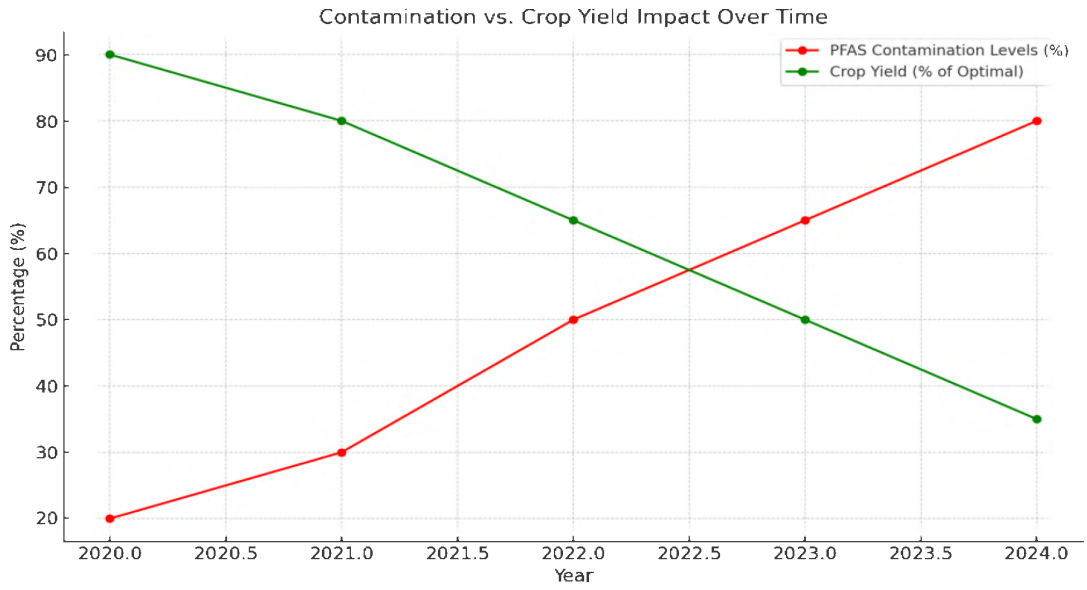

Figure 8. The correlation between increasing Cost-Benefit Analysis of PFAS Remediation.

3.2. Findings the Challenges and Opportunities in PFAS Remediation

High Costs of Advanced Remediation Technologies

Advanced PFAS remediation technologies, such as plasma-based treatments, thermal desorption, and bioaugmentation, require significant upfront investment and operational expenditures. Depending on the chosen method and contamination levels, estimates for remediating PFAS-contaminated soil range from €50,000 to €100,000 per hectare (Goldenman et al., 2019). Advanced PFAS remediation technologies, like plasma-based treatments and thermal desorption, can cost between €50,000 and €100,000 per hectare, especially for small-scale farms or regions with widespread contamination. However, the true cost of inaction far outweighs the expense of remediation. PFAS contamination permanently renders farmland unsuitable for food production, leading to economic and social impacts. Abandoned farmland in Northern Europe has led to food shortages, increased reliance on imports, and economic losses exceeding €500 million over a decade. Importing food to compensate for lost arable land also introduces new costs, further burdening consumers. Despite the high remediation costs, they represent an investment in preserving irreplaceable resources critical to regional food supply chains.

Knowledge Gaps in PFAS Degradation Pathways and Long-Term Impacts

Recent advances in PFAS research have made significant progress in understanding degradation pathways and the potential long-term effects of residual contamination. However, gaps remain in

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understanding the toxicity and environmental persistence of secondary compounds, as well as the cumulative impact of PFAS on soil microbiomes and nutrient cycling. Addressing these knowledge gaps is crucial for developing safe and effective remediation technologies, (Ross et al., 2018).

Global

The global response to PFAS contamination has been uneven, the Stockholm Convention, a United Nations initiative, aims to phase out certain PFAS compounds, but implementation varies across countries. The lack of standardized thresholds complicates international cooperation, and developing nations lack infrastructure and financial resources, underscoring the need for global knowledge-sharing and funding mechanisms.

The United States

The United States has adopted a fragmented approach to addressing PFAS contamination, with the EPA focusing on enforceable standards and monitoring. State governments like Michigan and California have implemented stricter regulations, but face challenges like high remediation costs.

Europe

The EU has established robust frameworks for PFAS management, including the Green Deal and REACH regulation. These regulations focus on reducing PFAS and supporting innovative remediation technologies. However, disparities in enforcement and funding across member states hinder comprehensive PFAS control. Success stories from Denmark and Sweden demonstrate the effectiveness of plasma-based remediation and bioaugmentation.

Greece and Mediterranean Countries

In Greece, PFAS contamination awareness and remediation efforts are still in their early stages. The lack of enforceable regulations and monitoring infrastructure has left significant gaps in addressing contamination risks. Greece's reliance on small-scale farms further complicates the issue, as these farms often lack the financial and technical resources to adopt advanced remediation technologies. The Mediterranean region faces unique challenges, including arid climates that limit natural PFAS attenuation and sandy soils that increase the risk of groundwater contamination. These factors necessitate tailored solutions, such as low-cost phytoremediation and bioaugmentation adapted to local conditions.

Opportunities in PFAS Remediation

Leveraging EU Green Deal Initiatives

The EU Green Deal provides a framework for scaling up PFAS remediation while promoting sustainable agricultural practices. Horizon Europe funding opportunities support research and pilot projects, reducing farmers' financial burdens and enabling the adoption of advanced technologies. For example, integrating solar-powered thermal desorption systems aligns with both remediation and the EU's renewable energy goals.

Collaboration Between Stakeholders

Public-private partnerships and cross-sector collaboration are critical for accelerating PFAS remediation. Partnerships between academia, industry, and policymakers can:

- Facilitate knowledge sharing and reduce technological barriers.
- Enable large-scale deployment of innovative solutions, such as plasma-based systems or hybrid remediation methods.

In Denmark, such collaborations have reduced PFAS levels in contaminated farmland by 85% while generating valuable data on scalability and cost-effectiveness (Liu et al., 2019).

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The Cost of Inaction vs. the Value of Remediation

The cost of inaction must be contextualized against the irreversible consequences of leaving farmland contaminated. Current estimates suggest that 3% of agricultural land in the EU is already impacted by PFAS, resulting in an annual loss of 2.5 million tonnes of food output (Goldenman et al., 2019). Beyond economic losses, the ethical and environmental implications of failing to address PFAS contamination include threats to food security, public health, and ecological integrity. Investing in remediation ensures that agricultural land remains productive, reduces dependence on food imports, and protects rural communities from collapse. While the challenges are significant, proactive action offers a pathway to sustainable food systems and environmental recovery.

Key actions include training farmers and local residents to monitor PFAS levels using low-cost detection tools. Awareness campaigns educate communities about PFAS contamination risks and preventive measures. Public dashboards provide real-time data on PFAS levels and health impacts. Implementing these recommendations requires sustained commitment, collaboration, and partnerships to achieve 100% remediation of contaminated farmland and sustainable food production.

4. Discussion

Environmental research is increasingly utilizing AI to address PFAS pollution, trained with real-world data, can suggest solutions and support actionable decisions, (Stensson et al.). For instance, AI can estimate PFAS accumulation in human bloodstreams, detect leaching from food packaging, and predict PFAS transportation via air currents, (Di et al.2022). This collaboration between AI and environmental monitoring and regulatory practice has expanded beyond technical dialogues to developing prototype scenarios demonstrating how AI can be integrated into characterization efforts, (Draghi et al.2024). AI has the potential to revolutionize monitoring of PFAS transport and distributions, offering improved cleanup strategies, (Li & MacDonald Gibson, 2022). By utilizing neural networks and machine learning techniques (Hu et al.2023), AI can analyze new and historical data streams, enabling real-time monitoring systems, (Breitmeyer et al.2024). This integration of AI can predict future trends and provide just-in-time mitigation options (Tokranov et al.2024), making it an exciting prospect for monitoring PFAS in-situ, (Jeong et al.2024). Text data modeling is used to enhance solid waste management, with results consistent with environmental monitoring, (Tatarinov et al.2022). The use of AI in environmental science is growing, offering innovative solutions to address environmental problems and promote resilience in society, (Stahl, 2021). Research on emerging pollutants in the field of environmental science is focusing on ethical, legal, and environmental hygiene impacts from wastewaters and reuse water in communities , (Adamopoulo et al., 2023; Gill & Germann, 2022). Artificial intelligence is expected to play a crucial role in detecting and managing these pollutants (Yigitcanlar et al., 2021), demonstrating its potential to significantly improve the detection and management of pollutants in the future, (Dauvergne, 2022). AI plays a transformative role in environmental research, particularly in PFAS contamination. It has significantly improved modeling of PFAS presence in food and correlations around predicted values in environmental waters and sediments, (Tao et al.2024) . Ensemble regression modeling has shown a 50% decrease in human serum half-lives for six PFSAs (Iulini et al.2024) , while artificial neural networks have shown correlations in serum half-life predictions, (Li & MacDonald Gibson, 2022). AI increases the efficiency of research, providing quicker and more accurate reported results, offering new opportunities for environmental scientists, (Lei et al.2023). AI-driven solutions, utilizing machine learning and molecular simulations (Kibbey et al., 2021), are being utilized to predict and prioritize PFAS in various systems (Karbassiyazdi et al.2022), thereby enhancing their environmental relevance and facilitating life cycle assessments and hazard risk assessments, (Zhang & Zhang, 2022; Han et al.2023).The overarching challenge in AI and PFAS studies is the reliance on untrusted sources and statistical tests, (Feinstein et al.2021). This lack of quantitative measurements and comprehensive validation necessitates collaboration among diverse stakeholders to improve data quality, (Su et al.2024). Interdisciplinary approaches, considering both short and long-term goals, are needed to

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create less toxic PFAS, shifting the normal logic of isolating variables, (Li & MacDonald Gibson, 2022). Finally, the implications of climate change and extreme weather events (Adamopoulos et al. 2023c; Adamopoulos et al., 2024b), on water supplies and traditional water management in Europe must be emphasized (Adamopoulou et al., 2023), since they have a negative impact on public health, agriculture, and food safety, (Adamopoulos et al., 2022). The discussion above shows that incorporating AI into mitigating PFAS contamination and ecosystem risk is not without challenges, but it also provides enormous opportunities to reveal hidden insights and patterns, generate increasingly reliable predictions, and accelerate the development of sustainable management plans.

Recommendations

Addressing PFAS contamination in Europe's agricultural sector requires strategic investments, robust regulatory frameworks, and active collaboration among stakeholders. The recommendations outlined here focus on creating sustainable, scalable, and inclusive approaches to remediate contaminated farmland and prevent future contamination. The EU is focusing on PFAS remediation and PFAS monitoring to combat contamination in agriculture. Key actions include increasing EU funding for research on cost-effective, scalable remediation technologies, strengthening regulatory frameworks to enforce PFAS monitoring, fostering public-private partnerships for comprehensive remediation, and promoting community involvement in PFAS monitoring and awareness. The EU Green Deal's Chemical Strategy for Sustainability aims to accelerate the development of innovative PFAS remediation technologies, while REACH regulations must be enhanced to include stricter thresholds for PFAS levels in agricultural soils and water sources. Public-private partnerships can mobilize resources and expertise for large-scale PFAS remediation, while promoting community involvement builds trust and promotes sustainable agricultural practices. These actions aim to reduce costs, improve efficiency, and promote sustainable agricultural practices.

Future Directions

The developed world, natural sciences, and social sciences are all interrelated subjects that are critical to comprehending the built environment. By combining data scientists' knowledge with insights from these domains, we can design effective policy initiatives to solve the pressing PFAS issues. It is critical to avoid dismissive rules and laws, instead focusing on cautious principles and dealing with the rebirth of history. The combination of human knowledge and AI capabilities has the potential to solve these complicated problems.

5. Conclusions

The requirement to address PFAS pollution in the environment and in communities is urgent. AI technology hold tremendous promise for environmental research. Implementing cross-disciplinary monitoring frameworks, top-down legislation, and analytical and remedial tools are all necessary for an effective response. Collaboration, democratic engagement, and self-management of affected communities are critical in decision-making processes. The rapid speed of AI advancement highlights the revolutionary potential of these technologies in combating PFAS contamination. Public Health in Europe faces a significant challenge in addressing PFAS contamination, which poses a significant environmental threat to food security and agricultural sustainability. These "forever chemicals" accumulate in soil, water, crops, and livestock, threatening food systems and community health. To combat PFAS contamination, a multi-pronged approach is needed, integrating innovative technologies like plasma-based remediation, bioaugmentation, and nanofiltration with comprehensive regulatory frameworks. Investing in research to develop cost-effective and scalable solutions is crucial, while regulatory reform and public-private partnerships can accelerate the deployment of advanced remediation technologies. Community engagement is also vital, as farmers, local residents, and other stakeholders must be equipped with the knowledge and tools to monitor contamination, participate in remediation efforts, and advocate for sustainable practices. Inaction will

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lead to escalating costs in terms of food imports, public health expenditures, and environmental degradation. By combining innovation, regulation, and community empowerment, Europe can lead the way in addressing PFAS contamination and ensuring the well-being of future generations.

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Review article

Accumulation of perfluorinated alkyl substances (PFAS) in agricultural plants: A review

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ABSTRACT

PFASs are a class of compounds that include perfluoroalkyl and polyfluoroalkyl substances, some of the most persistent pollutants still allowed - or only partially restricted - in several product fabrications and industrial applications worldwide. PFASs have been shown to interact with blood proteins and are suspected of causing a number of pathological responses, including cancer. Given this threat to living organisms, we carried out a broad review of possible sources of PFASs and their potential accumulation in agricultural plants, from where they can transfer to humans through the food chain. Analysis of the literature indicates a direct correlation between PFAS concentrations in soil and bioaccumulation in plants. Furthermore, plant uptake largely changes with chain length, functional group, plant species and organ. Low accumulations of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) have been found in peeled potatoes and cereal seeds, while short-chain compounds can accumulate at high levels in leafy vegetables and fruits. Significant variations in PFAS buildup in plants according to soil amendment are also found, suggesting a particular interaction with soil organic matter. Here, we identify a series of challenges that PFASs pose to the development of a safe agriculture for future generations.

1. Introduction

Contamination of regional, rural and urban ecosystems by anthropic activities has been a constant concern in environmental sciences in recent decades, and environmental regulations have to be continuously updated as new pollutants emerge (Manzetti et al., 2014). Poly- and perfluorinated alkyl substances (PFASs), especially perfluoroalkyl acids (PFAAs), which include perfluoroalkyl carboxylic acid (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs), are among the world's major pollutants. PFASs have been detected in oceans, across continents, and in remote parts of the globe, including the North Pole (Bossi et al., 2005; Cai et al., 2011; Holmström et al., 2005; Paul et al., 2008; Zhao et al., 2012), with several ecosystems in the USA, China and Europe affected (Ahrens et al., 2010; Herzke et al., 2013; Jin et al., 2009; Kowalczyk et al., 2013; Loos et al., 2009; So et al., 2004, 2007; Washington et al., 2010).

PFASs are a family of molecules composed of a carbon chain that can be linear or branched, and fully or partially fluorinated (Buck et al., 2011). They have unique physicochemical characteristics due to the larger size of the fluorine atom with respect to the hydrogen atom, and the strength of the carbon-fluorine bond, which give fluoroalkyl moieties their high thermal, chemical and biochemical stability. (Krafft and Riess, 2015; O'Hagan, 2008). This group of molecules also includes compounds with low aqueous surface tension and the property of interacting with water and non-polar phases, and they therefore exhibit amphiphilic behavior. These features have been widely exploited in several industrial and commercial applications for the preparation of fluorinated polymers and surfactants since 1950s. Examples include oil-repellent coatings for food containers and cookware, firefighting foams, coloring and waterproofing agents for fabrics, detergents, floor waxes, and pesticide formulations (Buck et al., 2011; Kissa, 1994). As a result of their wide use and high persistence, a broad range of these

Abbreviations: PFASs, Poly- and Perfluoroalkyl substances; PFAAs, Perfluoroalkyl acids; PFCAs, Perfluoroalkyl carboxylic acids; PFSAs, Perfluoroalkyl sulfonic acids; PFBA, Perfluorobutanoic acid; PFPeA, Perfluoropentanoic acid; PFHxA, Perfluorohexanoic acid; PFHpA, Perfluoroheptanoic acid; PFOA, Perfluorooctanoic acid; PFNA, Perfluorononanoic acid; PFDA, Perfluorodecanoic acid; PFUnA, Perfluoroundecanoic acid; PFDoA, Perfluorododecanoic acid; PFBS, Perfluorobutane sulfonic acid; PFHxS, Perfluorohexane sulfonic acid; PFOS, Perfluorooctane sulfonic acid

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substances have been detected in the environment, and in wildlife and humans. PFASs accumulate in the body, mainly in the liver, with mean half-life values of 3.8 years for PFOA and 5.4 years for PFOS, though with considerable variability (EFSA, 2008). They saturate protein surfaces (Bischel et al., 2010; Chen and Guo, 2009; Manzetti et al., 2014; Qin et al., 2010), and are transferred through the maternal cord from the placenta in rat models as well as in humans (Inoue et al., 2004; Loccisano et al., 2012; Winkens et al., 2017). Studies on rats showed that PFOA and PFOS accumulate mainly in liver, kidneys and serum (EFSA, 2008).

The toxicity of PFASs, particularly PFOA and PFOS, has been extensively studied with clear results on animals, but less statistical evidence regarding humans (for details, see individual chapters in DeWitt, 2015). Less is known regarding other compounds, including those with a short chain, such as perfluorobutanoic acid (PFBA), perfluorohexanoic acid (PFHxA) and perfluorobutane sulfonic acid (PFBS), which have been recently introduced by the manufacturing industry following concerns about the undesired effects of PFOA and PFOS on human health and the environment (Ritter, 2010). Although some short-chain PFASs, i.e. $C < 7$ for PFCAs and $C < 6$ for PFSAs (Buck et al., 2011), are not considered bioaccumulative, they are as recalcitrant to degradation as long-chained PFASs, are highly soluble, and have lower sorption to solids, which lead to greater mobility in the environment (Vierke et al., 2012; Wang et al., 2015)

Adverse effects associated with PFAS exposure in animal models include hepatotoxicity, tumor induction, developmental toxicity, immunotoxicity, neurotoxicity and endocrine disruption (Table 1; DeWitt, 2015 for a recent review). Regarding human health, the most comprehensive epidemiological data linking PFOA exposure and health outcomes were gathered by the C8 Science Panel (http://www.c8sciencepanel.org), which studied Mid-Ohio Valley communities that had been potentially affected by releases of PFOA since the 1950s. Probable links between serum levels and several health endpoints, such as increased levels of cholesterol, liver enzyme activity and uric acid, altered thyroid parameters, and pregnancy-induced hypertension, have emerged from these studies. The same investigations also report a probable link between PFOA exposure and testicular and kidney cancers (http://www.c8sciencepanel.org). The International Agency for Research on Cancer (IARC, 2017) has recently classified PFOA as possibly carcinogenic, while the US Environmental Protection Agency (EPA) concluded that both PFOA and PFOS are possibly carcinogenic to humans.

Among the most common PFASs found in the environment (Table 1), PFOA and PFOS are considered some of the most widespread organic pollutants for biota and humans (So et al., 2004; Ahrens, 2011; Houde et al., 2011; Kannan et al., 2001, 2005; Yamashita et al., 2005). However, other PFASs have been detected in the environment, such as PFBA, perfluoroundecanoic acid (PFUnA), and perfluorododecanoic acid (PFDoA) (Ahrens et al., 2009). Their persistence is strongly associated with carbon chain length, and PFASs with more than five ($C > 5$) carbon atoms are biopersistent and may have higher K_{ow} values than those with shorter chain lengths (Eriksen et al., 2010; Latała et al., 2009; Parsons et al., 2008). Reactivity, on the other hand, is less dependent on chain length, but is instead related to the non-fluorinated part of the molecules and/or the functional groups of the PFAS end-region (i.e. the sulfonic or carboxylic groups). The fluorinated part can generally be regarded as inactive, given the high strength of the carbon-fluoride bond, 109–130 kcal/mol (O'Hagan, 2008; Fuchibe and Akiyama, 2006). It is important to underline that the substances containing a perfluoroalkyl moiety have the potential to be transformed abiotically or biotically into perfluoroalkyl substances through changes in the non-fluorinated part of the molecule (Buck et al., 2011). Therefore, the corresponding carboxylic acids and sulfonic acids are formed from several precursors, such as fluorotelomer alcohols, sulfonamide derivatives, and polyfluoroalkyl phosphates. For this reason, we focus our review on the uptake and distribution in plants of PFAAs (PFCAs and PFSA) rather than their precursors.

2. Sources of PFASs in humans

Water and food consumption are generally considered the two major sources of PFASs in humans, although air and air-suspended dust, food packaging and cookware also contribute to the overall PFAS load in the human body (Tittlemier et al., 2007). Seafood, especially when fished in hotspot waters with much higher PFAS concentrations than the background, is considered one of the most important food sources of PFASs in humans (Brambilla et al., 2015; Christensen et al., 2017; D'Hollander et al., 2010a, 2010b; Fromme et al., 2009; Hloušková et al., 2013).

Herzke et al. (2013) and D'Hollander et al. (2015) recently reported the presence of PFASs, particularly PFCAs, in various vegetables, cereals and fruits sampled in Belgium, the Czech Republic, Italy and Norway. Furthermore, a series of scientific articles on PFAS accumulation in plants is emerging across the world (vide infra). Plants may, therefore, be considered possible contributors to the PFAS body burden in humans, either directly as part of the human diet (Klenow et al., 2013) or indirectly as fodder for livestock (Kowalczyk et al., 2013).

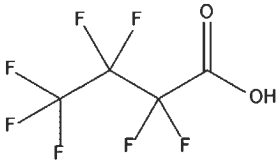
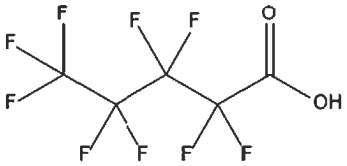
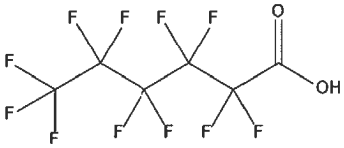
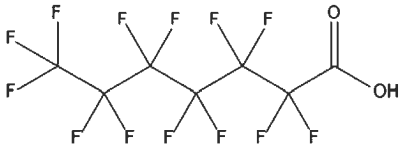
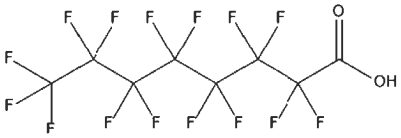
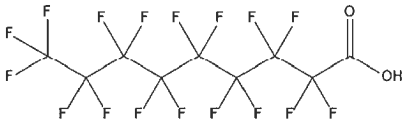
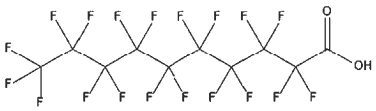
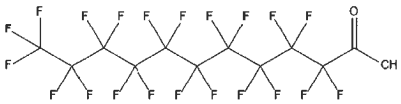
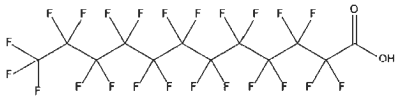
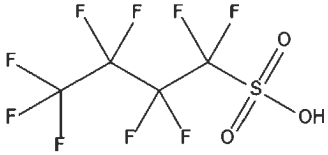
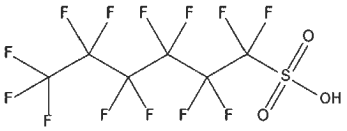
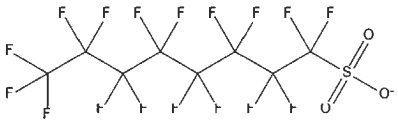
The purpose of this paper is to review the recent literature regarding the uptake and partitioning of the most common PFASs in agricultural plants, taking into account both cereals and vegetables, in an attempt to draw a picture of the role of the biotic and abiotic factors affecting these processes.

3. Sources of PFASs in plants

Due to the partial water solubility of PFASs, water is assumed to be their main vehicle of transfer across environmental compartments and biota, making a substantial contribution to their diffusion. High concentrations of PFAAs, which encompass both PFCAs and PFSAs, are often reported in effluents from wastewater treatment plants (WWTPs) (Llorca et al., 2012; Xiao et al., 2012), indicating that both urban and, particularly, industrial purification plants contribute to PFAS delivery to the environment. It has also been shown that PFOS and other long-chained PFSAs and PFCAs are mobilized from landfills and released into water bodies (Busch et al., 2010; Huset et al., 2008; Weber et al., 2011). In addition, runoff and leaching from areas treated with firefighting foams make a considerable contribution to PFAS pollution (Ahrens, 2011; Backe et al., 2013; Brambilla et al., 2015; Filipovic et al., 2015; Korucu et al., 2015). PFOS levels ranging from $< 10\ ng\ L^{-1}$ to $100\ ng\ L^{-1}$ have been reported in surface and ground waters (Brambilla et al., 2015), but PFOA concentrations at the $\mu g\ L^{-1}$ level have been measured in surface waters in Italy and in the Ruhr region of Germany (Valsecchi et al., 2015; Skutlarek et al., 2006). Alarming values at the $mg\ L^{-1}$ level in ground water and surface water close to fire-training locations and airports have also been measured (Ahrens, 2011; Moody and Field, 1999). Field irrigation with contaminated surface or ground waters may be an important source of soil and plant contamination by PFASs.

On agricultural land, a further important source of perfluorinated compounds comes from the use of contaminated sewage sludge as a soil conditioner (Sepulvado et al., 2011; Zareitalabad et al., 2013). In fact, PFASs entering WWTPs, although with differences due to their chain length and functional group, can partly escape into the effluent and partly sorb to sludge due to their amphiphilic behavior (Chen et al., 2013). In some areas of Germany, PFOS and PFOA have been measured in concentrations of 1000 and 910 μg/kg soil dry weight (d.w.), respectively, in the top 0.3 m layer of the soil, and in concentrations of 5500 and 810 μg/kg d.w., respectively, at 0.3–0.6 m, as a result of the application of soil amendments derived from industrial waste (Wilhelm et al., 2008, 2009). In the USA, the use of contaminated sludge as a soil conditioner has caused contamination ranging from 500 to 2000 μg/kg to 4000–6000 μg/kg d.w. of total PFASs, according to the year and field surveyed. Perfluorodecanoic acid (PFDA) was the major PFAS

Table 1
A list of the most common perfluorinated alkyl substances (PFASs) with their applications and properties.

Compound	Use	Structure	MW	Solubility in water	Bioaccumulative properties	Confirmed toxic effects
Perfluorobutanoic acid, PFBA	Synthetic chemistry		214.04	Soluble	No (Conder et al., 2008; Wang et al., 2015)	–
Perfluoropentanoic acid, PFPeA	Breakdown product of stain- and grease-proof coatings on food packaging, couches, carpets		264.05	–	No (Hölzer et al., 2008) Binds to serum (Qin et al., 2010).	Increase in liver sAST protein (Upham et al., 2009)
Perfluorohexanoic acid, PFHxA	Unknown		314.05	Miscible	Yes, Log K_{ow}: 4.37 (Zhao et al., 2013; Fan et al., 2014)	Yes Kidney accumulation (Klaunig et al., 2014)
Perfluoroheptanoic acid, PFHpA	Breakdown product of stain- and grease-proof coatings on food packaging, couches, carpets.		364.06	9.5 g/l	Yes (Haukås et al., 2007) Log K_{ow}: 5.33 (Zhao et al., 2013)	Yes (Rosen et al., 2013; Vongphachan et al., 2011; Wolf et al., 2012)
Perfluorooctanoic acid, PFOA	Water and oil repellant in fabrics and textiles, food packaging.		414.07	No	Log K_{OC}: 2.4 ± 0.12 (Ahrens et al., 2011b) Log K_D: − 0.22–0.55 (De Voogt and Sáez, 2006)	Yes (Abbott et al., 2007; Kudo and Kawashima, 2003; Wolf et al., 2007)
Perfluorononanoic acid, PFNA	Surfactant for synthesis of textiles and polymers		464.08	9.5 g/l	Yes (Henderson and Smith, 2007) Log K_{ow}: 6.3 (Zhao et al., 2013)	Yes (Henderson and Smith, 2007; Fang et al., 2008; Upham et al., 2009; Wolf et al., 2010)
Perfluorodecanoic acid, PFDA	Breakdown product of stain- and grease-proof coatings on food packaging, couches, carpets		514.08	No	Yes (Buck et al., 2011; Zhao et al., 2013; Jones et al., 2003; Martin et al., 2003a, 2003b) Log K_{ow}: 8.23 (Zhao et al., 2013)	Yes (Harris and Birnbaum, 1989; Heuvel et al., 1992)
Perfluoroundecanoic acid, PFUnA			564.09	9.5 g/l	Yes, Log K_{ow}: 9.20 (Zhao et al., 2013; Bjermo et al., 2013)	Yes (Takahashi et al., 2014)
Perfluorododecanoic acid, PFDoA			614.10	9.5 g/l	Yes, Log K_{ow}: 10.2 (Zhao et al., 2013)	Yes (Liu et al., 2016; Ren et al., 2015; Zhang et al., 2013)
Perfluorobutane sulfonic acid, PFBS	Stain repellant		300.10	1.4 µg/l (Martin et al. (2003a)	Yes (Jones et al., 2003; Martin et al., 2003b)	Yes (Hu et al., 2003)
Perfluorohexane sulfonic acid, PFHxS	Surfactant for textiles		400.11	–	Yes, Log K_{ow}: 4.34 (Wilhelm et al., 2009; Zhao et al., 2013; Calafat et al., 2006)	–
Perfluorooctanesulfonate, PFOS	Firefighting foam, textiles		500.13	680 mg/l (You et al., 2010)	Yes. Log K_{ow}: 6.28 (Houde et al., 2011; Zhao et al., 2013). Globally distributed in wild-life (Giesy and Kannan, 2001)	Yes (Ankley et al., 2005; Seacat et al., 2002, 2003)

(< 990 μg/kg d.w.), while PFOA was measured at 320 and PFOS at 410 μg/kg d.w. (Washington et al., 2010). The use of PFAS as an emulsifier in phytosanitary product formulations may also contribute to the presence of these substances in the above-ground edible parts of vegetables (Lassen et al., 2012). Furthermore, an alternative pathway of PFAS accumulation in plants is the aerial transport of volatile or particle-bound precursors, which can be absorbed by plant leaves with further metabolization to PFCAs and PFSAs (Rankin et al., 2016; Simonich and Hites, 1995; Yoo et al., 2011). Air emissions of PFASs from landfills and WWTPs (Ahrens et al., 2011a), and accumulation of neutral and ionizable PFASs in leaves around landfills (Tian et al., 2018) have been reported. The spread of PFASs via dust particles has been documented as reaching the polar regions (Schenker et al., 2008; Rankin et al., 2016). Lindim et al. (2016) estimated that the contribution of river discharge to PFOS input to the Baltic proper, the Black Sea and the Adriatic Sea was on average 3.5 times greater than that of atmospheric deposition, whereas atmospheric deposition of PFOA was similar to or larger than riverine disposal.

4. PFAS accumulation in plants

4.1. Cereals

As far as we know, the first systematic research on PFOA and PFOS transfer from soil to plants was conducted by Stahl et al. (2009). In their investigations on spring wheat, oats, potatoes, maize and perennial ryegrass grown in soil spiked with PFOA and PFOS in the range 0–50 mg/kg soil, they found that these pollutants accumulated in all the plant species, the tissue concentrations of PFOA being generally higher than those of PFOS. Accumulation was found to increase with soil pollutant concentrations, with values higher in green parts (vegetative) than in grains. Large differences were found among plant species, possibly due to their different protein contents, as indicated by Wen et al. (2016), or to the different compositions and surface areas of their root systems, or to biomass accumulation, as suggested by Müller et al. (2016). The amount of water transpired during growth has also been suggested to account for different uptakes and translocation capabilities among crops (Blaine et al., 2014a; 2014b). Therefore, all the climatic parameters influencing stomatal opening, such as irradiance, temperature and humidity, can influence plant uptake of perfluorinated compounds. In Stahl et al.'s (2009) experiments, PFAS concentrations in cereal straw generally followed the hierarchy maize < < oats < wheat, as shown in Table 2, which reports values obtained at a soil concentration of 1 mg/kg, a level comparable to those reported by Wilhelm et al. (2008) and Washington et al. (2010). Stahl et al. (2009) also showed that perennial ryegrass accumulated PFAS to different extents at four successive cuttings, with the highest concentrations at the second. Given the high level of PFAS accumulation (PFOA in particular), which ranged from 408 to 7520 μg/kg d.w. (Table 2), where the soil is contaminated this forage species can represent an entry point of PFASs into the food chain via animal feeding.

Krippner et al. (2015) deepened the study of PFAS accumulation in the straw and grains of maize plants by looking at ten C4 to C10 substances containing the carboxylic (PFCAs) or sulfonic (PFSAs) functional groups, all individually spiked at 0.25 and 1 mg/kg soil. They confirmed the direct correlation between soil and plant tissue concentrations: the higher the soil PFAS content, the more the plants accumulate. Differences in the accumulation levels of compounds in plants depend on chain length, with the longer-chained compounds generally having lower accumulation rates, and on the associated functional groups, with PFSAs generally having lower accumulation rates than PFCAs. Krippner et al. (2015) also confirmed straw as having a much greater rate of accumulation than kernels, the former having a total PFAS content of around 52,000 μg/kg d.w., the latter around 900 μg/kg d.w. at the individual spiking dose of 1 mg/kg soil for a total of 10 mg/kg soil (Table 2). At the same spiking dose, the highest

Table 2
Initial PFAS concentrations (mg/kg soil) and their accumulation in different crop plant parts (μg/kg d.w.).

Plant species and organ	Compounds	Type of treatment and initial soil concentrations (mg/kg soil)	Concentrations in plants (μg/kg d.w.)	References
Maize straw	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/kg: PFOA: 126 PFOS: 104	Stahl et al. (2009)
Maize straw	PFAA mixture, ten compounds	Spiked soil. 0.25 and 1 for each substance	At 1 mg/kg: PFBA: 35233 PFBS: 1843 PFOA: 645 PFOS: 617 ΣPFASs = 52200	Krippner et al. (2015)
Maize stover	PFAA mixture	Soil + urban 2x biosolids. 0.00716, as Σ. PFBA: 0.00010 PFBS: 0.00039 PFOA: 0.00128 PFOS: 0.00282	PFBA: 4.21 PFBS: < LOQ PFOA: < LOQ PFOS: < LOQ ΣPFASs = 4.49	Blaine et al. (2013)
Maize leaves	PFOS	Spiked soil. 38.5 PFBS as by-product: 0.020	PFOS: 23100 PFBS: 120	Navarro et al. (2017)
Maize ears	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/kg soil: PFOA: 4 PFOS: 3	Stahl et al. (2009)
Maize kernels	PFAA mixture, ten compounds	Spiked soil. 0.25 and 1 for each substance	At 1 mg/kg: PFBA: 229 PFBS: 5.44 PFOA: 2.45 PFOS: < LOQ ΣPFASs = 860	Krippner et al. (2015)
Maize roots	PFOS	Spiked soil. 38.5	254000	Navarro et al. (2017)
Oat straw	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/kg: PFOA: 690 PFOS: 150	Stahl et al. (2009)
Oat grains	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/k: PFOA: 54 PFOS: 17	Stahl et al. (2009)
Ryegrass, four cuttings	PFOA, PFOS	Spiked soil. 0–50 for each compound	Range at 1 mg/kg within the four cuttings: PFOA: 408–7520 PFOS: 92–470	Stahl et al. (2009)
Wheat straw	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/kg: PFOA: 1900 PFOS: 270	Stahl et al. (2009)
Wheat straw	PFAA mixture	Soil + biosolids at different rates: 0.0414–0.220, as ΣPFASs. At the highest rate: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	At 0.220 mg/kg as total amount: PFBA: 22.2 PFBS: 21.8 PFOA: 22.1 PFOS: 11.0 ΣPFASs = 178	Wen et al. (2014)
Wheat grains	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/kg: PFOA: 9 PFOS: 0	Stahl et al. (2009)
Wheat grains	PFAA mixture	Soil + biosolids at different rates: 0.0414–0.220, as ΣPFASs. At the highest rate: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	At 0.220 mg/kg as total amount: PFBA: 6.49 PFBS: < MDL PFOA: 2.90 PFOS: 2.53 ΣPFASs = 35.6	Wen et al. (2014)

(continued on next page)

Table 2 (*continued*)

Plant species and organ	Compounds	Type of treatment and initial soil concentrations (mg/kg soil)	Concentrations in plants (μg/kg d.w.)	References
Wheat husks	PFAA mixture	Soil + biosolids at different rates: 0.0414–0.220, as ΣPFASs. At the highest rate: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	At 0.220 mg/kg as total amount: PFBA: 5.77 PFBS: < MDL PFOA: 4.19 PFOS: 2.20 ΣPFASs = 37.8	Wen et al. (2014)

MDL = method-detection limit.
Σ = total amount.

concentrations of PFCAs and PFSAs in straw were those of the C4 perfluorinated chemicals PFBA and PFBS (35,233 ± 6535 and 1843 ± 687 μg/kg d.w., respectively), whereas the concentrations of PFOA and PFOS (both C8 compounds) were lower and similar to each other: 645 ± 121 and 617 ± 178 μg/kg d.w., respectively (Table 2). High amounts of perfluoropentanoic (PFPeA), i.e., 8335 ± 1499 μg/kg d.w., were also recorded. In kernels, the highest PFAS concentrations were those of the C4 - C6 PFCAs, especially PFPeA, with a mean value of 275 ± 91 μg/kg d.w., whereas PFOA content was only 2.45 ± 1.54 μg/kg d.w., and PFBS was the only PFSA detected (5.44 ± 1.90 μg/kg d.w.). Variability in the data, due to the complexity of the extraction procedures and analytical methods utilized, is also evident.

In a full-scale field study using urban biosolids, Blaine et al. (2013) found that PFBA and PFPeA were present only in maize stover, while the other PFASs were below the limit of quantification (LOQ) (Table 2). These results indicate that risk assessment studies based exclusively on PFOA and PFOS accumulation in plants do not give a real picture of the potential dangers to humans of food contamination, as many different PFASs may be present in soil and waters, and consequently in plants.

Comparison of the results reported by Stahl et al. (2009) and Krippner et al. (2015) reveals differences in the extent of PFOA and PFOS accumulation in maize plants, which are particularly evident in the straw (Table 2). Krippner et al. (2015) suggested that these differences could be due to different maize cultivars, growth conditions, and soil characteristics. They also hypothesized that interactions among PFAAs may influence their sorption to soil, and ultimately their uptake and distribution in maize plants.

Hydroponic studies on the uptake of perfluorinated chemicals by cereal plants are less realistic than those using soil as the growth medium, although they can provide more easily interpretable results regarding the mobility of PFASs through the plants without the interference of soil components. Krippner et al. (2014) investigated the accumulation of ten PFCAs and PFSAs with chain lengths from C4 to C10 in maize seedlings grown in nutrient solution and found a U-shaped pattern: five days into the experiment accumulations in the whole plants of the PFCAs with chain lengths up to the C7 compound perfluoroheptanoic acid (PFHpA) decreased, those with chain lengths C7 to C10 increased. Similar results obtained by Müller et al. (2016) with *Arabidopsis thaliana* were attributed to different mechanisms of interaction between plants and PFAS, the long-chained compounds being mainly adsorbed to the root surfaces, the short-chained ones more easily absorbed and translocated. Hydroponic experiments can also reveal information on the partitioning of different PFASs in the various plant compartments, particularly the shoot-to-root concentration ratio, i.e. the translocation factor (TF), which cannot be easily calculated in soil experiments. In maize plants, TF regularly decreased with chain length (< 1 for PFOA and PFCAs with chain lengths > C8), indicating that these chemicals are mainly retained by the root, as also reported by Wen et al. (2013) for PFOA and PFOS. PFSAs also had a similar, or even more pronounced, dependence of retention in roots with increasing chain length, PFBS being the only compound with a high TF. In summary, maize seedlings tend to accumulate C4 - C7 PFCAs and PFBS in the shoots, while the PFAS with higher chain lengths tend to be restricted to the roots. Interestingly, unlike experiments with spiked soil, with hydroponics PFOS accumulated to a greater extent than PFOA, as Wen et al. (2013) also showed, highlighting the important role played by soil components in determining PFAS uptake by plants.

Further results on PFAS accumulation in food crops were obtained by Stahl et al. (2013), who investigated the accumulation of PFOA and PFOS in straw and grain in a five-year lysimeter experiment with the crop sequence winter wheat, winter rye, canola, winter wheat, and winter barley, and with a spiking dose of 25 mg/kg soil. Results confirm that more of these perfluorinated compounds were allocated in the straw than in the grains of all the species examined, in agreement with other reports (Krippner et al., 2015; Stahl et al., 2009). With the exception of canola, PFOS concentrations in the straw were similar to or higher than those of PFOA, with the highest levels measured in the first year of the experiment in winter wheat (14,500 μg/kg d.w. of PFOS vs. 2030 μg/kg d.w. of PFOA). The unusual preponderance of PFOS over PFOA observed in this study was explained by high PFOA losses by leaching. PFBA and PFBS were also detected, these being by-products of the technical mixtures of PFOA and PFOS that were applied to the soil.

A field study on PFAS accumulation in wheat plants grown in biosolid-amended soils confirmed sewage sludge to be an important source of perfluorinated chemicals, and found their concentrations in wheat plants to increase with increasing amendment rate, although in a less-than-proportional manner (Wen et al., 2014). Soil organic matter was suggested as an important factor limiting the uptake of PFASs by wheat roots. Regarding the distribution within the plant of the nine C4–C14 PFCAs and three PFSAs studied, results confirmed the order of accumulation rate straw > grain reported by Krippner et al. (2015) and Stahl et al. (2009, 2013), and the more general order root > straw > grain ≥ husk. The presence of PFAS in husks in concentrations similar to those in grains raises concerns about the possible intake of PFAS with whole wheat flour, which is considered an important source of dietary fiber. PFAS chain length was also a determining factor in plant translocation: the longer the C-chain, the lower the transfer to wheat straw and grains, as shown in maize plants (Krippner et al., 2014, 2015).

Once again, comparison of results from different experiments reveals important differences. For example, Wen et al. (2014) found that at the lowest biosolid dose PFOA and PFOS in wheat grains were at levels of 1.01 ± 0.10 and 0.80 ± 0.02 μg/kg d.w., respectively, starting from soil concentrations of 4.33 ± 0.45 and 10.4 ± 1.0 μg/kg, respectively. Conversely, Stahl et al. (2009) found PFOS in grains only starting from a spiking dose of 10 mg/kg soil, with the contents of PFOA and PFOS in grains at 1300 and 2 μg/kg d.w., respectively.

A more comprehensive parameter with which to compare different experiments and plant species with respect to the plant's ability to accumulate pollutants could be the bioconcentration factor (BCF), i.e. the plant tissue-to-soil concentration ratio (Table 3), for which values > 1 indicate the tendency of plants to accumulate PFAS against a concentration gradient. The C4 compound PFBA had the highest BCF among all the PFASs in both maize and wheat straw. In maize plants grown at spiking doses of 0.25 and 1 mg/kg soil, the BCF for this compound was significantly higher at the lowest pollutant levels i.e. 63.64 *vs* 35.23 (Table 3, Fig. 1) (Krippner et al., 2015). This may indicate some kind of adaptation by plants to high PFBA concentrations or a sort of competition/interaction among PFAS molecules, as suggested by the same authors, although further studies are needed to support these hypotheses. A similar BCF of 64.8 was calculated in the stover of maize plants grown in soil amended with urban biosolids containing 0.10 μg/kg of PFBA (Blaine et al., 2013). In wheat straw, the BCFs for PFBA were more than an order of magnitude lower, ranging

Table 3
Crop plant part analyzed, initial PFAS concentrations (mg/kg soil), soil organic matter (SOM) or organic carbon (OC) contents and bioconcentration factors (concentration in plant parts/initial concentration in soil).

Plant species and parts	Compounds and initial concentrations (mg/kg)	PFBA, PFBS bioconcentration factors	PFOA, PFOS bioconcentration factors	SOM or OC contents	References
Maize straw	PFOA: 0–50 PFOS: 0–50	–	Values at 0.25 and 1 mg/Kg: PFOA: 0.272; 0.126 PFOS: 0.132; 0.104	na	Stahl et al. (2009)
Maize straw	PFAA mixture: 0.25 and 1 per each of 10 compounds	Values at 0.25 and 1 mg/Kg: PFBA: 63.64; 35.23 PFBS: 3.85[a]; 1.84[a]	Values at 0.25 and 1 mg/Kg: PFOA: 0.56[a]; 0.65[a] PFOS: 0.32; 0.62	OC: 0.274%	Krippner et al. (2015)
Maize stover	Full-scale field study, PFAA mixture from urban 2x biosolid addition. PFBA: 0.00010 PFBS: 0.00039 PFOA: 0.00128 PFOS: 0.00282	PFBA: 64.8 PFBS: –	PFOA: – PFOS: –	OC: 0.57%	Blaine et al. (2013)
Maize leaves	PFOS: 38.5 PFBS: 0.02	PFBS: 4.00[b]	PFOS: 0.80[b]	na	Navarro et al. (2017)
Maize ears	PFOA: 0–50 PFOS: 0–50	–; –	Values at 0.25 and 1 mg/Kg: PFOA: 0.008; 0.004 PFOS: 0; 0.003	na	Stahl et al. (2009)
Maize grains	PFAA mixture: 0.25 and 1 per each of 10 compounds	Values at 0.25 and 1 mg/Kg: PFBA: 0.133[a]; 0.229[a] PFBS: 0.008[a]; 0.005[a]	Values at 0.25 and 1 mg/Kg: PFOA: –; 0.002 PFOS: –; –	OC: 0.274%	Krippner et al. (2015)
Maize root	PFOS: 38.5 PFBS: 0.02	PFBS: 5.00[b]	PFOS: 8.82[b]	na	Navarro et al. (2017)
Oat straw	PFOA: 0–50 PFOS: 0–50	–	Values at 0.25 and 1 mg/Kg: PFOA: 0.88; 0.69 PFOS: 0.224; 0.150	na	Stahl et al. (2009)
Oat grains	PFOA: 0–50 PFOS: 0–50	–	Values at 0.25 and 1 mg/Kg: PFOA: 0.048; 0.054 PFOS: 0.004; 0.0170	na	Stahl et al. (2009)
Ryegrass, four cuttings	PFOA: 0–50 PFOS: 0–50	–	Range within the four cuttings at 0.25 and 1 mg/Kg: PFOA: 0.128–7.52 PFOS: 0.048–0.47	na	Stahl et al. (2009)
Wheat straw	PFOA: 0–50 PFOS: 0–50		Values at 0.25 and 1 mg/Kg: PFOA: 3.20; 1.90 PFOS: 0.20; 0.27	na	Stahl et al. (2009)
Wheat straw	PFAA mixture from the highest biosolid addition: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	PFBA: 1.64 PFBS: 0.635	PFOA: 0.847 PFOS: 0.270	SOM: 2.76%	Wen et al. (2014)
Wheat grains	PFOA: 0–50 PFOS: 0–50	–	Values at 0.25 and 1 mg/Kg: PFOA: 0.096; 0.009 PFOS: –; –	na	Stahl et al. (2009)
Wheat grains	PFAA mixture from the highest biosolid addition: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	PFBA: 0.48 PFBS: –	PFOA: 0.111 PFOS: 0.062	SOM: 2.76%	Wen et al. (2014)
Wheat husk	PFAA mixture from the highest biosolid addition: PFBA: 0.0135 PFBS: 0.0312 PFOA: 0.0261 PFOS: 0.0408	PFBA: 0.43 PFBS: –	PFOA: 0.160 PFOS: 0.054	SOM: 2.76%	Wen et al. (2014)

na = not available.
Σ = total amount.
[a] No significant difference between the two different concentrations.
[b] Obtained from mean values of t = 0 and t = final soil concentrations.

from 2.56 to 1.64 with increasing rates of biosolid amendment (Wen et al., 2014). The opposite pattern was observed in grains, where the BCFs for PFBA were higher in wheat, ranging from 0.916 to 0.481 with increasing rates of biosolid amendment (Wen et al., 2014), but were on average 0.18 ± 0.07 in maize grown at spiking doses of 0.25 and 1 mg/kg (Krippner et al., 2015) (Fig. 1). PFPeA was also detected in maize grains at both soil concentrations, but was detected in wheat only at the highest biosolid dose. Recently, Liu et al. (2017) reported very high BCFs for PFBA in maize and wheat grains (on average 2.5 and 33.1, respectively) from an area around a mega-fluorochemical industrial park, suggesting that the contribution of PFASs derived from precipitation as well as from irrigation water. According to these authors, the higher BCFs observed in wheat grains compared to maize might be related to their higher protein content, in accordance with Wen et al.'s (2016) results.

Regarding PFBS in straw, the BCFs of maize, i.e. 3.85 and 1.84 at

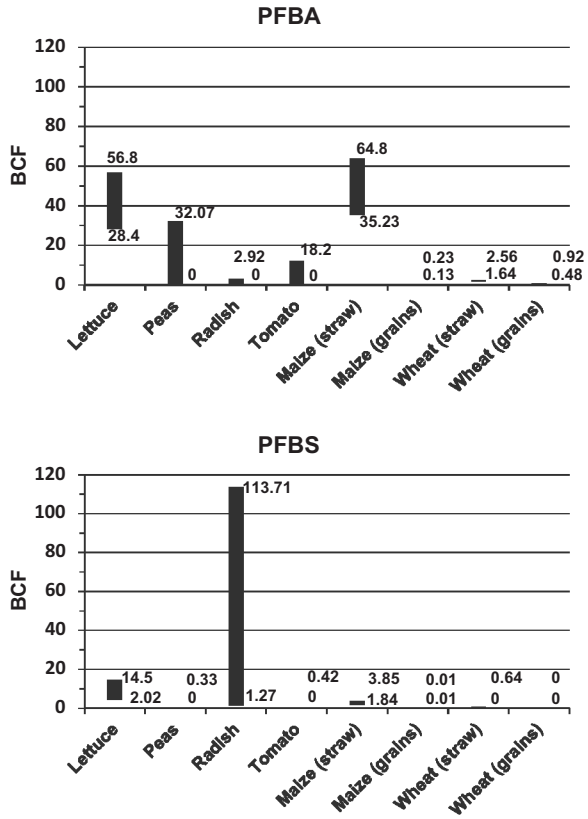


Fig. 1. Bioconcentration factors (BCF) for PFBA and PFBS in vegetable edible plant parts and in maize and wheat straw and grains. Data from Blaine et al., (2013, 2014a) for vegetables, from Krippner et al. (2015) and Blaine et al. (2013) for maize, from Wen et al. (2014) for wheat.

0.25 and 1 mg/kg spiking doses, respectively, (Krippner et al., 2015) were higher than those of wheat, for which the highest value registered was 0.635 (Wen et al., 2014). In grains, PFBS had very low BCFs of 0.008 and 0.005 in maize, and was not detected in wheat by Wen et al. (2014), although Stahl et al. (2013) found PFBS in wheat grains in their lysimeter experiment (448 μg/kg d.w in the first year).

Concerning the C8 PFASs, from Stahl et al.'s (2009) data on maize straw we were able to calculate BCFs of 0.272 and 0.126 for PFOA, and 0.132 and 0.104 for PFOS at 0.25 and 1 mg/kg, respectively. Higher BCFs were reported by Krippner et al. (2015), on average 0.60 ± 0.06 for PFOA, and 0.32 and 0.62 for PFOS at the same 0.25 and 1 mg/kg spiking doses, respectively. BCF values for PFOA in wheat straw were higher than in maize, ranging from 1.54 to 0.847 in Wen et al.'s (2014) experiments, while Stahl et al. (2009) obtained values of 3.20 and 1.90 at the 0.25 and 1 mg/kg doses, respectively, in their trials with wheat. These two authors reported more similar BCFs for PFOS, ranging between 0.20 and 0.27. The BCFs for PFOA and PFOS in maize grains were quite low, ranging from 0 to 0.008 in Stahl et al.'s (2009) and Krippner et al.'s (2015) experiments at the 0.25 and 1 mg/kg spiking doses. The BCFs for PFOA in wheat seeds calculated by Wen et al. (2014) were higher, varying between 0.233 and 0.111, whereas in Stahl et al.'s (2009) experiments they were 0.096 and 0.0090 at the 0.25 and 1 mg/kg spiking doses. BCF values for PFOS obtained from Wen et al.'s (2014) trials were roughly constant with a mean value of 0.0654, whereas from Stahl et al.'s (2009) data they were 0 at both the 0.25 and 1 mg/kg spiking doses.

In summary, overall results regarding the two most important cereal species, maize and wheat, showed a certain variability in BCFs within the same species when comparisons were made between different authors, and between different PFAS spiking concentrations reported by the same authors (see ranges in Fig. 1 for PFBA and PFBS). This confirms the importance of individuating and assessing the parameters that influence PFAS accumulation in plants. Anyway, despite the small number of papers dealing with this topic, it appears that maize tends to accumulate high levels of low-chained fluorinated compounds, particularly PFBA but also PFPeA, in the green parts of the plants to a far greater extent than wheat does. Concerning the C8 compounds, wheat straw seems to accumulate more PFOA than maize straw. Therefore, leaving maize or wheat residues in the soil as amendment in a polluted area may pose serious problems of PFAS spread into the environment, while using them in animal feeds can contaminate the food chain. Conversely, wheat grains and husks, and their derivatives might be a greater direct source of PFBA for humans than maize. However, as there are few papers dealing with these cereals, it is necessary to continue research in order to obtain more data with which additional, more comprehensive analyses can be carried out.

4.2. Vegetables and fruits

Stahl et al. (2009) produced the first report on PFAS accumulation not just in cereals but also in vegetable plants. They analyzed PFOA and PFOS concentrations in potato peel and peeled potato tubers from plants grown in soil spiked with the two chemicals in increasing doses from 0 to 50 mg/kg soil. At 1.0 mg/kg of soil, neither PFOA nor PFOS were detectable in peeled potato tubers, whereas in the fibrous structure (peel) they were found in quantities of 2 and 13 μg/kg fresh weight (f.w.), respectively. Studies have also been made of PFOA and PFOS accumulation in carrot, potato and cucumber plants grown in soil treated with contaminated sewage sludge, and in some cases spiked with the two contaminants (Lechner and Knapp, 2011). This research shows that shoot is again an important site of accumulation, whereas the edible parts of these vegetables were, fortunately, less contaminated, following the hierarchy peeled potatoes < < cucumber < peeled carrots. Carrots were the most contaminated of these three vegetable species (Table 4), probably because of their role in absorbing nutrients and water for the whole plant, whereas potatoes are storage organs and cucumber are fruits. Carrot peel contained roughly the same level of PFASs as peeled carrots. Conversely, in the case of potatoes, the peel contained about 2.3 times the concentration of PFOA and 21 times the concentration of PFOS as the peeled tubers, i.e. 17.6 ± 1.4 and 15 ± 2.2 μg/kg f.w., respectively, at the highest spiking dose. Concentrations similar to those found by Lechner and Knapp (2011) were reported by Cornelis et al. (2012) for potatoes sampled in Belgium. However, the PERFOOD study conducted in four European countries with the purpose of studying the risk to human health due to the presence of PFASs in European foods, found that the sum of PFHxA, PFHpA, PFOA and perfluorononanoic acid (PFNA) levels in starchy roots and tubers were on the scale of tens of nanograms per kg of f.w. (Herzke et al., 2013), i.e. approximately two orders of magnitude lower than the PFOA concentrations found by Lechner and Knapp (2011). Bizkarguenaga et al. (2016) used LUFA 2.4 soil (LUFA Speyer, Speyer, Germany) mixed with compost spiked with PFOA and PFOS at levels similar to those used by Lechner and Knapp (2011), and found comparable amounts of these two compounds in carrot peel and cores, with PFOS in generally higher quantities than PFOA. The authors also reported lower plant accumulation of the two PFASs when a commercial universal substrate was used, which they attributed to its higher organic matter content (53% vs 2.3% of total organic carbon). Large differences in the PFAS concentrations between the two carrot varieties studied were found, but only with the commercial universal substrate for PFOA (121 ± 36 μg/kg in *Chantenay vs* 41 ± 11 μg/kg in *Nantesa*), and with the LUFA 2.4 soil for PFOS (240 ± 22 in *Chantenay* vs 162 ± 17 in *Nantesa*), indicating different effects of soil characteristics on PFAS uptake by the two varieties.

Other studies on PFAS accumulation in vegetables were conducted with lettuce leaves and tomato fruits (Blaine et al., 2013), and in different organs of radish, celery, tomato, and sugar snap pea plants

Table 4
Initial PFAS concentrations (mg/kg soil) and their amount in different vegetable plant parts (μg/kg d.w.).

Plant Species and parts	Compounds	Type of treatment and initial soil concentrations	Concentrations in plant tissues	References
Carrots (peeled)	PFOA; PFOS	Soil + spiked biosolids.	PFOA: 31.3[a]; 333[b]	Lechner and Knapp (2011)
	Tub 1	0.681; 0.010	PFOS: 0.5[a]; 5.3[b]	
Carrots (peeled)	PFOA; PFOS	Soil + spiked biosolids.	PFOA: 30.8[a]; 328[b]	Lechner and Knapp (2011)
	Tub 2	0.676; 0.458	PFOS: 18.4[a]; 196[b]	
Carrots (peeled), *Chantenay* variety	PFOA and PFOS in separate pots	Soil 2.4 + spiked compost. 0.528; 0.445	PFOA: 148 PFOS: 240	Bizkarguenaga et al. (2016)
Carrots (peeled), *Nantesa* variety	PFOA and PFOS in separate pots	Soil 2.4 + spiked compost. 0.485; 0.335	PFOA: 144 PFOS: 162	Bizkarguenaga et al. (2016)
Celery shoots	PFAA mixture	Soil + biosolids. Industrially impacted: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASs = 0.329;	PFBA: 231.69 PFBS: 107.13 PFOA: 55.40 PFOS: 69.27 ΣPFASs = 817.3	Blaine et al. (2014a)
Celery shoots	PFAA mixture	Soil + biosolids. Municipal: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASs = 0.427	PFBA: < LOQ PFBS: 4.49 PFOA: 1.99 PFOS: 17.21 ΣPFASs = 39.3	Blaine et al. (2014a)
Cucumbers Tub 1	PFOA; PFOS	Soil + spiked biosolids. 0.406; 0.010	PFOA: 11.3[a]; 323[c] PFOS: ND	Lechner and Knapp (2011)
Cucumbers Tub 2	PFOA; PFOS	Soil + spiked biosolids. 0.805; 0.556	PFOA: 23.8[a]; 680[c] PFOS: 1.3[a]; 37.1[c]	Lechner and Knapp (2011)
Lettuce leaves	PFAA mixture	Soil + biosolids. Industrially impacted: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASs = 0.329	PFBA: 266.08 PFBS: 205.24 PFOA: 197.91 PFOS: 82.90 ΣPFASs = 1245	Blaine et al. (2013)
Lettuce leaves	PFAA mixture	Soil + biosolids. Municipal: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASs = 0.427	PFBA: 25.50 PFBS: 3.03 PFOA: 20.01 PFOS: 101.62 ΣPFASs = 240	Blaine et al. (2013)
Lettuce leaves	Field-scale trial plots PFAA mixture	Soil + biosolids, 4× PFBA: 0.00069 PFBS: 0.00080 PFOA: 0.00517 PFOS: 0.01391 ΣPFASs = 0.03477	PFBA: 27.5 PFBS: 1.62 PFOA: < LOQ PFOS: 1.39 ΣPFASs = 47.43	Blaine et al. (2013)
Lettuce leaves	PFOA and PFOS in separate pots	Soil 2.4 + spiked compost: 0.560; 0.508	PFOA: 1029 PFOS: 77	Bizkarguenaga et al. (2016)
Pea fruits	PFAA mixture,	Soil + biosolids. Industrially impacted: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASs = 0.329	PFBA: 150.14 PFBS: 16.18 PFOA: 2.65 PFOS: 1.28 ΣPFASs = 236	Blaine et al. (2014a)
Pea fruits	PFAA mixture	Soil + biosolids. Municipal: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASs = 0.427	PFBA: < LOQ PFBS: < LOQ PFOA: < LOQ PFOS: < LOQ ΣPFASs ≤ LOQ	Blaine et al. (2014a)
Potatoes (peeled)	PFOA, PFOS	Spiked soil. 0–50 for each compound	At 1 mg/Kg soil: PFOA: 0 PFOS: 0	Stahl et al. (2009)
Potatoes (peeled)	PFOA; PFOS	Soil + spiked biosolids.	PFOA: 2.9[a]; 17.9[b]	Lechner and Knapp (2011)
	Tub 1	0.276; 0.015	PFOS: < LOD	
Potatoes (peeled)	PFOA; PFOS	Soil + spiked biosolids.	PFOA: 7.7[a]; 47.5[b]	Lechner et al. (2011)
	Tub 2	0.795; 0.317	PFOS: 0.7[a]; 4.3[b]	
Radish roots	PFAA mixture	Soil + biosolids. Industrially impacted: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASs = 0.329	PFBA: 13.67 PFBS: 61.89 PFOA: 66.89 PFOS: 34.86 ΣPFASs = 279	Blaine et al. (2014a)

(*continued on next page*)

Table 4 (*continued*)

Plant Species and parts	Compounds	Type of treatment and initial soil concentrations	Concentrations in plant tissues	References
Radish roots	PFAA mixture	Soil + biosolids. Municipal: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASs = 0.427	PFBA: < LOQ PFBS: 23.88 PFOA: 8.11 PFOS: 21.03 ΣPFASs = 79.3	Blaine et al. (2014a)
Spinach	PFAA mixture	Soil + biosolids W1: PFBA: 0.00045 PFBS: ND PFOA: 0.00019 PFOS: 0.00035 ΣPFASs = 0.00144	PFBA: ND PFBS: ND PFOA: 2.37 PFOS: 1.62 ΣPFASs = 5.33	Navarro et al. (2017)
Spinach	PFAA mixture	Soil + biosolids W2: PFBA: 0.00043 PFBS: ND PFOA: 0.00018 PFOS: 0.00022 ΣPFASs = 0.00096	PFBA: ND PFBS: ND PFOA: ND PFOS: 0.99 ΣPFASs = 0.99	Navarro et al. (2017)
Tomato fruits	PFAA mixture	Soil + biosolids. Industrially impacted: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASs = 0.329	PFBA: 56.11 PFBS: 19.38 PFOA: 8.81 PFOS: < LOQ ΣPFASs = 337	Blaine et al. (2013)
Tomato fruits	PFAA mixture	Soil + biosolids. Municipal: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASs = 0.427	PFBA: < LOQ PFBS: < LOQ PFOA: < LOQ PFOS: < LOQ ΣPFASs = 21.4	Blaine et al. (2013)
Tomato fruits	Field-scale trial plots PFAA mixture	Soil + biosolids, 4× PFBA: 0.00069 PFBS: 0.00080 PFOA: 0.00517 PFOS: 0.01391 ΣPFASs = 0.03477	PFBA: 12.56 PFBS: < LOQ PFOA: < LOQ PFOS: < LOQ ΣPFASs = 37.7	Blaine et al. (2013)
Tomato fruits	PFAA mixture	Soil + biosolids W1: PFBA: 0.00074 PFBS: ND PFOA: 0.0012 PFOS: 0.00047 Σ = 0.00782	PFBA: 12.45 PFBS: ND PFOA: 0.18 PFOS: 0.03 ΣPFASs = 61.30	Navarro et al. (2017)
Tomato fruits	PFAA mixture	Soil + biosolids W2: PFBA: 0.00007 PFBS: 0.00013 PFOA: 0.00024 PFOS: 0.00030 ΣPFASs = 0.00088	PFBA: 2.48 PFBS: ND PFOA: ND PFOS: ND ΣPFASs = 3.47	Navarro et al. (2017)

LOD = limit of detection.
LOQ = limit of quantitation.
ND = not detected.
Σ = total amount.
[a] On a fresh weight basis.
[b] On a dry weight basis calculated assuming the % dry matter values of 9.4 and 16.2 (author's data) for peeled carrots and potatoes, respectively.
[c] Calculated on a dry weight basis assuming a 3.5% dry matter content (http://nut.entecra.it/646/tabelle_di_composizione_degli_alimenti.html).

(*Pisum sativum* var. *macrocarpon*) grown in soil amended with municipal biosolids or industrially impacted municipal biosolids (Blaine et al., 2014a). Examination of the concentrations of different PFAAs, consisting of C4 to C10 PFCAs and PFSAs, revealed much higher levels of contamination in plants grown in soil amended with biosolids, particularly those containing industrial residues (Table 4), compared with control soil. For example, PFOS concentrations were < LOQ in lettuce grown in control soil, 82.90 ± 15.93 μg/kg in soil amended with municipal biosolids, and 101.62 ± 5.80 μg/kg in soil amended with industrially impacted municipal biosolids, confirming sludges as potential sources of PFASs in plants. Similar conclusions were reached by Navarro et al. (2017), who detected PFASs in anaerobically digested thermal drying sludge (W1) and in anaerobically digested municipal solid waste compost (W2). They found high rates of PFBA and PFPeA accumulation in fruits of tomato plants grown in amended soil, especially with W1 (Table 4). Analyses carried out by Blaine et al., (2013, 2014a) of the bioconcentration factors for plants grown in soil amended with industrially impacted municipal biosolids revealed that PFBA had the highest values in almost all the organs of the lettuce, radish, celery and pea plants examined, increasing from the roots to the aboveground part of the plants, and reaching the highest levels in the edible parts of the lettuce (56.8 ± 3.45), celery (49.49 ± 4.50) and pea (32.07 ± 32.07) plants, and in tomato shoot (26.03 ± 2.28). However, tomato fruits had a slightly higher BCF for PFPeA than for PFBA (17.06 ± 3.74 *vs* 12.16 ± 1.71). Conversely, in the aboveground parts of these plants, PFOA and PFOS tended to have somewhat lower BCF values (Table 5). BCFs for plants grown in soil amended with industrially impacted municipal biosolids were generally higher than

Table 5
Vegetable plant part analyzed, initial PFAS concentrations (mg/kg soil), soil or sludge organic carbon (OC) contents and bioconcentration factors (concentration in plant parts/initial concentration in soil).

Plant species and parts	Compounds and initial concentrations (mg/kg)	PFBA, PFBS bioconcentration factors	PFOA, PFOS bioconcentration factors	Soil or sludge OC contents	References
Carrots (peeled)	PFOA: 0.681 PFOS: 0.010	–	PFOA: 0.49[a] PFOS: 0.53[a]	na	Lechner et al. (2011)
Carrots (peeled)	PFOA: 0.676 PFOS: 0.458	–	PFOA: 0.49[a] PFOS: 0.43[a]	na	Lechner et al. (2011)
Carrots (peeled), *Chantenay* variety	PFOA: 0.528 PFOS: 0.445 in separate pots	–	PFOA: 0.28 PFOS: 0.55	Soil OC: 4.9%[b]	Bizkarguenaga et al. (2016)
Carrots (peeled), *Nantesa* variety	PFOA: 0.485; PFOS: 0.335 in separate pots	–	PFOA: 0.30 PFOS: 0.49	Soil OC: 4.9%[b]	Bizkarguenaga et al. (2016)
Celery shoots	PFAA mixture, from Industrially impacted biosolids: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASS = 0.329	PFBA: 49.49 PFBS: 2.21	PFOA: 0.71 PFOS: 1.39	Soil OC: 2.24%	Blaine et al. (2014a)
Celery shoots	PFAA mixture from Municipal biosolids: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASS = 0.427	PFBA: – PFBS: 21.4	PFOA: 0.13 PFOS: 0.05	Soil OC: 6.34%	Blaine et al. (2014a)
Cucumbers Tub 1	PFOA: 0.406 PFOS: 0.010	–	PFOA: 0.79[b] PFOS: –	na	Lechner et al. (2011)
Cucumbers Tub 2	PFOA: 0.805 PFOS: 0.556	–	PFOA: 0.85[b] PFOS: 0.067[b]	na	Lechner et al. (2011)
Lettuce leaves	PFAA mixture from Industrially impacted biosolids: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASS = 0.329	PFBA: 56.8 PFBS: 4.22	PFOA: 2.52 PFOS: 1.67	Soil OC: 2.24%	Blaine et al. (2013)
Lettuce leaves	PFAA mixture from Municipal biosolids: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASS = 0.427	PFBA: 28.4 PFBS: 14.5	PFOA: 1.34 PFOS: 0.32	Soil OC: 6.34%	Blaine et al. (2013)
Lettuce leaves	Field-scale trial plots, PFAA mixture from biosolid addition 4×: PFBA: 0.00069 PFBS: 0.00080 PFOA: 0.00517 PFOS: 0.01391 ΣPFASS = 0.03477	PFBA: 40.0 PFBS: 2.02	PFOA: – PFOS: 0.10	Soil OC: 3.51%	Blaine et al. (2013)
Lettuce leaves	PFOA: 0.560 PFOS: 0.507 in separate pots	–	PFOA: 1.85 PFOS: 0.15	Soil OC: 4.9% [b]	Bizkarguenaga et al. (2016)
Pea fruits	PFAA mixture from Industrially impacted biosolids: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASS = 0.329	PFBA: 32.07 PFBS: 0.33	PFOA: 0.03 PFOS: 0.03	Soil OC: 2.24%	Blaine et al. (2014a)
Pea fruits	PFAA mixture from Municipal biosolids: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASS = 0.427	PFBA: – PFBS: –	PFOA: – PFOS: –	Soil OC: 6.34%	Blaine et al. (2014a)
Potatoes (peeled)	PFOA: 0 − 50 PFOS: 0 − 50	–	Values at 0.25 and 1 mg/Kg: PFOA: 0; 0 PFOS: 0; 0	na	Stahl et al. (2009)
Potatoes (peeled) Tub 1	PFOA: 0.276 PFOS: 0.015	–	PFOA: 0.065[b] PFOS: –	na	Lechner et al. (2011)
Potatoes (peeled) Tub 2	PFOA: 0.795 PFOS: 0.317	–	PFOA: 0.045[b] PFOS: 0.010[b]	na	Lechner et al. (2011)

(*continued on next page*)

Table 5 (*continued*)

Plant species and parts	Compounds and initial concentrations (mg/kg)	PFBA, PFBS bioconcentration factors	PFOA, PFOS bioconcentration factors	Soil or sludge OC contents	References
Radish roots	PFAA mixture from Industrially impacted biosolids: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASS = 0.329	PFBA: 2.92 PFBS: 1.27	PFOA: 0.85 PFOS: 0.70	Soil OC: 2.24%	Blaine et al. (2014a)
Radish roots	PFAA mixture from Municipal biosolids: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASS = 0.427	PFBA: – PFBS: 114	PFOA: 0.54 PFOS: 0.066	Soil OC: 6.34%	Blaine et al. (2014a)
Tomato fruits	PFAA mixture from Industrially impacted biosolids: PFBA: 0.00468 PFBS: 0.04858 PFOA: 0.07852 PFOS: 0.04966 ΣPFASS = 0.329	PFBA: 12.2 PFBS: 0.42	PFOA: 0.11 PFOS: –	Soil OC: 2.24%	Blaine et al. (2013) and Blaine et al. (2014a)
Tomato fruits	PFAA mixture from Municipal biosolids: PFBA: 0.00090 PFBS: 0.00021 PFOA: 0.01491 PFOS: 0.31949 ΣPFASS = 0.427	PFBA: – PFBS: –	PFOA: – PFOS: –	Soil OC: 6.34%	Blaine et al. (2013) and Blaine et al. (2014a)
Tomato fruits	Field-scale trial plots, PFAA mixture from biosolid addition 4×: PFBA: 0.00069 PFBS: 0.00080 PFOA: 0.00517 PFOS: 0.01391 ΣPFASS = 0.03477	PFBA: 18.2 PFBS: –	PFOA: – PFOS: –	Soil OC: 3.51%	Blaine et al. (2013)

na = not available.

[a] Calculated on a dry weight basis from Table 4 data.

[b] Calculated taking into account the % OC content of both soil and spiked compost, and their mixture ratio.

Table 6
Bioconcentration factors calculated for lettuce and celery grown in soil amended with industrially impacted biosolids or municipal biosolids.

Compound	Soil + industrially impacted biosolids Lettuce leaves	Soil + municipal biosolids Lettuce leaves	Soil + industrially impacted biosolids Celery shoots	Soil + municipal biosolids Celery shoots
PFBA	56.8	28.4	49.5	< LOQ
PFPeA	20.4	10.2	12.8	1.12
PFHxA	9.90	11.7	11.9	3.00
PFHpA	2.66	3.33	2.51	0.66
PFOA	2.52	1.34	0.71	0.13
PFNA	2.85	0.77	0.69	0.27
PFDA	0.52	0.34	0.32	0.20
PFBS	4.22	14.5	2.21	21.4
PFHxS	7.56	1.08	2.31	0.07
PFOS	1.67	0.32	1.39	0.05

Values taken or calculated from soil and plant data of the greenhouse study of Blaine et al. (2013, 2014a).

when the soil was amended with municipal biosolids, with the exception of PFBS, for which celery and lettuce plants treated with municipal biosolids had the higher BCF values (Table 6; Blaine et al., 2013; Blaine et al., 2014a). This clearly shows that the amount of PFASs accumulated from biosolid-treated soils by the same plant species depends on the kind of sludge utilized, not only with respect to the total and individual concentrations of these pollutants, but also because different biosolids have different effects on the availability and/or uptake of individual PFASs by plants. Blaine et al. (2013) hypothesized that differences in the bioavailability of PFAAs may also be due to the different nature of the organic carbon of the two kinds of biosolids.

Hydroponic studies on the uptake of perfluorinated chemicals are probably more realistic when they concern vegetable plants rather than cereals as many vegetables can be grown in this manner. However, calculating BCFs and comparing them with those obtained from plants grown in soil is not easy owing to the changes in nutrient solution volumes, and consequently in PFAS concentrations, due to evapotranspiration. Felizeter et al. (2012, 2014) studied PFAS accumulation in lettuce, tomato, cabbage and zucchini plants grown in a nutrient solution containing 14 PFASs, comprising the C4 to C14 PFCAs, and the C4, C6 and C8 PFSAs. Lettuce tended to accumulate the lighter C4 - C7 chemicals in the leaf, in accordance with Blaine et al. (2013), but PFOA and PFCAs with higher chain lengths tended to accumulate in the roots. Likewise, the three PFSAs considered accumulated mainly in the roots, again in increasing percentages with increasing chain lengths. The higher sorption of longer-chained PFASs to plant organic components, and their reduced ability to cross the Casparian strip have been put forward to explain these results (Felizeter et al., 2012). The same effect of chain length on PFAS distribution across plant organs was also found in tomato, cabbage and zucchini plants, with the longer-chained chemicals having a preference for the roots, the shorter-chained ones for the leaves and fruits or heads, see Fig. 2 for the mass distribution of

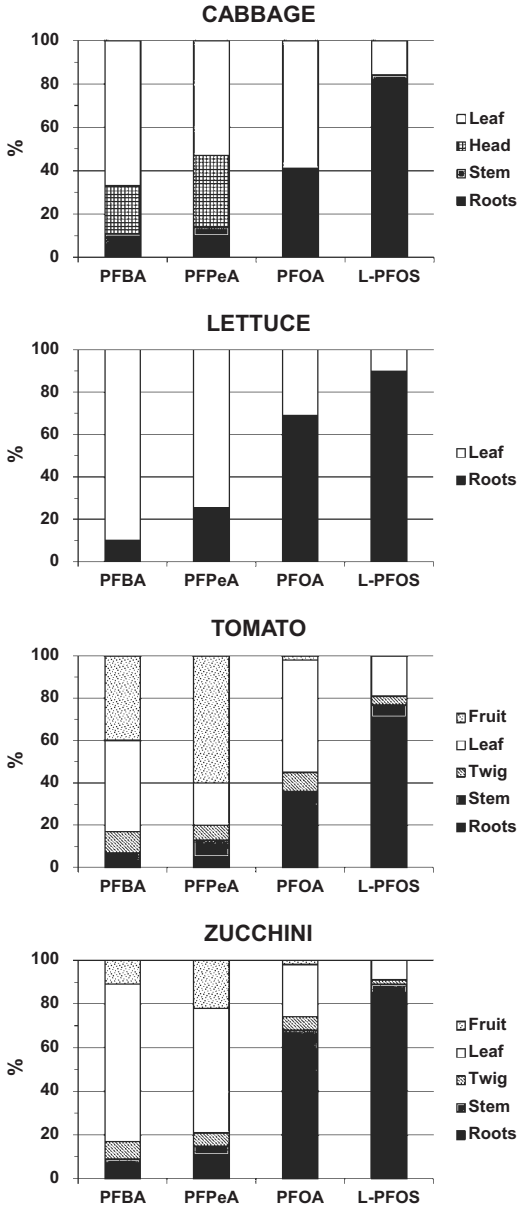


Fig. 2. Mass distribution of some PFAAs in different tissues of cabbage, lettuce, tomato and zucchini plants, expressed as percent of the total amount of PFAA found in the plant. Data from Felizeter et al. (2012, 2014).

some PFAAs in different tissues of cabbage, lettuce, tomato and zucchini plants (Felizeter et al., 2012, 2014). However, it is important to point out that PFAS distribution across roots, stem, leaf, fruit and head varied among species, as seen in Blaine et al.'s (2013; 2013 a) experiments and in cereals. PFOA and PFOS spiked at a nominal concentration of 1 μg/L, with several renewals of the treatment solutions, were accumulated, respectively, to 1.55 ± 0.61 and 0.50 ± 0.31 μg/kg f.w. in lettuce leaves, 0.34 ± 0.10 and 0.10 ± 0.041 μg/kg f.w. in cabbage head, 0.29 ± 0.06 and 0.12 ± 0.023 μg/kg f.w. in zucchini fruit, and 0.28 ± 0.035 μg/kg f.w. and < LOQ in tomato fruit (Felizeter et al., 2012, 2014). It is important to mention that the authors concluded that the quantities of PFOA and PFOS measured in these plants were well below the levels that could pose a risk to human health, as established by the European Food Safety Authority (2008) tolerable daily intake (TDI) values for PFOA and PFOS, i.e. 1500 ng/kg body weight and 150 ng/kg body weight, respectively (EFSA, 2008). Recently, the US EPA specified the lower oral non-cancer reference doses (RfDs) of both PFOS and PFOA at 0.00002 mg/kg/day = 20 ng/kg/day (EPA, 2016a, 2016b). To exceed these values, a person of 70 kg body weight would need to eat about 0.9 kg (for PFOA) and 2.8 kg (for PFOS) of lettuce grown at a concentration of 1 μg/L. At this spiking dose, PFBA accumulated to a greater extent than PFOA and PFOS, that is, to 11.7 ± 2.4 μg/kg f.w. in lettuce, 7.21 ± 3.09 μg/kg f.w. in cabbage, 5.56 ± 1.12 μg/kg f.w. in tomato, and 1.24 ± 0.11 in zucchini. PFPeA accumulated to a similar or greater extent than PFBA in all the vegetables except lettuce. The C4 PFBS accumulated to a lesser extent than PFBA, as is the case with cereals, with a mean concentration of 0.61 ± 0.26 μg/kg f.w. in the vegetable species analyzed (Felizeter et al., 2012, 2014).

In the context of the European project PERFOOD, D'Hollander et al. (2015) found that PFAA detection rates in fruits were generally higher than in other food groups (cereals, sweets, salts), with values depending on species and origin. The most frequently detected PFASs were PFOA followed by PFOS, their concentrations in fruits ranging from < LOQ to 139 and 539 ng/kg f.w., respectively. Dietary intake assessments for PFOA and PFOS made by these authors showed that the daily intake of PFASs was far below the current tolerable levels established by the EFSA. Regarding the short-chain compounds, Klenow et al. (2013) found that fruits accounted for 39%, 58%, 60% and 20% of PFHxA total dietary intake in the Czech Republic, Italy, Norway and Belgium, respectively. The PERFOOD studies contain no data on PFBA and PFPeA, which can accumulate in plants to a greater extent than PFHxA (Table 6).

4.3. Parameters affecting PFAS accumulation in plants

As far as we know, Higgins and Luthy (2006) were the first researchers to point to organic carbon content as a dominant parameter affecting PFAS sorption to sediments. Zhao et al. (2011) showed that soil organic matter, followed by cation exchange capacity (CEC), has the greatest influence on PFOA and PFOS phytotoxicity for *Brassica chinensis* plants. In a soil leaching experiment, Gellrich et al. (2012) found strong adsorption of PFASs to sewage sludge, which they attributed to its high organic carbon content. Zhao et al. (2016) compared PFAA sequestration and bioavailability in farmland soil with two peat soils, and showed that the higher the soil organic matter content, the higher the sequestration of these substances by soils. It must be added that PFAAs were not irreversibly sequestrated, as they were accumulated to some extent by earthworms. Still regarding the role of soil organic matter, Wen et al. (2014) found that BCF values were less variable if they were calculated on the basis of soil organic matter. Likewise, Bizkarguenaga et al. (2016) reported lower plant accumulation of PFOA and PFOS when a substrate rich in organic matter was used. However, Blaine et al. (2014 b) studied the accumulation in lettuce of PFAS from soils differing in their organic carbon contents (0.4%, 2%, 6%), and found greater accumulation of the short-chained compounds PFBA, PFPeA and PFBS at 2% soil organic content than at 0.4 or 6%. Furthermore, as discussed before, Blaine et al. (2014a) suggested that the nature of organic carbon can also influence PFAS bioavailability to plants. Other soil characteristics can affect the amount of PFAS that can be absorbed by plants. Stahl et al. (2013) attributed the unusual lower accumulation of PFOA compared with PFOS to the higher leaching of PFOA from the soil. Zhao et al. (2014) found that the presence of the redworm *Eisenia fetida* in soil induces increased bioaccumulation of PFCAs with C ≤ 7, and decreased accumulation of those with C > 7 and of PFSAs in wheat plants. A further parameter influencing the amount of PFAS accumulated by plants may be their composition. Gellrich et al. (2012) found that PFBA was strongly adsorbed by soil only in the absence of long carbon chain PFAS, probably because of competition for sites of adsorption.

Further environmental parameters that can affect PFAS absorption by plants are temperature and salinity. Zhao et al. (2013, 2016) showed that increases in these parameters induce higher PFCA and PFOS uptake and translocation in wheat plants grown in hydroponics. High

temperatures induce higher transpiration rates, which can lead to higher contaminant uptake. High salinity may trigger increased partitioning of PFAS into the organic components of the roots with a salting-out mechanism, as suggested by You et al. (2010) to partially justify the higher sorption of PFOS onto sediments at increasing $CaCl_2$ levels. Regarding the effect of pH, Zhao et al. (2013) found the highest PFOS accumulations in wheat plants grown in nutrient solutions at pH values ranging from 6 to 8, with lower uptake at pH 4 and particularly at pH 10. In contrast, Krippner et al. (2014) did not observe pH dependence for PFOS uptake by maize plants grown in nutrient solution, but found that PFDA accumulated to a greater extent at pH 5 than at pH 7, while the influence of this parameter on other PFCAs was inconsistent. Wen et al. (2015) found soil pH to play a relatively unimportant role in PFOS and PFOA bioavailability to earthworms in biosolid amended soil. Similar views were expressed by Zhao et al. (2011) regarding the role of soil pH and CEC on PFOS and PFOA toxicity for *Brassica chinensis* plants. Additional insights by Blaine et al. (2014 b) suggest that the mobility and bioavailability of PFAAs may be greater when delivered with irrigation water than through biosolid-amended soil. To conclude, Li et al. (2018) recently reviewed available data on the role of soil and sediment properties in determining PFAS sorption and argued that the behavior of these substances in the environment is more complex than can be explained by a single soil or sediment property.

4.4. Challenges for the future and research needs

As discussed previously, PFASs are highly persistent compounds. Even the recently-adopted fluorinated alternatives and their precursors are highly persistent and environmentally mobile, so even if emissions cease in the future, it will take decades or even centuries to reverse the global contamination of short-chain PFAAs (Wang et al., 2015). Owing to the current unfeasibility of removing PFASs from foods, the first challenge to growing safer agricultural products must be to identify the sources of contamination in irrigation water and soil. As already mentioned, waters and soils close to airports, fire-training locations, industrial sites and landfills are particularly at risk of PFAS pollution. However, the use of biosolids on agricultural soil must also be considered a potential source of agricultural plant contamination. In polluted areas, to reduce dietary intake of PFAS, it is important to adopt agricultural measures that decrease their absorption by plants, in particular the addition of unpolluted manure to the soil, as many studies provide evidence for a decrease in PFAS accumulation in plants at increasing levels of soil organic matter. Furthermore, the use of plant residues, such as maize stover, in animal feeds should be avoided as this practice can contaminate the food chain. Growing vegetables in hydroponics should also be discouraged in polluted areas, as it allows pollutants to interact directly with plants in the absence of soil and its ability to adsorb pollutants. The adoption of phytoremediation techniques may counter the spread of PFASs in the environment, as it has been shown that tree plants and constructed wetlands are able to remove these pollutants (Gobelius et al. (2017); Yin et al. (2017). However, as discussed previously, more studies are needed, as many research results display discrepancies regarding the tendency of the same plant species to accumulate PFASs, and the role of soil components in PFAS bioavailability. There is also the need to fill the gaps in the knowledge of the accumulation capacity of PFAS by other plant species. The role of volatile PFAS precursors in the accumulation of these substances by plants also needs to be clarified. The general variability observed in the examined data further complicates our understanding of how PFASs interact with soils and plants.

5. Conclusions

Current research suggests that the spread of PFASs in agricultural soil is mainly a result of irrigation with contaminated water, the use of polluted sewage sludges or industrial wastes as soil conditioners. Agricultural soils especially at risk may be those close to airports and fire-training locations. PFASs are absorbed by plants to different extents according to their concentrations, chain lengths, functional group, plant species and variety, growth media (hydroponics vs. soil), and soil and biosolid characteristics. In particular, the abundance and characteristics of soil organic matter are considered one of the most important factors. Once inside the plants, partitioning among organs depends on species, and, particularly, on functional group and chain length. The C4–C6 compounds, which have recently replaced C8 PFOA and PFOS in many industrial processes, appear to accumulate particularly in leaves and fruits, whereas the compounds with higher chain lengths tend to be more concentrated in roots. However, owing to the variability of available data, the complexity of the interactions between PFASs and soil components, and between PFASs and plants, further studies need to be undertaken in order to clarify the role of each factor influencing the uptake and distribution of these compounds within plants in order to reduce human exposure to these hazardous substances.

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PFAS

'There are no tears left' Arundel dairy farmer devastated by PFAS fights for state help

State agriculture officials are expected to roll out an income replacement program soon.

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ARUNDEL, Maine — A state income replacement program for Maine farms struggling with financial losses caused by contamination from toxic chemicals, known as PFAS, is expected to be rolled out in a few weeks. But the owner of a York county farm worries about falling through the cracks.

Fred Stone and his wife Laura took over the century-old Stoneridge Farm in the mid-70s, supplying several thousand gallons of milk weekly for Oakhurst dairy. But instead of looking forward to retirement, the couple is stuck in a toxic nightmare.

"We have cried and killed a lot of these cows, and there are no tears left," Stone said.

The family's legacy took a huge hit five and a half years ago when high levels of PFAS chemicals were discovered in his cow's milk, soil, feed, and drinking water. Stone alerted state regulators, pulling milk from the shelves. He eventually had to put down the majority of the farm's Brown Swiss and Holstein cows, which resulted in hundreds of thousands of dollars in lost income.

RELATED: Toxic disaster of PFAS contamination a nightmare for Maine farmers

"We were the ones that brought this forward, and we have paid a hell of a price for this," Stone said.

Earlier this year Maine Department of Agriculture, Conservation, and Forestry officials announced plans for a short-term income replacement program to help farms devastated by PFAS contamination.

Stone sent Nancy McBrady, the director of the Bureau of Agriculture, Food and Rural resources at the DACF, information on expenses and lost income with hopes of receiving financial assistance.

Earlier this month, Stone received an email from McBrady informing him the income replacement program is designed for farms that have recently suffered losses from contamination.

Stone worries he and other farmers whose contamination was discovered before 2022, may fall through the cracks.

"It is a situation where it kills the messenger not the message," Stone explained.

He is also fighting to be compensated for his dead cows, through the US Department of Agriculture's Dairy Indemnity Program.

"They are facing the extreme financial hardship," said Susan Collins, (R) Maine.

At a congressional hearing last month, Senator Susan Collins took US Agriculture Secretary Tom Vilsack to task for not promptly responding to inquiries she made about financial assistance for Maine dairy farmers with contaminated cows.

"We are working with EPA to try to establish a national standard on what is an acceptable level of PFAS, so we can basically help define the level of assistance and help that is required," Vilsack testified.

Jim Britt, the spokesman for DACF tells NEWS CENTER the department plans to roll out the income replacement program in the near future, which will be open to farms with current and prior PFAS contamination. The department says it will also work with Fred to begin the application process.

"The income replacement program is new and getting underway. It is open to farms with current PFAS contamination and those who discovered contamination before 2022. The Department has been working with three currently contaminated farms as we roll out the program. We anticipate the program will continue to evolve, including working with farms that have ceased operations, such as Mr. Stone. This was the context that Director McBrady provided to Mr. Stone in her communication earlier this month. To date, Fred has not applied for the income replacement program. He shared some preliminary information with Director McBrady earlier this year before the income replacement program was fully formulated. The Department will work with Mr. Stone to officially begin the application process and looks forward to finding ways to assist him.

"The new PFAS Fund included in the Governor's budget specifically focuses on assistance to impacted farms. Farms with current and prior PFAS contamination will be able to seek assistance from this Fund, which will have $60M in funding. To the extent the DACF's new income replacement program may not satisfy some farms' needs, the Fund will likely allow for additional support. Maine Farmland Trust and MOFGA also have some funding available to work with impacted farms – you may want to connect with them regarding their ability to support farms with PFAS contamination before 2022.

"In addition to the programs above the DACF and the Maine Department of Environmental Protection currently and have historically worked with farms to ensure that water is clean for both residential and farm use. When a farm is found to have drinking water that exceeds Maine's interim drinking water standard for PFAS, DEP and DACF coordinate with the farm to ensure the appropriate type of filtration system is installed. Sometimes the farm may want to do the installation on its own and seek reimbursement. In these cases, the farm is provided guidance from DEP and DACF as to parameters for design and reimbursement. To be clear, the Department does not want to see any farms fail and understands the tremendous strain on farms that have ceased operations. We intend to work closely with all farms and look forward to working with Mr. Stone. Again, this is a new program, and we appreciate Mr. Stone's patience and perspective about his experience and needs. The Department will contact Mr. Stone to help with the application and begin discussing his needs." Britt said.

But the couple who still raise a small number of cows to show at agriculture fairs in New Hampshire, says no amount of help will make them whole again.

agriculture

MDPI

Review

Making the Connection Between PFASs and Agriculture Using the Example of Minnesota, USA: A Review

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Abstract

Exposure to per- and polyfluoroalkyl substances (PFASs) can cause detrimental health effects. The consumption of contaminated food is viewed as a major exposure pathway for humans, but the relationship between agriculture and PFASs has not been investigated thoroughly, and it is becoming a pressing issue since health advisories are continuously being reassessed. This semi-systematic literature review connects the release, environmental fate, and agriculture uptake of PFASs to enhance comprehension and identify knowledge gaps which limit accurate risk assessment. It focuses on the heavily agricultural state of Minnesota, USA, which is representative of the large Midwestern US Corn Belt in terms of agricultural activities, because PFASs have been monitored in Minnesota since the beginning of the 21st century. PFAS contamination is a complex issue due to the over 14,000 individual PFAS compounds which have unique chemical properties that interact differently with air, water, soil, and biological systems. Moreover, the lack of field studies and monitoring of agricultural sites makes accurate risk assessments challenging. Researchers, policymakers, and farmers must work closely together to reduce the risk of PFAS exposure as the understanding of their potential health effects increases and legacy PFASs are displaced with shorter fluorinated replacements.

Keywords: PFAS; agriculture; environmental fate; knowledge gaps; mitigation; Midwest

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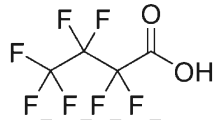
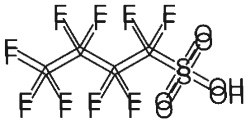
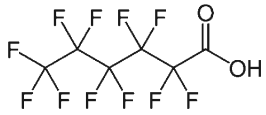
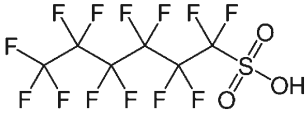
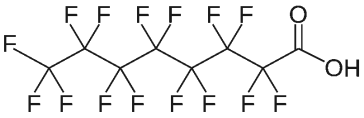
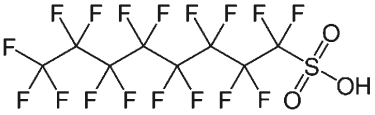
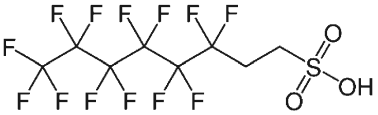
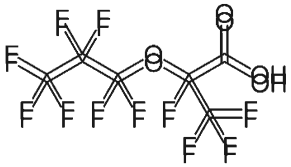
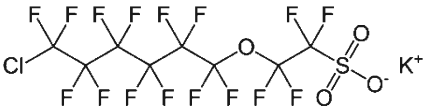
1. Introduction

PFASs are defined as aliphatic substances which contain at least one carbon atom on which all hydrogen atoms have been replaced with fluorine atoms. Perfluoroalkyl substances are substances in which all hydrogen atoms attached to the carbon chain have been substituted by fluorine, and polyfluoroalkyl substances are not fully fluorinated [1]. The Organisation for Economic Co-operation and Development proposes a broader definition by suggesting that any compound containing at least one fully fluorinated methyl or methylene group be referred to as a PFAS [2]. There are currently over 14,000 chemicals listed as PFASs in the United States Environmental Protection Agency toxicity databases [3]. Some of the most relevant representatives are presented in Table 1.

Table 1. A list of the most relevant PFASs. * The ammonium salt of HFPO-DA is also known as GenX.

Name	Abbreviation	Chemical Formula	Structural Formulas
Perfluorobutanoic acid	PFBA	$C_4HF_7O_2$	
Perfluorobutanesulfonic acid	PFBS	$C_4HF_9O_3S$	
Perfluorohexanoic acid	PFHxA	$C_6HF_{11}O_2$	
Perfluorohexanesulfonic acid	PFHxS	$C_6HF_{13}O_3S$	
Perfluorooctanoic acid	PFOA	$C_8HF_{15}O_2$	
Perfluorooctanesulfonic acid	PFOS	$C_8HF_{17}O_3S$	
6:2-Fluorotelomersulfonic acid	6:2 FTS	$C_8H_5F_{13}O_3S$	
Hexafluoropropylene oxide dimer acid *	HFPO-DA *	$C_6HF_{11}O_3$	
6:2 chlorinated polyfluoroalkyl ether sulfonate	F-53B	$C_8ClF_{16}KO_4S$	

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are ubiquitous and can be detected in the environment, wildlife, human serum, and tissue [4–7]. PFASs can accumulate in the human body over time and cause different toxicological effects [8,9], such as cancer, kidney and liver disease, and adverse effects on reproductivity and development [10]. The mean half lives of some of the most common PFASs, namely perfluorooctane sulfonic acid (PFOS), perfluorohexane sulfonic acid (PFHxS), and perfluorooctanoic acid (PFOA), are 5.4 years, 8.5 years, and 3.8 years, respectively [11].

PFASs can be found in many products, such as firefighting foams, food packaging materials, household products, medical devices, pesticide formulations, and surfactants [12]. The widespread industrial and commercial utilization of these substances is based on their chemical and physicochemical properties. The carbon–fluorine bond is the strongest bond in organic chemistry, with a dissociation energy of up to 536 kJ/mol [13]. Therefore, the PFAS family of chemicals shows exceptionally high chemical and thermal stability [14–16]. Due to their polar nature, most PFASs show high solubility in water [17] and are therefore environmentally mobile [18]. Four main activities are responsible for the contamination of PFASs in the air, water, and soil. These include leakages and emissions of PFASs at

manufacturing sites, use of aqueous film-forming foam (AFFF) for firefighting activities, leaching from landfills, and the application of biosolids for wastewater treatment [19–22].

Although PFASs have multiple potential exposure paths to humans, a significant exposure route of concern is the consumption of contaminated food [23]. PFASs can accumulate from the environment in crops and produce that are consumed directly by humans [24], and the PFASs in fodder and grain used to feed livestock can lead to the contamination of the animals and their related food products [25,26]. In Maine, USA, dairy farms were shut down due to PFAS contamination in their products, with PFASs being detected on more than 50 farms [27–30]. Most environmental research has primarily focused on PFAS sources and the fate of these substances in the air, water, and soil [31]. Research on downstream biological systems, such as agricultural crops and livestock, experiencing PFAS contamination is lacking. Understanding the relationship between PFASs in the environment and agriculture is critical as humans are dependent on food production and approximately 44% of habitable land surface is used for agricultural purposes [32].

The research objective of this study is to detail the sources of PFAS contaminations in agriculture, determine the environmental fate of PFASs, and outline the connection to agriculture to fill knowledge gaps. The history, contamination levels, and actions regarding PFASs are discussed for a typical Midwestern state (Minnesota, USA) so that the findings can be put into the historical context, and other agricultural areas can assess their paths towards protecting their agricultural sector from contamination issues. This work aims to make meaningful contributions to public health by reducing the risk of PFAS exposure by suggesting sensible actions based on our findings. Furthermore, we suggest future research directions to close dangerous knowledge gaps.

2. Materials and Methods

A semi-systematic literature was carried out to address the link between PFAS contamination and agriculture. Different databases and search engines (i.e., SciFinder (web-based, Chemical Abstracts Services, Columbus, OH, USA), Scopus (web-based, Elsevier, Amsterdam, Netherlands), Web of Science (web-based, Clarivate, Philadelphia, PA, USA), PubMed (web-based, U.S. National Library of Medicine, Bethesda, MD, USA), Google Scholar (web-based, Google LLC, Mountain View, CA, USA), and ScienceDirect (web-based, Elsevier, Amsterdam, Netherlands)) were utilized to identify relevant peer-reviewed articles. Governmental databases (e.g., Minnesota Groundwater Atlas) and reports (e.g., Minnesota's PFAS Blueprint) were examined from multiple agencies to gather Minnesota-related and other important information. Various single and combined search terms were employed due to the complexity and diversity of the presented research objective. For example, the number of publications per year for a typical query (i.e., PFAS * and agriculture *) is shown in Figure 1. Excluding most sources from Section 3.1.1, the majority of relevant papers were published from 2019 onward and no publications were found prior to 2007 regarding PFAS in agriculture specifically. The exponential growth of the number of publications indicates the increasing scholarly attention regarding this issue to address pressing knowledge gaps and lacking mitigation strategies.

Multiple qualitative criteria were used for the inclusion or exclusion of the respective sources. These encompassed the (1) relevance, (2) originality (primary and field studies were preferred), (3) publication date (papers published between 2000 and 2024 were considered; this does not apply to historic documents and a few exceptions), (4) overall quality (e.g., methodology, sample size, transparency, etc.), and (5) impact (i.e., number of citations or legislative documents).

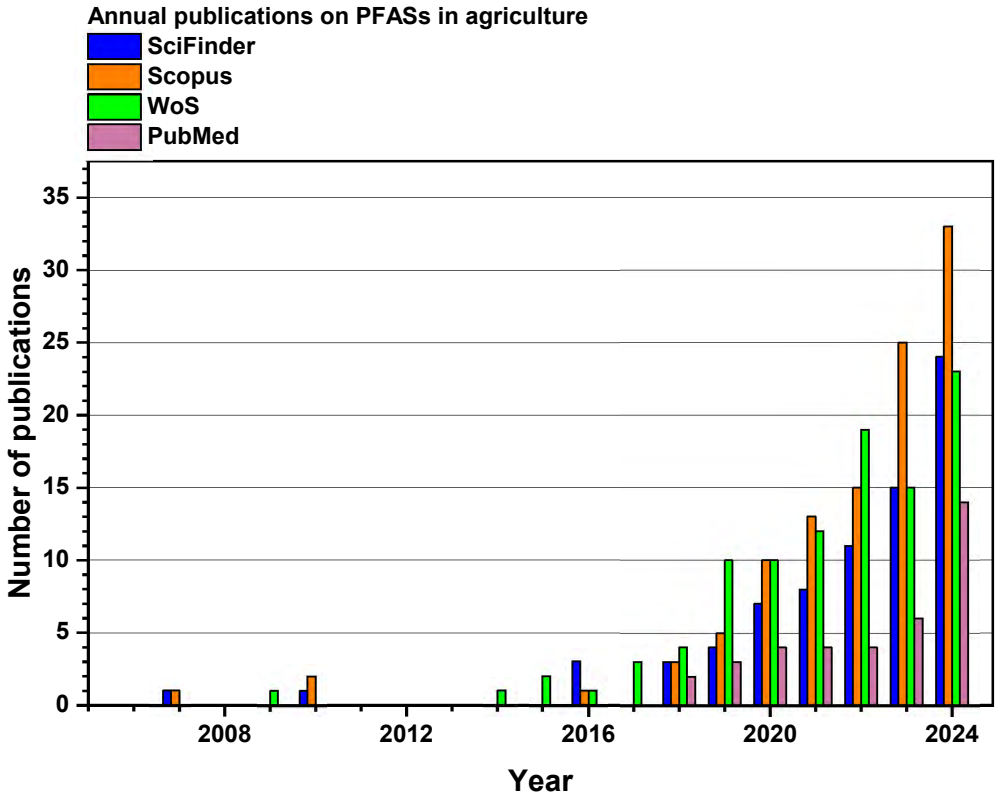


Figure 1. A plot of annual publications for the query "PFAS * AND agriculture *" between 2005 and 2024.

3. Results and Discussions

3.1. PFASs in Minnesota

3.1.1. History of PFASs in Minnesota

Minnesota's experience with PFASs is deeply intertwined with the beginning of PFAS production in the 1940s. PFOS- and PFOA-related substances and other fluorochemicals were manufactured at Minnesota Mining and Manufacturing (officially renamed 3M in 2002) production facility in Cottage Grove, MN [33]. As early as 1955, research indicated that PFOS could impact biological systems, as protein binding properties were observed in PFOS [34]. In 1961, DuPont researchers found that these chemicals can potentially increase the liver size in rats and rabbits [35]. In 1997, 3M was selling almost 1900 metric tons of PFASs in the US. [36]. Waste products, sludges, and wastewater were generated, disposed of at landfills or discharged to the Mississippi River [37]. In 2002, 3M informed the Minnesota Pollution Control Agency (MPCA) that PFOA and PFOS had been detected in groundwater reservoirs used near production and disposal sites [38].

A number of locations linked to PFAS wastes in and around Cottage Grove, MN, were investigated to examine the extent of contamination [39]. While Cottage Grove and the surrounding areas with contamination were more rural areas when 3M began disposal of PFAS wastes, their population grew to become part of the greater Twin Cities (Minneapolis–St. Paul) metropolitan area. According to the MPCA, 3M's manufacturing activities led to the contamination of groundwater covering more than 150 square miles, which affected the drinking water quality of more than 140,000 Minnesotans. Different PFASs were detected at varying concentrations in all samples, including groundwater, surface water, municipal wells, sediment, sludges, influent/effluent, fish, plants, leachates, and gas condensates at landfills [37,39]. Further research analyzed the PFAS contamination in the air [40], fish and bald eagles [41,42], leachate and gas condensates from landfills [43], and yard

waste sites [44]. Statistical evaluations showed that residents of affected communities (e.g., Oakdale) were 30% more likely to be diagnosed with prostate cancer, and infants were 34% more likely to be born with low birth weight compared to unaffected communities [45].

3M decided to terminate the manufacturing of PFOS- and PFOA-related compounds after the initial discovery of contamination [5,46]. In 2010, the Minnesota Attorney General sued over contaminated drinking water and natural resource findings [47]. More recently, 3M announced that it will cease all PFAS manufacturing and stop selling products containing PFASs by 2025 due to the "rapidly evolving external regulatory [. . .] landscape" [48]. Communities are still tremendously concerned about PFAS exposure and the associated health effects and urgently demand that appropriate actions be taken.

3.1.2. Contamination in Minnesota

The detection of major contamination around the Twin Cities metropolitan area led to a larger effort to examine PFAS contamination and spread than is seen in other communities and states. While Cottage Grove was the single most sampled area, rural areas throughout Minnesota were also heavily sampled. Much of the data has been reported to the MPCA, who publishes it as part of their role in investigating and regulating pollution in the state [49]. With this extensive data set, Minnesota provides a temporally and geographically complete example for assessing issues related to PFAS contamination. That said, these datasets are complex and difficult to interpret because of the many unique PFAS chemical species and behaviors, in addition to changing testing protocols as the field has advanced.

Drinking Water Standards as a Guide

The potential human health risks of consuming contaminated drinking water have meant that most Minnesota and nationwide PFAS investigations have concentrated on the contaminant levels in drinking water [49,50]. However, the extent of the problem has meant that Minnesota has expanded testing to surface and groundwater statewide. While the dangers represented by contaminated drinking water are different than agricultural PFAS contamination, water data provides the best guide to understanding contamination in agriculture. The vast PFAS water data from different sites and examining different types of PFASs require that a metric be established to help evaluate locations where contamination levels are potentially detrimental to health.

The Health Index (HI) was adopted by Minnesota in 2002 to evaluate the associated risk of consuming drinking water, and it is continuing to be updated as new data emerges [51]. The HI uses drinking water guidance values for perfluorobutanoic acid (PFBA) (7.000 µg/L), perfluorobutanesulfonic acid (PFBS) (2.000 µg/L), perfluorohexanoic acid (PFHxA) (0.200 µg/L), PFHxS (0.047 µg/L), PFOA (0.035 µg/L), and PFOS (0.015 µg/L) (see Equation (1)) [52].

$$\begin{aligned} HI = ([PFBA])/(7\ \mu g/L) + ([PFBS])/(2\ \mu g/L) + ([PFHxA])/(0.2\ \mu g/L) \\ + ([PFHxS])/(0.047\ \mu g/L) + ([PFOA])/(0.035\ \mu g/L) + ([PFOS])/(0.015\ \mu g/L) \end{aligned} \quad (1)$$

If the HI is greater than 1, the drinking water is considered to have potentially adverse health implications. More information on the regulation of PFAS contamination levels is provided in Section 3.5.

As of July 2024, 919 Community Water Systems out of 966 have been tested by the MPCA, with 5 exceeding the proposed HI. The contaminated Community Water Systems are in larger urban areas, except for Swanville, a small community in central Minnesota. Most Community Water Systems showing PFAS contamination are located in the Twin Cities metropolitan area, which can be linked to 3M production in Cottage Grove (see Section 3.1.1) [49].

The geospatial distribution of wells that contain at least one PFAS that exceeds health-based guidance values and their proximity to wellhead protection areas, which surround public water supply wells and where groundwater contributes to the respective wells, are shown in Figure 2. Out of 13,884 locations/timepoint sampling combinations, 17% contained at least one exceedance and 4189 total exceedances were detected [53]. These were found both in urban and rural/agricultural regions (see Figure 2).

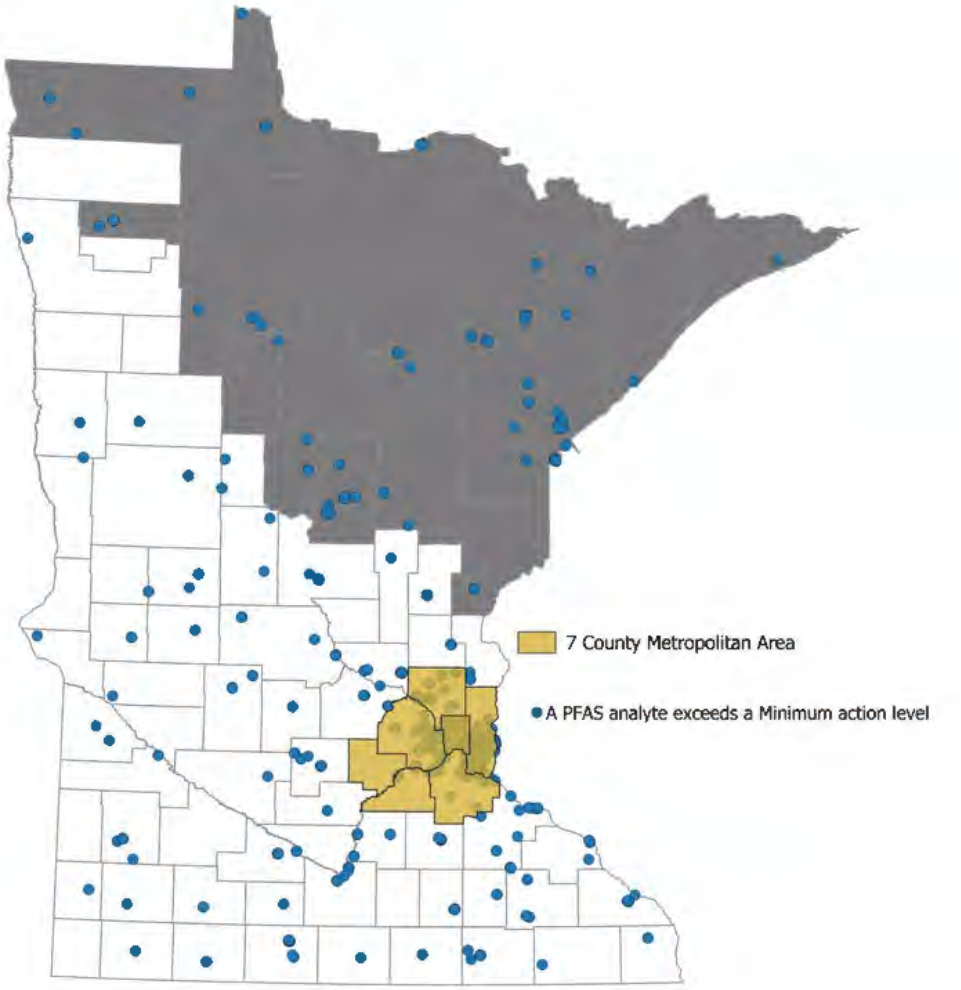


Figure 2. PFAS exceedance data obtained from the MPCA by request. Regions in black represent counties with less than 2.7% of county land area used for corn production. The Twin Cities metropolitan area is excluded. White regions represent counties with greater than 2.7% of county land area used for corn production.

Air Pollution

The first extensive study on PFAS levels in ambient air was published by the MPCA in April of 2022 [40]. Researchers sampled the ambient air at four different sites. Three sites were located near PFAS-emitting sites in urban areas (Duluth, St. Louis Park, and Eagan), while the fourth site (Grand Portage) was in a rural area and was expected to be a low-PFAS reference site. During the observation duration, 17 different PFASs were detected, varying in concentration and time of observation. The MPCA found PFBA, PFBS, PFOA, and PFOS in all air samples, with PFBA contributing approximately between 47% and 70% of the total PFAS concentration. The authors attribute the abundant occurrence of PFBS to the direct emission and degradation of precursors in the atmosphere. Precursors are used as intermediates in the manufacturing process of surfactants or fluoropolymers. They are also used as substitutes for chlorofluorocarbons, coolants, solvents, or fire suppressors [54].

Additionally, 6:2 fluorotelomer sulfonate is known to replace PFOS at chrome plating facilities (e.g., at the St. Louis Park site) and is additionally applied in AFFFs. Therefore, 6:2 fluorotelomer sulfonate was regularly detected in ambient air [40].

Though Grand Portage was chosen due to its remote location, low population (684 in 2019), and limited industry, it showed the second highest airborne PFAS contamination with a mean concentration of about 100 pg/m^3. The authors hypothesize that this surprising finding is likely due to emissions from the local fire department and the solid waste transfer station. They also speculate that long-distance atmospheric transport might contribute to the relatively high concentrations. The findings that the rural community of Grand Portage could have high detectable levels of airborne PFAS indicates that rural/agricultural areas should not assume that their relative isolation is potentially sufficient to limit exposure to airborne PFASs.

3.2. Significant Sources of PFASs in the Environment

3.2.1. Release of PFASs During Their Production, Use, and Disposal Phases

Manufacturing plants that produce PFASs or use PFASs in production of other products contribute significantly to environmental contamination by PFASs [55]. During the manufacturing process, PFASs can be emitted into the atmosphere [40,56] or discharged as contaminated wastewater that is often not sufficiently treated at wastewater treatment plants before it enters the aquatic environment [37,55]. Hazardous wastes containing PFASs can also be generated during the production process, which may not always be mitigated when incinerated or disposed of; improper storage or burial might contaminate the environment [57].

PFASs can enter the environment during the use of the PFAS-containing products [15]. For example, some carpets contain PFASs to repel stains, which might be released from carpet fibers over time [58]. Functional textiles (e.g., outdoor clothing) are also treated to have water-repelling properties. They are exposed to radiation, rain, heat, and abrasive stress that can cause the emission of repellents [59]. Many non-stick pans are coated using PFAS precursors, which can flake off during cooking or during the cleaning process. PFASs are present in more than 200 use categories [15], which have the potential to lead to unintentional PFAS release.

After their use, roughly 146 million metric tons (50%) of solid wastes were landfilled in 2018 in the US. [60]. Solid waste management facilities are viewed as important point sources of PFAS pollution, and thus, many researchers have investigated the PFAS concentration in landfill leachates [61–63]. PFASs undergo long-term leaching, and precursors degrade [64] as they are exposed to sunlight, microbial activity, varying redox conditions (available oxygen, pH-value, content of redox active species, etc.), or wastewater treatment [65]. PFAS leachate and gas condensates at solid waste landfills were evaluated across the state by the MPCA between 2005 and 2008. Down-gradient (points where water flows away from landfill) and upgradient (points where water flows to the landfill) groundwater samples were taken and found to have significant differences in concentrations of PFBA and PFOA, with PFAS concentrations being higher in the water leaving the sites. PFOS was detected in 10% of the samples. There were noteworthy differences between the median PFOA and PFBA concentrations in the groundwater at lined and unlined landfills. The median concentrations at lined landfills were 2.39 ng/L and 33.2 ng/L, respectively. The median concentrations of PFOA and PFBA at unlined landfills were 0.36 ng/L and 7.29 ng/L, respectively. The same study also showed that gas condensates were contaminated [43].

In total, the MPCA investigated the groundwater at 102 out of 111 landfills of interest. Out of those, 98% showed elevated PFAS levels, and the drinking water guidance values of the Minnesota Department of Health (MDH) were exceeded at 62 sites [66]. The landfills

included both metro and rural landfills. Rural landfills are often outside cities and adjacent to active farming operations. Most of the rural landfills analyzed in these efforts are landfills that were closed or consolidated as more stringent environmental rules increased requirements for siting and management. However, these closed landfills are likely to have been leaching PFASs for decades. They are typical of small town/county size landfills found throughout the US Midwest.

Mass balancing PFAS leachates and estimating total releases is challenging as they depend on various fluctuating factors (e.g., rainfall, number/concentration of PFASs) [67]. Lang et al. estimated that the total PFAS mass release from leachates at landfills in the US is approximately 600 kg/year [61].

3.2.2. Aqueous Film-Forming Foam (AFFF)

Over 240 individual PFAS compounds (e.g., perfluoroalkyl carboxylic acids (PFCAs), perfluorosulfonic acids (PFSAs), and perfluoroalkyl sulfonamido compounds) are used in aqueous film-forming foam (AFFF) formulations [68,69] to enhance wetting at the hydrocarbon–water and air–water interfaces [15,69]. In practical terms, this allows fire-fighting foams with AFFF to more easily encapsulate flammable liquid fuels and starve them of oxygen when attempting to stop a fuel fire. Firefighting and training activities (e.g., at airports, fire training areas, or military sites) have resulted in the contamination of surrounding surface waters and groundwater. However, data describing the lower-volume AFFF use at smaller airports has not been collected, so it is difficult to assess how significant this problem may be at rural airports. As many precursors present in these foams undergo transformation processes [70,71], the fate of AFFF-contaminated sites is not fully understood [68].

Many rural airports are located immediately adjacent to agricultural fields, which provide the open land needed for runway safety. The use of AFFF at these sites, either for training or to combat an actual fire, has the potential to contaminate nearby groundwater and fields.

3.2.3. Biosolids from Wastewater Treatment

Biosolids consist of organic matter that is recovered from sewage during wastewater treatment. They are often applied in agriculture to improve soil fertility by enriching it with nutrients and organic matter [72]. Biosolids are heterogeneous and have varying concentrations of organic matter, microorganisms, bacterial constituents, inorganic materials, and water [73]. As a result, PFASs can bind to various sites of these components with different strengths [74–76]. PFAS-contaminated biosolids are generated in the wastewater treatment process because PFASs cannot be efficiently removed from sewage being treated during biosolid separation. Some studies even showed that the concentration of stable perfluoroalkyl acids increases during treatment due to the oxidation of precursors (e.g., fluorotelomer alcohols) [77,78]. Hence, wastewater treatment plants are considered an important point source of PFAS [16]. Although sewage and treated biosolids from residential sanitation systems are known to contain somewhat higher levels of background PFASs, sewage from industrial wastewater treatment is a primary source of more highly contaminated biosolids [79].

Once the contaminated biosolids are applied on a field, the fate of the PFASs is influenced by physicochemical properties of the respective substances and soil. It was shown that the half-lives of different PFASs in soils can range from a few days to years depending on their chain length, functional groups, and their dissociation constant [80,81]. Furthermore, substances can be accumulated by various plants [82–84] or leach into the groundwater [23,85]. As a result, PFASs were detected in livestock, and thus, food pro-

duction can be an exposure pathway to consumers [86]. Lindstrom et al. investigated well water and surface water close to fields where contaminated biosolids were applied for 12 years and observed elevated PFAS concentrations (up to 11 µg/L for PFOA in surface water) [87].

The Environmental Protection Agency (EPA) estimated that 1.15 million dry metric tons of biosolids were applied to agricultural land in 2021 [88]. The yearly loading of PFASs on agricultural land in the US is estimated at 1375–2070 kg/year [89]. Thus, concerns arise about whether and to what extent biosolids pose a threat to food supply and, therefore, the economic security of farmers [90]. While a typical application of biosolids is not expected to dramatically increase PFAS concentrations in the soil, there is a concern that multiple biosolid applications or the use of heavily contaminated biosolids may increase soil contamination to the point where food or livestock feed would have high levels of PFASs.

Regulatory agencies became aware of the contamination potential of biosolids in 2016, when elevated PFAS levels were detected in milk products of a dairy farm in Maine. The Maine Department of Environmental Protection found combined PFOA and PFOS concentrations of up to 1.420 µg/L. In addition, a PFOS concentration of 32.200 µg/L was detected in dairy products on another farm in 2020 [90].

3.2.4. PFASs in Pesticide Formulations

Approximately 68,000 metric tons of pesticides are applied yearly in the Midwestern US [91]. PFASs have been part of some pesticide formulations as active ingredients or identified as inert ingredients in the past [92]. However, it is not fully clear how many currently used pesticides in the US contain PFASs as active ingredients or for other reasons. Lasee et al. found that 6 out of 10 insecticide formulations contained PFAS concentrations between 3.92 mg/kg and 19.2 mg/kg [93]. However, these results could not be verified in a separate investigation, and no PFASs were detected above the detection limit of 0.2 µg/L [94].

International pesticide active ingredient and application differences also increase the difficulty of quantifying PFAS applications to land. N-ethyl perfluorooctane sulfonamide, also called sulfuramid, is an effective insecticide used to control leaf cutting ants (Atta and Acromyrmex) [95]. It can be transformed into PFOS in the environment. Even though sulfuramid was internationally phased out by 2016 [96], it is still used in South America. Estimates are that Brazilian sulfuramid use has been responsible for the release of up to 487 metric tons of PFOS/perfluorooctanesulfonamide between 2004 and 2015 [97].

Storage containers can also be a PFAS source in pesticides via leaching. Nguyen observed gradual leaching from fluorinated high-density polyethylene containers. The total PFAS concentration was 15 µg/L when methanol was used as a solvent and 3 µg/L in water [98]. However, the relatively small contribution of PFAS from a container to any applied chemical would be difficult to detect. Consider an example of a container that holds 9 L of pesticide covering 4 hectares of crops, which could leach 15 µg PFAS per liter of pesticide, and leaching would result in 34 µg/ha or 3.3 ng/m^2. This is below the background levels, as noted by Qian and French [94]. Generally, pesticide use is very crop, weather, and geographically specific. Thus, the load of pesticide-related PFASs must be calculated on a case-by-case basis.

3.2.5. PFASs in Rural Versus Urban Communities

It is evident that rural and urban areas are impacted by different PFAS contaminant sources as they vastly differ in their industrial activities, population density, and land use [99]. This is reflected by site-specific contamination patterns, which reveal information about PFAS discharge in the area [100].

The background levels should be lower in rural areas due to reduced human activities, but similarly to urban areas, point sources are primarily responsible for pollution in rural areas. It is important to explicitly discuss sites likely to face contamination issues in agricultural and rural areas. While a significant amount of contamination has been observed in areas near PFAS production sites and in industries utilizing PFASs, such occurrences are relatively uncommon in rural areas. Very few facilities produce PFASs nation-wide, and facilities using large volumes of PFASs in industrial application tend to be closer to urban centers that can supply labor. Therefore, most contamination in rural areas occurs from the application of high-PFAS-concentration biosolids, the use of AFFF, the application of PFAS containing pesticides, or leaching from landfills. While some species of PFASs are spread by air in rural environments, it should not be significantly different than the global background levels. Lastly, in total, there are 321 airports, seaplane bases, and heliports statewide in Minnesota that are generally linked to enhanced contamination levels [101].

3.3. The Behavior and Transport of PFASs in the Agricultural Environment

The fate of PFASs in the environment and its entry into agricultural ecosystems depends on various parameters. The physicochemical properties of particular PFAS species determine their movement, partitioning, and transformation behavior, also called environmental fate [102]. The PFAS chain length [103] and functional groups [104] significantly impact their solubility, mobility, and bioavailability. Thus, each substance has a unique environmental fate, which makes it challenging to develop predictive models in agricultural soils, plants, and livestock [105].

3.3.1. PFASs in the Atmosphere

Non-ionic PFASs (typically precursors) are often sufficiently volatile such that they are transported in the atmosphere [106]. Long-range transport of PFASs, on the order of days to weeks, can occur in the atmosphere. For example, researchers attributed the contamination of the European Arctic to long-atmospheric transport processes, the resulting degradation processes and local sources (e.g., consumer products and AFFFs) [107,108]. The total historical global emissions of PFCAs due to indirect air emissions are estimated to be as high as 350 metric tons, which contributes between 4.8% and 10.9% of the total historical PFCA emissions [109].

Airborne PFASs can migrate to the soil or into surface water via dry (direct deposition) or wet deposition (ab- or adsorption by/at water) [107,110–112]. Compared with other types of movement of PFASs, airborne movement likely spreads material over a wider area at a lower concentration. This compares to PFAS movement in water, biosolids, or other solid/liquid mediums. This is likely a primary mechanism for the background PFASs found in many isolated rural communities, both agricultural and non-agricultural.

3.3.2. PFASs in Surface Water, Sediment, and Groundwater

About 40 different PFASs have been detected in the aquatic environment [106]. PFASs with low pKa values and vapor pressure (e.g., PFCAs and PFSAs) are ionic at common environmental pH values. These ionic PFASs are mainly observed in water or bound to particles and sediment [109,113]. The PFASs that persist in the aquatic environment can be transported downstream by rivers until they end up in the marine environment, where PFASs can reenter the atmosphere via sea spray [114]. During this process, the total PFAS concentration decreases downstream until it reaches the oceans [106].

Multiple studies have investigated the sorption behavior of PFASs between water and different sediments/soils [104,113,115,116]. Sediments can store organic pollutants over a long period since sediments show hydrophobic behavior in the aquatic atmosphere [117], and trends of released PFASs (e.g., replacing PFOA with HFPO-DA) can be observed in

sediment cores [118]. Other important parameters for sorption are the pH value, organic carbon content, and the coexistence of certain metal cations (e.g., Ca^{2+}) [104,113,115,116].

Generally, PFCAs with a chain length shorter than seven carbon atoms were exclusively found in the liquid phase [104], and every extra CF2 moiety on the molecule in-creases binding affinity to the solid phase. Moreover, the sulfonate group enhances the absorption strength further in comparison to the PFCA analogs [116]. Linear PFAS isomers tend to sorb to sediments due to their lower hydrophilicity compared to their branched isomers [119].

Many worldwide studies have investigated groundwater contamination levels [120–124]. Groundwater contamination can be attenuated by the vadose zone due to sorption phenomena [85], and surface water was identified as a key source of groundwater contamination [125–127]. In contrast to the atmosphere and deposition on surface water, which retain PFASs for short periods, groundwater (and soil column) contamination reflects the long-term retention of PFASs [128].

The biotransformation of precursors is another important factor in evaluating whether aquifer contamination is a potential source of danger for the environment [121] or human health [9,129]. Biotransformations are highly dependent on the toxic conditions of the soil [70,130,131].

As groundwater enters springs, lakes, rivers, and wetlands or is used for irrigation [132], PFASs that are present can reenter the water cycle [121] and thus might be reintegrated into the biosphere. Overall, PFAS groundwater contamination processes are not well understood, and further research is needed to improve modeling [133].

3.3.3. Behavior and Uptake of PFASs in Soils

PFAS Sorption

The modeling of soil PFAS adsorption is complicated by the wide variation in soil components and the resulting soil's physical and chemical properties. Soils are a mix of mineral, organic, and biologically active materials and other compounds that can change over the distance of a few meters. These variations interact in concert with the chemical property differences in PFAS species. Some soils simultaneously have surface sites that are charged positively and negatively. The surface charge strongly depends on the pH and on the soil type [134,135]. At the same time, several PFASs occur in an ionic or zwitterionic state (equal number of positively and negatively charged functional groups present in the molecule) in the environment [136,137].

Several studies investigated the vertical distribution of PFASs in the ground after contamination due to biosolids application or AFFF use [6,21,138–140]. In most cases, the observation was that the vertical loading of PFASs in the vadose zone decreased sharply with increasing depths. In contrast, Nickerson et al. found that total PFAS concentrations increased along the vertical profile of the soil, and the maximum concentrations were observed at a depth of 3–5 m [141]. The retention and lifetime of PFASs, especially precursors, differ vastly depending on the soil [142–144].

The factors influencing the adsorption of PFASs (mainly PFOA and PFOS) onto soils have been thoroughly investigated. It is apparent that the sorption PFASs is not dependent on one factor and that models must use multiple parameters to describe sorption accurately. Organic carbon is often attributed as the driving parameter of PFAS adsorption in the soil [116,145–149].

Another important factor is the content of dissolved inorganic salts in the soil column since they can change the properties of the aqueous phase. Investigations showed a positive correlation between the ionic strength and the sorption of anionic PFAS onto soils [116,146,147,150,151]. It was found that with increasing ion concentrations, the mo-

bility of PFASs along the soil profile decreased (especially for long-chain PFASs with C >10) [151].

Most sorption processes of PFASs might be explainable when considering the organic carbon, pH, and the clay content due to the importance of electrostatic interaction for the sorption processes. More data is needed to conduct multivariate data analysis techniques, which might provide a more accurate understanding [76]. This would be very helpful as values of the field- and laboratory-derived partitioning coefficients for PFOS, PFOA, PFNA, and PFDA in the literature vary by factors of 1.6, 10.9, 1.9, and 1.7, respectively. All field-derived values are larger than the laboratory-derived values [76]. More soil adsorption research is needed, which should include a precise characterization of the soil properties to facilitate future analysis of sorption processes. If the sorption mechanisms are fully understood, farms and areas that are likely to have the potential to have a high concentration of sorbed PFASs can be identified. This ensures effective employment of resources in mitigation efforts.

PFAS Retention

The retention of PFASs in soils depends not only on the interactions between the compound and the soil but also on (1) adsorption at the air–water interfaces, (2) partitioning to soil gases, (3) adsorption at non-aqueous phase liquid (NAPL)–water interfaces, and (4) partitioning to the NAPL [152,153]. The combination of general partitioning mechanisms might not be sufficient to predict retention in some cases [154]. One field study investigated the PFAS partitioning behavior at AFFF-affected sites and observed that at 87% of the examined sites, the concentration levels in the soil exceeded those of the groundwater, indicating the significance of the retention [154].

However, a laboratory study revealed that between 80% and 90% of the ten investigated PFASs were eluted in the flow-through soil column experiments [155]. It is questionable to what extent the results of the study can be transferred to environmental conditions. A further important contributor to PFAS transport is the presence of co-contaminations [156]. Those can increase sorption by providing additional binding sites or lower PFAS sorption by competing for existing sorption sites in the soil [157].

PFASs in Agricultural and Rural Soils

PFAS release mechanisms of concern for agriculture are all point sources of contamination that first impact specific release sites. Thus, preventing agricultural plant and animal systems from being exposed to PFASs requires an understanding of local point sources of contamination and the potential for movement into the agricultural environment.

Depending on factors such as surface and sub-surface water flows and soil types, the areas adjacent to the initial contamination sites can become polluted over time. As described above, the rate of movement is dependent on many different soil, water, and PFAS physical and chemical properties as well as concentration gradients. Based on the data from Minnesota contamination sites, it is likely that sub-surface movement of PFASs from a contamination site can take several years per kilometer of movement in typical soils. This time is reduced in soils with high groundwater flows. In contrast, contamination moves very quickly in surface waters.

The risk of PFAS translocation should be insignificant for the consumer if the products were grown on fields with no history of direct PFAS application and that underwent minimal industrial processing. However, the general lack of monitoring of agricultural sites makes it challenging to perform accurate risk assessments.

3.4. PFASs in Plant and Animal Systems

Agricultural plants and animals are in direct, constant interaction with the outdoor environment, and thus, they can take up PFASs from their surroundings. A better understanding of the uptake mechanism of agricultural plants and animals consuming plants is required to reduce the potential risks of human exposure to PFAS from agricultural products. This section describes the current understanding of these interactions and the potential for contamination of agricultural goods during production.

3.4.1. Plant Uptake of PFASs

PFAS Uptake Mechanisms in Plants

The predominant pathway for plant accumulation is root uptake [158–161]. Aboveground parts of the plant can also absorb airborne PFASs to a smaller degree [162–164]. In the first step of the root uptake process, the contaminants diffuse passively into the surface tissue of the root system. The PFASs are then gradually distributed and accumulate in the root tissue. This only occurs in cell walls that have not sufficiently developed protective layers (e.g., cuticle layer) [165]. If the molecule has sufficient lipid solubility, it can pass the lipid bilayer of the membrane and enter the aqueous phase inside the cell. Charged or polar particles might be hindered from entering the cell membrane by the lipid bilayer, but proteins can function as a transport system [166] due to the high affinity between PFASs and proteins [167]. After they enter the root tissue, PFASs can be absorbed by apoplastic cell wall tissue outside root cell membranes and be transferred through the xylem up to the plant's shoot (see Figure 3). Here, the Casparian strip inhibits the translocation of long-chain PFCAs, PFSAs, FOSAs, and chlorinated polyfluorinated ether sulfonates by sealing the space between the cell membranes and cells [83]. As a result, long-chain PFASs are mainly limited to the root system and accumulate in the shoot system in a smaller amount [84,159,168].

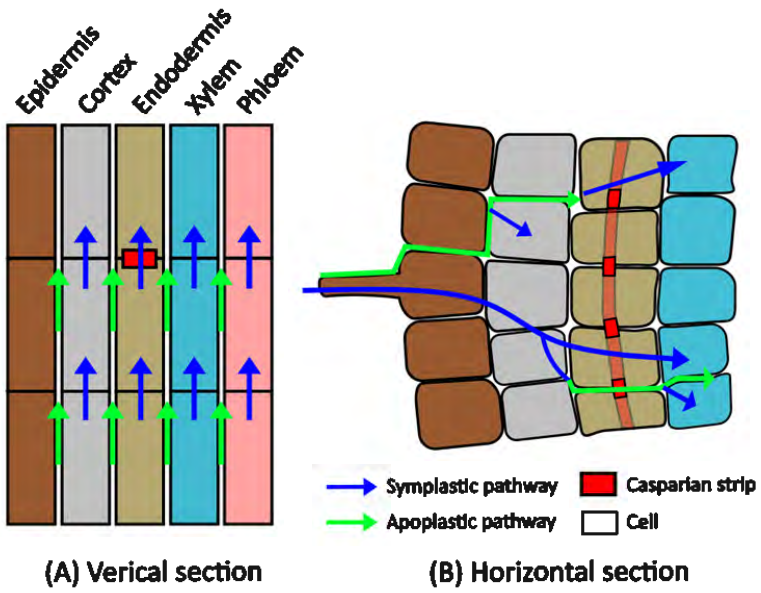


Figure 3. A schematic illustration of the possible pathways of PFAS translocation in plants. A vertical root section and transportation (**A**); PFAS movement in a root cross section (**B**). The movement along cell walls (apoplastic) is indicated with green arrows, and the translocation through cells (sym-plastic) is displayed with blue arrows [169].

PFAS concentrations in fruits decrease further due to additional barriers as the contaminants are translocated in the aboveground plant tissue [170]. It is important to note that uptake mechanisms are plant specific [83]. For example, Liu et al. linked differences

in PFAS uptake in different crops to the varying lipid and protein contents [56]. Another study showed that the shoot accumulation of long-chain PFASs might be related to the surface-area-to-tissue ratio and is enhanced with an increasing surface area [171].

Uptake Rates of Different Crops

Since most plant uptake experiments have utilized varying conditions (e.g., soil vs. hydroponic cultivation, different plants or soil types, contamination levels, etc.), uptake metrics have been developed to improve the comparability of plant uptake across different studies. The bioaccumulation factor (BAF) gives the ratio of the PFAS concentration in the plant to the concentration levels in the surrounding environment (Equation (2)) [172]. The second important factor is the translocation factor (TF), which describes the distribution of the compound of interest within the plant (e.g., TFleaf/root) and is calculated similarly to the BAF (Equation (3)) [173].

$$BAF = [PFAS_{plant\ tissue}]/[PFAS_{soil}] \quad (2)$$

$$TF = [PFAS_{leaf}]/[PFAS_{root}] \quad (3)$$

The literature shows a direct positive correlation between the PFAS concentration in the soil and the plant [83,84,174]. Moreover, the bioaccumulation of PFASs by plants depends on various plant and soil factors. The physicochemical properties of the respective PFASs, contamination levels, plant type, and environmental conditions all interact to determine the uptake rate. An interesting trend observed was that, while TFs varied throughout the plant, BAFs were higher for vegetative plant parts than reproductive and storage organs [84,173]. The Casparian strip plays an important role in impeding the translocation within the plant, and translocation is also decreased by a relatively low protein content in the aboveground tissue [169].

Krippner et al. investigated the influence of the chain length (C4 to C10 for PFCA; C4, C6, and C8 for PFSAs) on the uptake and distribution of PFCAs and PFSAs in corn and observed that the uptake rates (BAF) exhibited a U-shaped trend. The lowest uptake rates were observed for PFHpA and PFHxS [175]. In contrast, the TF showed a negative correlation with the chain length for the tested PFCAs and PFSAs. Longer-chain-length PFASs possess a higher affinity for the lipid bilayer of membranes, which reduces overall movement within the plant [175]. PFOS showed the highest mean uptake rates, possibly due to different uptake mechanisms [176]. However, the BAFs for PFOS in different plants are generally considerably lower than those for PFOA or PFHxS [177]. It is apparent that translocation is more hindered with increasing chain lengths, and the risk of exposure is higher for short-chain PFASs that are less studied [178]. It should be noted that Krippner et al. grew plants in nutrient solutions, which is a less realistic approach since the interactions with soil components are missing [175].

In a case study, Liu et al. [56] examined the PFAS concentration in the soil and crops from a mega fluorochemical industrial park at two distances (0.3 km and 10 km). The soil had total contamination levels between 79.9 µg/kg and 200 µg/kg, with PFOA being the most abundant substance. The PFAS levels in the crops were between 58.8 µg/kg and 8050 µg/kg, with the BAFs of edible parts reaching levels of up to 48.0 µg/kg (radish) in shoot vegetables. The ranking for BAFs in terms of highest bioaccumulation of the different plants grown at the field 0.3 km from the pollution source were shoot vegetables (24.3 µg/kg) > fruit vegetables (6.63 µg/kg) > flower vegetables (4.23 µg/kg) > grain crops (4.05 µg/kg) > root vegetables (3.58 µg/kg).

PFASs' impacts on plant health were examined by Stahl et al., who studied the uptake of PFOA and PFOS by different plants by spiking the soil with concentrations between 0

and 50 mg/kg. They found that exposure to a concentration of 50 mg/kg led to a significant reduction in corn (*Zea mays*) yield, mainly due to reduced ear mass [160]. However, these high contamination levels are only likely found at sites where PFASs are directly released (e.g., manufacturing or AFFF affected sites) [179], and studies investigating PFAS uptake at more typical environmentally relevant concentration levels are lacking [180].

3.4.2. PFAS Contamination in Livestock

The primary mechanism for PFAS contamination of livestock, and the resulting animal-based productions they provide, is the consumption of contaminated feed or water. PFASs can be absorbed by the gastrointestinal tract [181,182] and bind to blood serum proteins [183,184]. The distribution in animal tissue is PFAS and species dependent. For example, PFOA, PFOS, and PFBS accumulate to a higher degree in the liver in most species, whereas PFBA and PFHxS can be found to a higher degree in the blood serum [182].

Kowalczyk et al. [25] investigated the kinetics of PFBS, PFHxS, PFOS, and PFOA in dairy cows by feeding them contaminated grass silage and hay for 21 days. The plasma concentration levels of PFBS (mean = 1.8 ±0.8 μg/L) and PFOA (mean = 8.5 ± 5.7 μg/L) were relatively low compared to those of PFOS (2462 ± 411 μg/L on day 44) and PFHxS (419 ± 172 μg/L on day 29). Additionally, the concentrations in the milk samples were proportional to blood serum levels, and the highest cumulative secretion was observed for PFOS (14 ± 3.6% μg/L). PFBS (0.01 ± 0.02% μg/L) and PFOA (0.1 ± 0.06% μg/L) were secreted into the milk in a low amount. Similarly, it was shown that PFOA is excreted significantly faster compared to PFOS in sheep. PFOA was mainly excreted with urine (51–55%) and could not be detected after day 42. In contrast, the highest PFOS excretion was detected in feces (4–5%) followed by milk. The PFOS levels in the tissue of the sheep did not decrease during the 21-day PFAS-free feeding period [26]. Since the animals were exposed to contaminated fodder for a relatively short time, serum and/or tissue concentrations might have not reached a steady state, possibly leading to a significant underestimation of these values [185].

In general, few studies have focused on PFASs in livestock, and their focus was on uptake and elimination. Potential adverse health effects were hardly investigated for different animal species, especially livestock [86]. Studies in monkeys showed a link between the exposure of PFASs and liver toxicity, altered thyroid hormone concentrations, decreased cholesterol serum concentrations, weight loss, and glycogen metabolism [186]. It is assumed that animals are capable of tolerating high PFAS concentrations [86], but more research is needed to investigate potential adverse health effects in livestock. Hlousova et al. found that PFAS concentrations from common farm livestock decrease in the following order: pig/bovine liver > egg > meat > dairy products (butter) [187]. Pasecnaja et al. summarized several studies that assessed the contamination levels in the European food market. They found that the most important sources were fish, meat, eggs, fruits, and vegetables, with fruits and vegetables exceeding the levels found in meat [188].

3.4.3. PFAS Transfer Through the Food Chain

Several parameters influence PFASs as they moves through the food production chain (Figure 4). The descriptions of the environmental fate of PFASs described above can be transferred to food production. Bioaccumulation can occur at every step if a contamination source is present. This is critical if the source continuously emits PFASs or if large amounts are released over a brief period. Crops grown on soil with high PFAS loadings will uptake the surrounding contaminants to an often times unknown extent. If these crops are used as feed, the livestock and the produced meat will be affected as well and might show stronger contamination levels compared to the fed crop. For example, Tefera et al. estimated the

total daily mean dietary PFAS intakes for firefighters. They showed that food (i.e., eggs, fruits, and vegetables) which was produced at fire stations was responsible for 82% (i.e., 1250 ng/d) of the total PFAS intake [189].

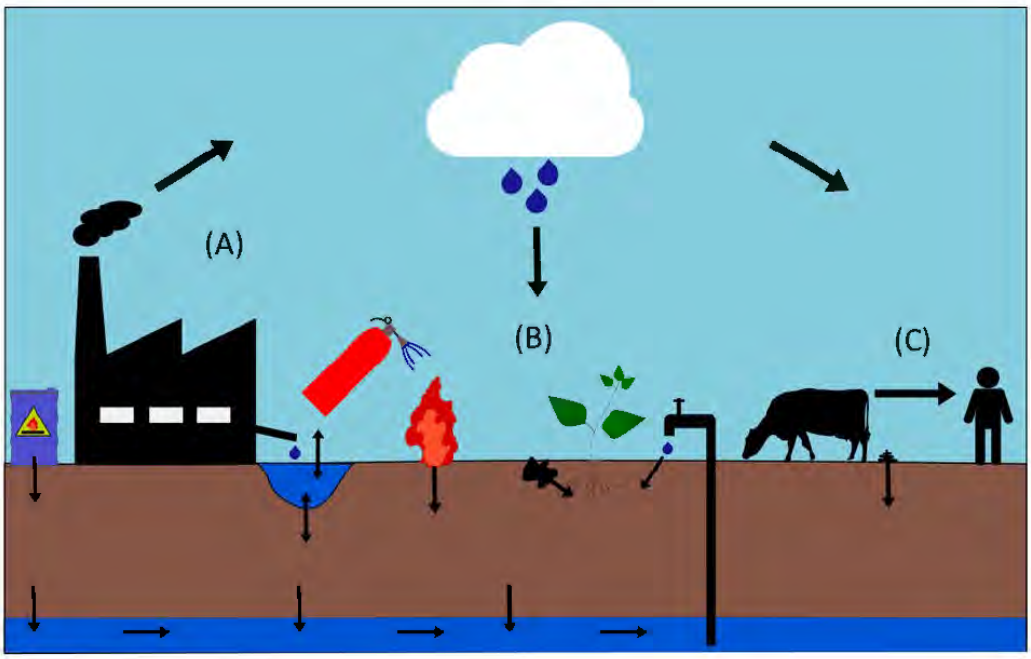


Figure 4. An illustration of the PFAS cycle in relation to agriculture. (**A**) shows the release of PFASs by point sources (manufacturing sites, landfills, AFFFs, and biosolids), which is followed by (**B**) uptake by crops and livestock. Lastly, (**C**) contaminated products are consumed. The arrows indicate the direction of PFAS translocation and accumulation along the food production process.

The large number of individual PFAS molecules and their unique fates make it challenging to conduct highly accurate PFAS risk assessments [23] for food production systems. While short-chain PFASs have not been the focus of research, they show the highest mobility in the environment and across the food web, and their emissions are expected to increase. Recent studies showed that they might cause adverse health effects similar to long-chain PFASs [178,190,191], indicating the dire need for intensive investigations on shorter-chain PFASs.

There seems to be a disconnection between the fact that diet is a major exposure pathway [23] and the limited resources being used to address PFASs in agriculture. This might be partially due to early studies suggesting that dietary exposure is neglectable [192]. However, recent studies suggest that even exposure to an extremely low concentration might cause detrimental health effects, which is underlined by the EPA's updated drinking water health guidance values [193] (see Figure 5).

The complexity of soil-to-plant, plant-to-human/livestock, and milk/meat-to-human PFAS interactions combined with the vast number of PFAS species limits our understanding of how and which PFASs are transferred through the food web. Another dimension of complexity is added when considering PFASs present in food contact materials (e.g., fast food wrappers or baking plates) [194]. This makes accurate assessments of the posed threat to consumers nearly impossible without more information.

3.5. Policy and Regulations

In the US, regulation and policy for pollution issues often occur at both the federal and state levels. There are currently no nationwide restrictions on PFAS contamination levels in food in the US. The State of Maine Department of Agriculture, Conservation and Forestry established PFOS action levels for beef (3.4 µg/kg) and milk (0.21 µg/L) [195].

The European Food Safety Authority set a tolerable weekly intake (TWI) for the sum of PFOA, PFNA, PFHxS, and PFOS at 4.4 (ng/kg food)/(week · kg bw) [196].

Most PFAS regulation and research funding have been directed at drinking water standards. Due to Minnesota's longer history of PFAS pollution issues, Minnesota has had much earlier PFAS drinking water standards than the federal government. In the past few years, the federal government has begun to set more stringent requirements for drinking water (Figure 5).

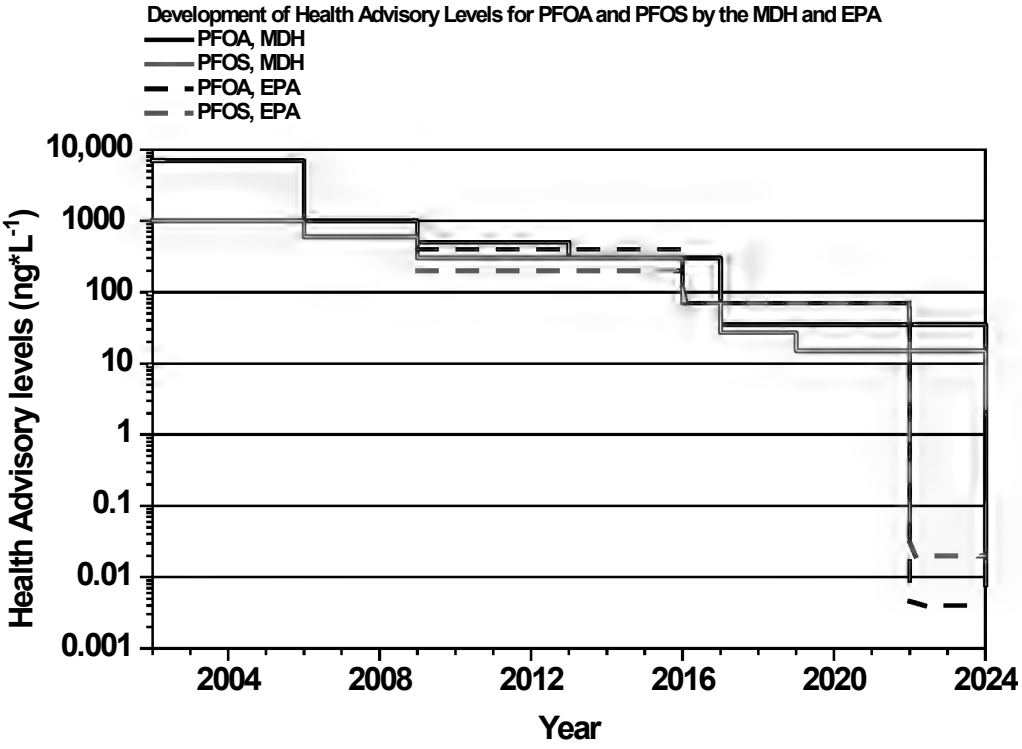


Figure 5. A comparison of the proposed health advisory levels for PFOA (black) and PFOS (gray) drinking water standards by the Minnesota Department of Health (MDH) (continuous) and US Environmental Protection Agency (EPA) (dotted) between 2002 and 2024 [196,197]. Note that the scale is a log scale, so proposed standards for 2024 are on the order of 100,000 times more stringent than in 2002.

Until recently, neither the state nor federal government had addressed PFAS contamination issues in agriculture. In 2023, the Minnesota Legislature passed bills (SF 1955 and HF 2310) regulating pesticide products that contain intentionally added PFASs [197]. One of the new laws requires pesticide registrants to inform the Minnesota Department of Agriculture if a pesticide product contains intentionally added PFASs. The EPA is studying this issue and determining the scope and scale of pesticide-related PFAS issues.

Both Minnesota and the federal government have set up near-term plans for further research and rulemaking on PFAS-related issues. Minnesota's PFAS staff group has observed that it is challenging to manage PFASs sufficiently through regulatory actions as several vital areas (e.g., pollution prevention and waste management or understanding risks and how they relate to exposure through food/water) are overlapping [198]. The EPA's roadmap's goals for 2021 to 2024 are to increase the understanding of the effects of PFAS exposure on human health and ecological systems, prevent the release of those sub-stances into the environment, and facilitate remediation efforts at contaminated sites [199].

In Minnesota, the MPCA, MDH, Department of Natural Resources, and Minnesota Department of Agriculture published Minnesota's PFAS Blueprint for addressing the PFAS problem systematically and efficiently at the beginning of 2021.

Minnesota's blueprint lists the following areas of concern: (1) preventing PFAS pollution, (2) measuring PFAS effectively and consistently, (3) quantifying PFAS risks to human health, (4) limiting PFAS exposure from drinking water, (5) reducing PFAS exposure from consuming fish and game, (6) limiting PFAS exposure from food, (7) understanding risks from PFAS in the air, (8) protecting ecosystem health, and (9) remediating PFAS-contaminated sites [198].

While all agree that the potential for PFAS contamination in food production needs to be addressed [83], regulating agriculture is often made more difficult due to political and practical issues. Minnesota's Department of Employment and Economic Development estimated that the agricultural sector in Minnesota generated about USD 17 billion in sales, with USD 8.85 billion being contributed from the cultivation of crops in the year 2022 [200]. Similar economic impacts are generated by agriculture in states throughout the US Midwest. Agriculture is an important industry that supplies the planet with needed food; thus, state and federal regulators are often cautious about changing rules for agriculture. However, care needs to be taken to reduce the potential that agricultural products are a source of exposure to PFAS in people. Poorly addressing PFAS issues has the potential to negatively impact human health, threaten economic sustainability, and cause consumer backlash.

The PFAS cycle must be broken long term to prevent further contamination on agricultural sites. This requires far-reaching regulations governing the production, use, and disposal of PFASs as it is very challenging to prevent the emission of PFASs into the environment over their life cycle [201]. Therefore, restricting production and utilization is the best option to prevent further contamination. For example, the state of Minnesota banned the use of PFAS-containing firefighting foam with a few exceptions and limited intentionally adding PFASs to food packaging material in 2024 [202]. Banning the commercialization of PFAS-containing products is especially important if the PFAS-provided properties are not necessarily required for the respective application. This step must also include thorough and mandatory testing of PFAS substitutes to ensure their biocompatibility and biodegradability. Biosolids for land application should be thoroughly investigated, and PFAS contaminants exceeding a set threshold should be treated as they are primary contamination sources in rural areas. The Maine legislature went so far as to ban land application in August 2022 [203]. Existing wastes should be handled adequately so they will not enter the groundwater or atmosphere. Furthermore, law makers should address PFAS-containing food contact material and restrict their use if necessary. Consequentially, legally binding maximum PFAS levels in food should be established, which should include the most common legacy PFASs (e.g., PFBA, PFBS, PFHxA, PFxS, PFOA, PFOS, and fluorotelomer sulfonates) and upcoming alternatives (e.g., GenX, F-53B, OBS).

Lastly, agencies should facilitate mitigation strategies, which are still energy- and cost-intensive. It is self-explanatory that the required actions require enormous resources and the parties responsible for the pollution to be held accountable.

3.6. Future Research Recommendations and Challenges

It is clear that the US agriculture community faces challenges in identifying the risks that PFASs present to today's agriculture-based food systems. More work across all areas is needed to understand the scope of these challenges and develop long-term strategies for mitigating related risks. It is also important to recognize that there is a difficult question of responsibility for PFAS contamination as farms are most likely contaminated without the knowledge of the farmers. However, they may suffer financial losses due to contamination or mitigation. A rapid response and sufficient resources are needed for establishing a framework to tackle the PFAS crisis.

3.6.1. Agricultural Challenges and Strategies for PFAS Mitigation

Arguably, most US farmers have not heard of the term PFAS. Thus, a knowledge gap in the farming community is an initial barrier to mitigating risks to agriculture and the food supply. Little guidance and studies in the literature are available that are geared towards informing agricultural practitioners about PFASs. The agriculture sector is already under heavy pressure to act on other environmental issues (e.g., greenhouse gases, nitrates, fossil fuels, and pesticides), which makes it difficult to incorporate additional information about PFASs in outreach materials to producers and their cropping advisors. Without this information, farmers are unlikely to even be aware that they should consider potential risks to their crop or livestock products from PFASs.

It is also clear that the outlined knowledge gaps need to be addressed regarding translocation in agricultural plants and livestock. Therefore, more research on PFAS translocation and presence is needed in every step of food production (soil sorption, plant uptake/translocation, transfer to livestock, and consumption). In many products, long-chain PFASs have been replaced with shorter alternatives (e.g., GenX), which are more mobile in agricultural systems. However, there are uncertainties about their toxicological potential that must be studied. Field studies that deal with plant uptake are needed to enhance the understanding of the respective uptake mechanisms. Clinical studies that expose the livestock to relatively high PFAS loads have limited applicability as well, which makes it challenging for regulators to establish suitable action plans. One helpful measure would be to develop TWI values to increase the transparency and trust of consumers.

These suggested efforts require immediate action, many resources, and the bundling of expertise across biological, chemical, and medical fields as the question of under which conditions crops and livestock can be safely grown must be answered.

Finding data on PFAS-contaminated agricultural sites that present risks to farm operations is also difficult. States with robust environmental protection policies or a known existing issue with PFASs may have a well-developed database on PFAS manufacture, use, or contamination sites. For example, Minnesota has a repository of PFAS-related test data available online in raw and searchable (map based) formats [50]. A further challenge for producers that have identified a potential PFAS contamination risk is the lack of resources on subsequent steps to verify the risks. Educational and consulting resources for farmers in the US are typically centered on state universities and their federally funded agricultural extension staff. Extension staff in some states have begun outreach efforts on agricultural PFAS issues [204], typically in states with known significant PFAS problems. However, the majority of state universities do not have staff focused on the issue. Farmers at high risk for PFAS contamination will likely need to refer to information from other states or outside the agricultural sector.

Should a producer believe that they may be at risk for PFAS issues in their operations, the next step would be testing. The availability of testing labs and methods needed for performing soil/plant/animal testing will be another challenge for farmers. Tests for PFASs are very sensitive, detecting these materials at the parts per billion or trillion level. A simple web-based search indicates that most certified labs testing drinking water samples in the US charge between USD 250 and USD 600 per sample. It is likely that testing soil or products would add additional costs. Between the costs of tests and sampling work, it is likely that farmers would find testing unaffordable. Moreover, financial support should be provided for contaminated farms so that farmers do not fear testing. It may be necessary for states to assist with the expertise and cost of conducting PFAS tests. There is also the issue of interpreting test results as there are currently no guidelines for defining soil contamination in an agricultural context.

3.6.2. Strategies for PFAS Risk Reduction on Farms

Understanding the sources and associated entry of PFASs into the environment is crucial to identify locations with higher associated risks and to develop mitigation strategies to ensure efficient use of resources [205]. Based on the findings in Minnesota, these include rural sites located by landfills or airports or sites that have undergone application of contaminated biosolids. In the case of landfills and airports, these are typically matters of public record. Farmers who have been applying biosolids will typically know how much and how often they have applied them to their land. This work can all be performed at the individual farm level or by mapping farms adjacent to known contamination sites.

Another approach to identifying risks of PFASs in agriculture that may play an increasing role in the future is machine learning (e.g., for farms in proximity to airports or military sites). Maine is already examining machine learning-related methods [195]. However, extensive PFAS and environmental monitoring data is required for these methods to be efficiently applied.

Once identified, sites with elevated contamination levels or at high risk for new contamination must be further evaluated to assess whether and how farming can be continued. Remediating agricultural sites is currently not economically feasible due to the energy and cost-intensive nature of the respective methods. Alternative crops that have lower BAFs or are not used for food production (e.g., bioenergy or fiber crops) may need to be produced on contaminated land. PFASs would still be present on both land and crops but would not put livestock or humans at risk.

Another contamination-related issue is irrigation and groundwater. Agricultural lands may not necessarily be heavily contaminated, but irrigation water pumped from contaminated aquifers could pose a risk. Again, the primary rural source of contamination for water is landfills; thus, it is prudent to test irrigation water being pumped from wells near landfills. This should also be performed for drinking water provided to livestock.

At present, these are concrete steps that can be taken to mitigate PFAS risks. They will likely need to be re-evaluated and updated as more information becomes available on both PFASs in general and its linkage to agriculture in particular. The rapid development of strict drinking water standards suggests that PFAS regulation will be developed for agriculture in the near future so that we can reduce the potential for PFAS contamination in food supply.

4. Conclusions

The experience with PFASs in Minnesota makes it evident that PFAS contamination in agriculture is an area of concern that needs to be more fully examined by the scientific community, policymakers, and farmers to mitigate risks for the environment and consumers. Minnesota, with its historical relationship to PFASs and a strong agricultural sector, provides a glimpse into the major sources of agricultural contamination (landfills, biosolids, and airports) and can be used as a model region for other rural areas. Adaptations must be made based on local geographical features, its contamination history, and individual food production chains.

Knowledge gaps are a critical issue, as consuming PFAS-contaminated foodstuffs is identified as an important exposure pathway. Our findings suggest that more field studies which investigate the PFAS partitioning behavior in all soil types, PFAS uptake by crops cultivated in different soil types, and PFAS uptake/distribution/accumulation in livestock are direly needed. Studies in a controlled environment can expand the understanding of certain factors but can only be applied to real conditions to a limited extent. Often times, environmental conditions are not investigated, but unrealistic high PFAS levels are used. Epidemiological studies are lacking in this area, as well as data and reports specifically

evaluating contamination levels at agricultural sites. The amount and type of specific PFASs detected in the environment will change in the near future due to manufacturing adaptions made by the industry. This poses a great challenge to the scientific community and must be tackled quickly so that research, regulations, and public health measures do not only address legacy PFASs.

The high complexity, knowledge gaps in critical areas, and interdisciplinary nature of the PFAS crisis do not allow reliant risk assessments at the present time. No simple approaches will adequately meet this problem, and complex approaches, such as machine learning or intergovernmental cooperation, will be of utmost importance.

Our findings suggest that the consumption of produce from areas with no direct PFAS application should pose no significant threat to the consumer. Animal products, especially offal, generally show higher contamination levels compared to plant-based foods. It is advisable to establish a monitoring system in agricultural areas and across the food chain. Farms and foods with a higher associated risk of contamination (e.g., close to manufacturing sites, airports, or landfills; those that have undergone historic biosolid application; or those in contact with contaminated surface water) should be preferred during the planning phase of such systems. Legally binding regulations dealing with PFASs in agriculture and food production are basically non-existent, which shows the need for immediate action.

The current strategies to limit agricultural PFAS contamination rely on avoiding known contamination from these sources or leachates spreading in groundwater. Remediation efforts will likely play a significant role in the future for some agricultural sites as the methods become economically competitive or are required by future regulations. However, there are currently no sensible remediation strategies in the context of agriculture. Established methods, such as the excavation or washing of contaminated soils, do not meet the requirements, and more attention needs to be paid to this aspect.

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JLTURE

h a second farm shuttered due to massive S contamination, Maine legislators weigh ing access to the courts

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Just days before the Maine Legislature's Judiciary Committee was scheduled to hold a virtual hearing on LD 2160 (https://mainelegislature.org/legis/bills/display_ps.asp?paper=HP1544&snum=129) — legislation to clarify when civil lawsuits may be filed for compensation for harm from Per- and Polyfluoroalkyl Substances (PFAS) contamination — a second dairy and beef farm was found by Maine's Department of Agriculture, Conservation, and Forestry to have "very startling" (https://pfascentral.org/news/state-investigating-very-startling-levels-of-pfas-chemicals-on-central-maine-dairy-farm)levels of PFOS, one of the PFAS family of chemicals.

In fact, the amount of PFOS in milk from the dairy herd at the Tozier farm in Somerset County was worse than "startling" — it may be the highest milk contamination levels ever recorded (https://www.ourhealthyfuture.org/sites/default/files/pdfs/fs_-_highest_milk_levels_-_updated_with_2020_farm_1.pdf) in North America. Measurements in late June and early July ranged from 12,700 to 32,200 parts per trillion (ppt). The highest reading is 153 times Maine's standard for determining that milk is "adulterated" (https://www.maine.gov/dacf/ag/pfas/docs/dacf-ats-pfaspresentation-011420.pdf) and unfit for sale (210 ng/l). As a result, the farm has been forced to stop selling its milk and beef.

These measurements are twenty times the level of PFAS that were discovered in 2016 in milk from the dairy herd at farmer Fred Stone's 100-year old Stoneridge Farm in Arundel, Maine. As the bill's sponsor State Rep. Henry Ingwersen explained (https://mainelegislature.org/legis/bills/getTestimonyDoc.asp?id=141460), Stone's problems began in November of 2016, when he received a letter from the local water district saying that tests showed a well providing his and his cows' drinking water was contaminated with over twice the Environmental Protection Agency's (EPA) advisory limit of 70 ppt for PFOS. The Maine Department of Environmental Protection later concluded that the source of the contamination was wastewater sludge spread on the farm as fertilizer under a state-sponsored program from 1983 to 2004. In 2016, Fred began voluntary testing of his water, soil, hay used for feed, cows, milk and both his and his wife

Laura's blood — and found it all to be contaminated with PFAS. There are currently soil locations on Fred's farm that still test as high as 800,000 ppt. Stoneridge Farm has been forced to shut down (https://www.reuters.com/article/us-usa-dairy-chemicals-idUSKCN1R01AJ) due to the contamination.

Despite the fact that all municipal sludge tested in the state has contained PFAS (https://theintercept.com/2019/06/07/pfas-chemicals-maine-sludge/), Maine still allows sewage treatment sludges (euphemistically called "biosolids" (https://www.theguardian.com/environment/2019/oct/05/biosolids-toxic-chemicals-pollution) by wastewater treatment and composting operations) to be used on farmland. Both Maine farms now shuttered because of PFAS contamination used sludge as fertilizer. This practice is common throughout the country and has the approval of the EPA as well as state environmental agencies.

In the absence of federal action (https://www.npr.org/2019/07/16/742109303/as-federal-regulations-lag-states-take-action-against-pfas-chemicals), state governments are struggling to address widespread PFAS contamination of water, soils and food. PFAS are a group of man-made chemicals that include PFOA, PFOS, GenX and many other chemicals (up to 4,000 variations). Sources of PFAS contamination are many because these chemicals are ubiquitous. They are found in foam fire suppressants and in various industrial wastes, including from paper, textile and tannery operations. They are used in the manufacture of rain and stain-repellent clothing treatments such as Scotchgard and Gore-Tex, car and floor waxes and even some dental floss. PFAS is also showing up in food and compost through PFAS-containing food packaging and other contamination sources, and high levels have been found in breast milk (https://theintercept.com/2019/04/30/breast-milk-pfas-chemicals/) around the world.

PFAS are persistent in the environment, meaning they don't break down for years and can bioaccumulate in both humans and farm animals. For this reason, they have been called "forever chemicals (https://www.foodandwaterwatch.org/insight/these-chemicals-are-forever-water-contamination-pfoa-pfos-and-other-pfas)." According to the federal Agency for Toxic Substances and Disease Registry (https://www.atsdr.cdc.gov/pfas/health-effects.html), exposure to certain PFAS may affect growth, learning and behavior of infants and older children, and cause endocrine disruption. Exposure to PFAS has been linked to kidney cancer and testicular cancer, as well as thyroid disease, compromised immune systems and infertility.

Maine's farmers are on the front lines of this PFAS-caused disaster. Their health is at risk from contaminated drinking water causing high PFAS blood levels (https://pfasproject.com/2019/08/16/maine-dairy-farmers-blood-tests-high-for-forever-chemicals-from-toxic-sludge/), and the viability of their farms and livelihoods is threatened by PFAS-contaminated beef and milk that is unsafe, inedible and unsaleable. Farmers and others who experience health problems, property damage and economic ruin from PFAS contamination should have clear access to the courts to sort out the blame and assess liability for actions taken by manufacturers and other responsible parties.

Unfortunately, Maine law creates unnecessary and unjust hurdles in the way of farmers and others injured by PFAS who seek compensation through civil actions. That's why IATP testified (https://www.iatp.org/documents/testimony-support-ld-2160-act-relating-statute-limitations-injuries-or-harm-resulting) in support of Rep. Ingwersen's legislation to update Maine law to recognize the unique characteristics of PFAS litigation. Access to the courts in all U.S. states is governed by statutes of limitation, which establish the timetable for filing lawsuits. While famously murder isn't subject to a statute of limitation, most civil actions are, and in Maine they must be filed "within 6 years after the cause of action accrues." (http://legislature.maine.gov/statutes/14/title14sec752.html)

Simply put, Maine's statute of limitations is both unclear and out of date. It was conceived of without understanding chemicals with properties such as PFAS, which silently and invisibly contaminate soil, water, plants and livestock; build up over time in food and in human bodies; travel far in groundwater and soils from where they were first applied; and persist for decades. Georgetown University adjunct law professor Scott Faber testified (https://mainelegislature.org/legis/bills/getTestimonyDoc.asp?id=141467) that Maine's law is unlike the statutes of limitations in 37 other states, all of which have been updated by legislatures or interpreted by the courts to incorporate the "discovery rule." That rule generally starts the clock ticking for filing a lawsuit at the time a plaintiff discovers or

reasonably should have discovered the harm or injury, as well as the link to the chemicals causing that harm. Dairy farmer Fred Stone found out about PFAS contamination of his drinking water and dairy herd decades after the sludge was spread as fertilizer. To require Stone to file a civil action soon after the sludge was spread, before he was even aware of the existence of PFAS, much less contamination of his land — as some interpret Maine's standard — would perpetrate a manifest injustice.

Despite applying for emergency aid from the U.S. Department of Agriculture (USDA), Fred Stone hasn't received help from the USDA or other sources, so a lawsuit against PFAS manufacturers may be his only option to receive some compensation. Importantly, LD 2160 implements the "polluter pays" principle that underlies Maine's longstanding approach to cleaning up and paying for pollution. This is why a majority of the members of Maine Governor Janet Mills' PFAS Task Force endorsed clarifying Maine's statute of limitations in their January 2020 final report (https://www.maine.gov/pfastaskforce/materials/report/PFAS-Task-Force-Report-FINAL-Jan2020.pdf). Taxpayer funding shouldn't be the first resort to pay for damage caused by these chemicals, especially where the manufacturers including 3M were well aware of the potential for harm (https://theintercept.com/2018/07/31/3m-pfas-minnesota-pfoa-pfos/) decades past, and have since discontinued production of some of these compounds to limit their liability. Unfortunately, the legacy of even discontinued PFAS formulations lives on, while newer PFAS compounds continue to be manufactured and remain ubiquitous in everyday consumer products.

Maine is not the only state where farms have been contaminated by PFAS and had to shut down. PFAS contamination has significantly harmed or shut down farming operations in Colorado (https://www.buzzfeednews.com/article/nidhisubbaraman/pfas-food-farms-milk-produce), New Mexico (https://www.aglaw.us/schroeder-ag-law-blog/2019/3/6/dairy-farm-pfas-contamination), Michigan (https://www.mlive.com/news/muskegon/2018/12/high-pfas-levels-found-at-grand-haven-horse-farm.html) and Wisconsin (https://www.milkbusiness.com/article/waste-pfas-spread-on-wisconsin-farmland), and it is likely that there are many more farms across the country that will be found to be contaminated if tested. Sewage treatment plants around the country have also developed composting operations that sell or give away composted fertilizer to home gardeners. Once thought to be an environmentally sound practice benefiting everyone, some of this compost has been found to be contaminated with PFAS including in Alaska (https://www.apnews.com/69b88cbcbcf34dfa9e68c980d6bc73c2) and Washington state (https://toxicfreefuture.org/toxic-pfas-chemicals-found-in-compost/;).

IATP will work with leading environmental organizations in Maine including the Environmental Health Strategy Group (https://www.ourhealthyfuture.org/media/mainers-urge-lawmakers-hold-%E2%80%98forever-chemicals%E2%80%99-polluters-accountable) and Conservation Law Foundation (https://legislature.maine.gov/legis/bills/getTestimonyDoc.asp?id=141464) to advocate to the full Maine Legislature to enact LD 2061 to protect the right to sue, once the legislature reconvenes after a hiatus caused by the COVID-19 pandemic. We will also push for more comprehensive measures to regulate PFAS and prevent future exposure. Phasing out land spreading of sewage treatment sludge is clearly long overdue, as this practice is proving to be a primary cause of agricultural PFAS contamination leading to economic devastation for farms and farmers, and is severely undermining the otherwise sterling reputation of Maine food as high quality and safe.

We submitted comments (https://www.iatp.org/documents/comments-maine-pfas-task-force-draft-final-report) to the Maine PFAS Task Force focusing on the need for more comprehensive testing of milk and other agricultural products, especially at the farm level, and expanded testing of soils and groundwater at hundreds of locations where sewage sludge was spread over decades. We have also called on the state to establish an enforceable Maximum Contaminant Level (MCL) for drinking water to protect health including that of vulnerable populations. The failure to establish an MCL makes Maine an outlier in the region, where Massachusetts, New Hampshire and Vermont have all acted to set a state MCL. These and other states establishing their own MCLs have rejected the voluntary EPA guidance (relied on by Maine) as insufficiently protective.

As states around the country tackle the daunting challenges of PFAS contamination, cleanup and compensation, IATP will be identifying best practices and working with allies at the state and federal level to ensure that agricultural, food and farming issues and the most effective solutions are front and center.

Filed under: Agriculture (/agriculture2)
PFAS (/taxonomy/term/721), Toxics (/keyword/toxics), Water (/keyword/water), Dairy (/issue/agriculture/dairy), Environment (/keyword/environment), Health (/keyword/health), State (/keyword/state)

Related documents

AGRICULTURE

Testimony in Support of LD 2160, "An Act Relating to the Statute of Limitations for Injuries or Harm Resulting from Perfluoroalkyl and Polyfluoroalkyl Substances" Joint Standing Committee on Judiciary, Maine Legislature (/documents/testimony-support-ld-2160-act-relating-statute-limitations-injuries-or-harm-resulting)

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