From: Pamela Castle
To: Benton Public Comment
Subject: References for verbal and written testimony for Pam Castle #31
Date: Wednesday, May 7, 2025 8:13:32 AM
Attachments: Reference A - townsend-et-al-2024-landfill-gas-a-major-pathway-for-neutral-per-and-polyfluoroalkyl-substance-(pfas)-release.pdf
Reference B - A critical review of perfluoroalkyl and polyfluoroalkyl substances (PFAS) landfill disposal in the United States.pdf
Reference C - Review of the fate and transformation of per- and polyfluoroalkyl substances (PFASs) in landfills.pdf
Reference D - tian-et-al-2018-occurrence-and-phase-distribution-of-neutral-and-ionizable-per-and-polyfluoroalkyl-substances-(pfass).pdf
Reference E - Per- and Polyfluoroalkyl Substance Toxicity and Human Health Review.pdf
Review F - Per- and polyfluoroalkyl substances (PFAS) in livestock and game species- A review.pdf

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Good morning,

While I provided the first page of each reference used in my verbal testimony last night, I only provided links in the written testimony I submitted yesterday (May 6). I understand now that commission members cannot use linked resources so I am submitting the references as PDFs of the complete papers.

Thank you and kudos for all the hard work you all are doing in this matter,

Pam Castle

Environmental Science & Technology LETTERS

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Letter

Landfill Gas: A Major Pathway for Neutral Per- and Polyfluoroalkyl Substance (PFAS) Release

Ashley M. Lin, Jake T. Thompson, Jeremy P. Koelmel, Yalan Liu, John A. Bowden, and Timothy G. Townsend*

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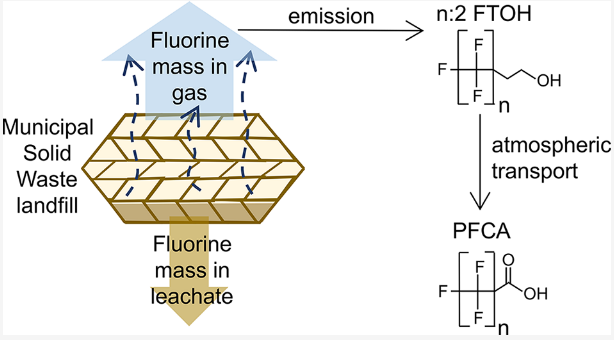
ABSTRACT: The undisclosed and ubiquitous use of perfluoroalkyl and polyfluoroalkyl substances (PFAS) in consumer products has led to a growing issue of environmental pollution, particularly within the solid waste community, where the fate of volatile (neutral) PFAS in landfilled refuse is not well understood. Here, three municipal solid waste landfills in Florida were assessed for neutral PFAS in landfill gas and ionic PFAS in landfill leachate to compare the relative mobility between the two pathways. Landfill gas was directly sampled using a high volume, XAD-2 resin based sampling approach developed for adsorption and analysis of 27 neutral PFAS. Across sites, 13 neutral PFAS were identified from fluorotelomer alcohol (FTOH), fluorotelomer olefin (FTO), secondary FTOH, fluorotelomer acetate (FTOAc), and fluorotelomer methyl acrylate (FTMAc) classes; however, FTOHs dominated concentrations (87−97% total neutral PFAS), with most detections surpassing utilized calibration levels. Even under conservative assumptions, the mass of fluorine leaving in landfill gas (32−76%) was comparable to or greater than the mass leaving in landfill leachate (24−68%). These findings suggest that landfill gas, a less scrutinized byproduct, serves as a major pathway for the mobility of PFAS from landfills.



KEYWORDS: *volatile, emissions, GC, fluorotelomer alcohol*

1. INTRODUCTION

Widespread per- and polyfluoroalkyl substance (PFAS) contamination has been a mounting environmental concern due to their chemical persistence and toxicity to human and biotic health.[1−4] While numerous industries are being confronted with PFAS-related management challenges, the burden of remediation and PFAS removal has often fallen on downstream industries—namely, the solid waste sector.[5−9] Discarded, PFAS-laden consumer products including textiles, wood products, and packaging and commonly landfilled industrial byproducts like MSW incineration ash and wastewater biosolids are known contributors to PFAS loading in landfills.[10−16] Existing research suggests most discarded PFAS mass is retained within landfills[9,17] with liquid-phase byproducts of waste decomposition, leachate and gas condensate, currently considered prevalent pathways for PFAS mobilization.[2,7,9] However, the extent of PFAS release to another major byproduct, landfill gas (LFG), has remained largely unscrutinized.

The bulk of PFAS characterization studies focus on nonvolatile/semivolatile (ionic) perfluoroalkyl acids (PFAAs) measured in liquid and solid matrices, in part because of a high presence and awareness of these species within the PFAS community but largely because analytical capabilities for ionic PFAS measurement are better established.[18−21] Volatile (neutral) PFAS are also utilized in consumer products[13,22−27,27] and have been determined in a few studies on ambient air surrounding landfills and near wastewater treatment plants,[28−32] but a lack of volatile analytical standards and latency in methodological development has hindered the progression of gas phase research in environmental matrices. Whereas PFAS characterization in leachate is established, concentrations ranging from thousands to tens of thousands of nanograms per liter are commonly encountered;[33−38] only two studies characterize volatile PFAS directly in LFG.[39,40] Titaley et al. identified fluorotelomer alcohol (FTOH), fluorotelomer acrylate (FTAc) and fluorotelomer olefin (FTO) homologues in LFG with combined concentrations ranging from 4,600 to 14,000 ng m^{-3} across three landfills. Goukeh et al., only assessing FTOHs, identified higher combined concentrations than Titaley et al., finding ∼18,000 ng m^{-3} (sum of 6:2 and 8:2

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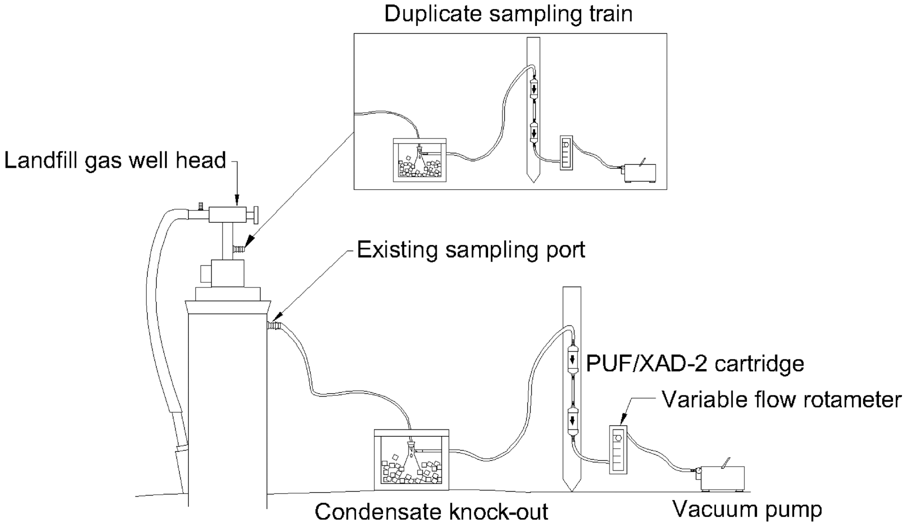

Figure 1. Developed system for sampling neutral PFAS directly from landfill gas well heads.

FTOH) in the one LFG sample examined. These studies suggest PFAS variability in LFG, which motivates further investigation, deploying higher sampling volumes[39] and larger analyte lists[40] to understand the potential presence of other neutral PFAS and distribution among landfills of different regions, compositions, and sizes.

With the ongoing development of PFAS regulation,[19] understanding the partitioning behavior of PFAS in major repositories like MSW landfills grows increasingly critical to minimize environmental and human risk. Unlike leachate, LFG is not always captured by collection systems, and management varies broadly across landfills, ranging from no treatment (i.e., passive venting) to some treatment (i.e., flaring, LFG to energy projects), but current treatment, if any, is not intended for PFAS.[41,42] Emerging research suggests the toxicity of volatile species (specifically 6:2 FTOH) to be significantly higher than their ionic counterparts via the inhalation pathway (a main route of exposure for volatile compounds).[43−47] Further, degradation of neutral species to ionic PFAAs once emitted to the atmosphere is well established.[48−58] The potential for long-range atmospheric transport of PFAS from landfills underscores the importance of considering neutral species and their fate during management to prevent further environmental contamination of highly scrutinized PFAAs such as perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS). As the only studies on LFG primarily identified FTOHs in LFG, the magnitude and significance of other neutral species remains unclear.

Here, LFG was sampled directly from gas well heads at three MSW landfill locations in Florida using a higher volume sampling protocol. XAD-2 resin sandwiched between polyurethane foam (PUF) was utilized for PFAS capture, then samples were analyzed for 27 volatile/semivolatile (neutral) PFAS via targeted gas chromatography high resolution mass spectrometry (GC-HRMS). To contextualize release in the gas phase, leachate was also collected at each landfill and analyzed for ionic PFAS (n = 93) using ultrahigh pressure liquid chromatography tandem mass spectrometry (UHPLC-MS/MS). The observed LFG and leachate concentrations were normalized on a mass of fluorine basis to compare the potential mobility in gas versus leachate matrices. This study provides foundational data critical for understanding the role of landfills in anthropogenic PFAS release and for informing LFG management.

2. METHODS AND MATERIALS

For brevity, materials and methods associated with ionic PFAS analysis in landfill leachate are provided in section 1 of the Supporting Information (Tables S-1 through S-4).

2.1. Standards and Reagents. Targeted neutral PFAS (≥97% purity, n = 27) were purchased from Wellington Laboratories Inc. (Guelph, ON, Canada), SynQuest Laboratories (Alachua, FL), and Chiron (Stiklestadveien, Trondheim, Norway). Nine classes of neutral PFAS (perfluoroalkane sulfonamides (FASAs), perfluoroalkane sulfonamidoethanols (FASEs), fluorotelomer acetates (FTOAcs), fluorotelomer methyl acrylates (FTMAcs), fluorotelomer iodides (FTIs), fluorotelomer secondary alcohols (sFTOHs), FTOHs, FTAcs, and FTOs) were measured using eight isotopically labeled internal standards (IS) from FASA, FASE, FTOH, and FTMAc classes for quantitation (Table S-5).

2.2. Sample Preparation and Collection. Polyvinyl chloride (PVC) cartridges filled with 4−5 g of Amberlite XAD-2 resin retained between two polyurethane foam (PUF) discs were utilized for PFAS capture.[29,59,60] Before use, XAD-2 sorbent was made PFAS-free through sequential Soxhlet extractions.[61] All cartridge components, sampling vessels, and tubing were sonicated in a mixture of Liquinox and PFAS-free water, rinsed, and then sonicated in methanol and methanol rinsed before use. Once dried and assembled, cartridges were stored in individually sealed polyethylene bags at 4 °C until sampling.

As neutral compounds were the focus of this investigation, aerosolized/particulate-bound PFAS were not specifically targeted for capture; however, a condensate collection system was included to prevent moisture interference. The developed sampling system (Figure 1) consisted of a condensate knockout (borosilicate, barbed Erlenmeyer flask contained in a cold box), two PUF/XAD-2 cartridges (installed in-series), a rotameter for flow control, a portable vacuum pump, and PFAS-free Tygon tubing. Before each sampling event, gas well head connection to the larger landfill gas collection system was disabled to create a neutral to positive pressure, workable for flow through the sampling system, then gas composition/

Table 1. Average Concentrations ($n = 2$) of 13 Neutral PFAS (ng m^{-3}) from Three Municipal Solid Waste Landfills in Florida (Site Characteristics Are Provided in Table S-9)[a]

	concentration (ng m^{-3})					
analyte	landfill 1	E	landfill 2	E	landfill 3	E
4:2 FTOH	220		ND		57	
6:2 FTOH	*>9,900*	**170,000**	*>6,000*	**22,000**	*>6,500*	**62,000**
8:2 FTOH	*>6,800*	**200,000**	*>6,000*	**140,000**	*>6,500*	**740,000**
10:2 FTOH	*>5,100*	**14,000**	*>3,000*	**23,000**	*>5,000*	**120,000**
12:2 FTOH	860		1,400		5,000	
5:2 sFTOH	*>2,900*	**8,800**	*>1,700*	**9,000**	*>1,900*	**5,900**
7:2 sFTOH	320		*>1,300*	**13,000**	*>1,400*	**11,000**
8:2 FTO	2,500		1,300		550	
10:2 FTO	650		840		540	
12:2 FTO	97		580		160	
8:2 FTOAc	610		90		490	
10:2 FTOAc	99		19		140	
6:2 FTMAc	3,800		56		150	

[a] Concentrations of 6:2, 8:2, and 12:2 FTOH and 5:2 and 7:2 sFTOH consistently exceeded the upper limit of developed calibration ranges; therefore, both a minimum concentration (assuming the highest calibration concentration) and a maximum extrapolated concentration are provided. Italicized values denote a minimum concentration. Column "E" presents average maximum concentrations. "ND" denotes non-detect measurements. FTACs, FASAs, FASEs, FTIs, and 8:2 FTMAc were not detected in any samples. Analyte acronyms and details are provided in Table S-5.

temperature was recorded using an Optimax Biogas analyzer (MRU Instruments, Humble, TX). Duplicate sampling trains were connected to existing gas well sampling ports. Approximately 1,200 L was sampled through each train at a flow rate of 5 L min^{-1}. After sampling, PUF/XAD-2 cartridges were sealed and individually stored at ≤4 °C for transport/storage. Quality control (QC) procedures are provided in the SI, section 2.

2.3. Extraction and Analysis. Spent XAD-2 from each cartridge was weighed and transferred to a 50 mL polypropylene centrifuge tube and vortexed, and approximately 2 g aliquoted for extraction. Samples were spiked with a mixture of mass labeled IS (Table S-5), rotated end-over-end for 18 h in 4 mL of 75/25% (v/v) ethyl acetate and methanol, and centrifuged for 10 min at 4,000 rpm. Supernatants were transferred to 15 mL centrifuge tubes, and the extraction process was repeated, combining supernatants from the two-fold extraction. Extracts were concentrated to 3 mL via gentle nitrogen evaporation, aliquoted, and stored no more than 30 days at −20 °C until analysis. QC details are provided in the SI, section 2 (Table S-6 and Figure S-1).

Targeted analysis of 27 neutral PFAS by positive chemical ionization (PCI) with selected ion monitoring (SIM) was conducted using a Thermo Scientific TRACE 1310 gas chromatograph coupled to a Thermo Scientific Orbitrap Exploris GC 240 mass spectrometer (GC-HRMS; see SI, section 2 for details regarding GC separations and instrumentation). A 12-point external calibration curve (from 1 to 2,000 pg μL^{-1}) was developed for quantitation, prepared through serial gravimetrically derived dilutions of primary stock solutions. A mixture of mass labeled IS at concentrations of 150 pg μL^{-1} was added to each calibration level. When a labeled standard was not available for a compound, a labeled standard with a similar retention time or structure was utilized for quantitation (Table S-5).

3. RESULTS AND DISCUSSION

Unexpectedly, several neutral PFAS concentrations in LFG exceeded the implemented calibration levels. Because of considerable exceedance for some compounds, dilution would reduce IS below instrument detection; therefore, in instances where sample concentrations exceeded calibration limits, two concentrations are presented (Equation S-1): a minimum value which assumes the highest calibration concentration and a maximum extrapolated concentration. Fluorine mass release calculations utilize minimum values, preventing overextrapolation while providing a conservative estimate for leachate comparison. Even under these assumptions, substantial concentrations of neutral PFAS, higher than those previously observed, were identified. Future assessments should deploy shorter sampling durations to refine findings.

3.1. Neutral PFAS in Landfill Gas. Except for 4:2 FTOH in one landfill, 13 PFAS were detected in duplicate samples across the three sites (site characteristics are provided in Table S-9). Observed concentrations are displayed in Table 1. At minimum, combined concentrations of neutral PFAS in LFG ranged from 22,000 to 33,000 ng m^{-3}. Considering extrapolated values, total concentrations ranged from 210,000 to 940,000 ng m^{-3}, an order of magnitude higher than those previously reported in LFG.[39]

3.1.1. FTOHs and sFTOHs in Landfill Gas. Like previous studies on LFG and air surrounding landfills, FTOHs dominated neutral PFAS concentrations;[28,31,32,39,40] however, extrapolated concentrations in this study surpassed previous reports in LFG, in some cases by 2 orders of magnitude, and were more comparable (although much lower) to concentrations recently identified in soil vapor near a PFAS manufacturing facility.[55] While there are uncertainties given the degree of extrapolation, the magnitude of FTOHs found in this study compared to existing research suggests fundamental differences potentially related to sampling methodology (e.g., much larger sampling volumes) and/or sampled landfill characteristics (e.g., waste type, age, air intrusion), although these data were not available for comparison. Across the three sites, 6:2, 8:2, and 10:2 FTOH, combined, made up 87 to 97% of total concentrations, but 8:2 FTOH alone constituted 50 to 79%. The shortest and longest analyzed homologues, 4:2 and

12:2 FTOH, were significantly lower in concentration (Table S-10). This is supported by previous FTOH distributions determined from source fluoro-telomer polymers[62] and observations in LFG, urban air, and air surrounding wastewater treatment/landfill sites.[28−31,39,40,63] Concentrations of 12:2 FTOH were of similar magnitude to those in Titaley et al., but 4:2 FTOH has not been detected in LFG, suggesting MSW landfills to be a previously unidentified potential source of atmospheric 4:2 FTOH.[39]

Secondary FTOHs have not been targeted in gas-phase landfill research but have been identified in condensate associated with LFG collection systems.[37] As intermediary byproducts of 6:2 and 8:2 FTOH biodegradation to PFAAs, 5:2 and 7:2 sFTOH, were unsurprisingly elevated, they were at least an order of magnitude lower than respective parent FTOH homologues.[64−66] All detections of 5:2 sFTOH and two out of three detections of 7:2 sFTOH were above calibration, combined sums attributed to 2 to 10% of total concentrations.

3.1.2. Other Neutral PFAS in Landfill Gas. Other neutral PFAS fell within acceptable calibration ranges and together accounted for 0.22 to 1.9% of total concentrations. FTO homologues have been encountered in other LFG and ambient air studies, but in past assessments 8:2 and 10:2 FTO were below limits of quantitation and 12:2 FTO concentrations were consistently an order of magnitude higher than those reported here.[39,59] To the authors' knowledge, 8:2 and 10:2 FTOAc and 6:2 FTMAc have not been determined in LFG. FTOAcs are not commonly assessed analytes but are associated with fluoropolymer textile treatments and have been identified in one indoor air study from Japan.[67,68] Similarly, 6:2 FTMAc has only been analyzed in a few studies on cosmetics and wastewaters but at lower concentrations.[69−71]

3.2. Comparative Fluorine Mass Release between Landfill Byproducts.

Normalizing PFAS concentrations on a fluorine basis allows comparisons to be drawn between different matrices and PFAS types (e.g., gas−liquid, neutral−ionic, precursor−terminal). This methodology is widely used to assess the "mass balance" of PFAS within systems, given that the long-term environmental fate of measurable PFAS is transient, whereas the mass of fluorine is conserved.[17,72,73]

Here, the same approach is utilized to compare the PFAS mobility in leachate versus LFG pathways. Neutral (Table 1, minimum values) and ionic (Table S-4) PFAS concentrations in LFG and leachate from this study were individually normalized to a mass of fluorine (Equation S-2) using compound specific fluorine mass fractions (Table S-8). Summed fluorine masses in leachate and LFG were then scaled according to site-specific annual generation volumes reported for each landfill (Table S-9).[41] A caveat of this comparison is the absence of measurements for neutral species in leachate and ionic species in LFG; however, the literature suggests FTOHs (the dominant neutral class identified) predominantly exist in the gaseous phase, while PFAAs exist in liquid or particulate phases.[28,74] Subsequent research should assess neutral and ionic compounds in both matrices to validate findings and further elucidate the PFAS behavior in landfills.

Even utilizing minimum concentrations observed in LFG, equal magnitudes of fluorine release are observed between LFG and leachate at each site (Figure 2)—contrasting from existing estimates of PFAS mass flow from landfills.[9] Existing estimates, based on limited data, suggest that most PFAS mass mobilized from landfills releases through leachate (~62%).[9] However, our data from Landfill 1, showing over 76% fluorine release in LFG, along with substantial masses released by LFG in Landfills 2 and 3 (at minimum 40% and 32%, respectively), indicate that LFG may serve as an equal, likely greater, conduit of PFAS mobility from landfills than leachate, concurring with previous reactor studies on FTOH volatilization and neutral/ionic PFAS assessments of select waste materials.[17,75,76]

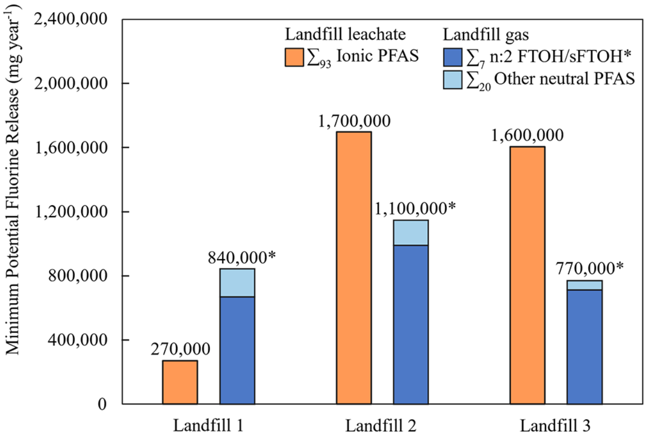

Figure 2. Annual fluorine mass release in landfill leachate versus landfill gas (LFG) from three municipal solid waste landfills in Florida. Fluorine masses in leachate are derived from ionic PFAS ($\sum_{93}$ PFAS) concentrations measured in leachate from each site (Table S-4) multiplied by the annual leachate generation volume and scaled using each detected compound's fluorine mass fraction (Tables S-8, S-9). The same methodology was applied for neutral PFAS ($\sum_{27}$ PFAS) in LFG by using the average of minimum concentrations (Table 1). Asterisked (*) values denote input FTOH/sFTOH concentrations which were above calibration levels developed for this study and therefore assumed to be at the highest calibration concentration. Consequently, these findings should be viewed as minimum values which conservatively estimate the magnitude of PFAS mobility in leachate versus LFG.

At least 79 to 92% of the fluorine mass in LFGs were derived from FTOH/sFTOH classes, with minimal contribution from FTOs, FTOAcs, and FTMAcs. In this conservative assessment, fluorine from LFG surpassed leachate in only Landfill 1. Although actual fluorine emission from LFG is higher than reported here, the elevated ratio of gas-to-leachate generation at Landfill 1 likely caused this difference (Table S-9). Landfill 2, the largest site, demonstrated the highest combined fluorine release from leachate and LFG, followed by Landfill 3, and then Landfill 1, corresponding to descending waste mass in place at each location.

4. IMPLICATIONS

This study provides fundamental data about neutral PFAS in LFG from MSW landfills. Unexpectedly, FTOH/sFTOH detections in LFG from this study exceeded implemented calibration levels; subsequent research should deploy shorter sampling durations. Regardless, even under more conservative assumptions these findings suggest that LFG, largely unscrutinized for PFAS, contains similar or greater magnitudes of PFAS compared to leachate, mostly attributed to midlength FTOH homologues. As landfills can be viewed as unabating PFAS repositories, the significance of LFG management in mitigating the long-term, long-range atmospheric transport of neutral PFAS, and subsequently derived PFAAs, cannot be understated. Unlike landfill leachate, LFG collection systems (when in place) are not fully efficient, collecting an estimated ~50−70% of generated biogases.[77] Though this is a considerable collection efficiency of biogas and presumably neutral PFAS, management of captured LFG fractions varies globally, from no treatment to degrees of carbon filtration and thermal treatment (i.e., flaring, advanced renewable natural gas technologies). Because the feasibility of PFAS destruction through thermal treatment remains unclear, research is needed to determine the treatment/removal efficiency of existing LFG management technologies. Considering the range of LFG capture efficiency, the retention and emission of neutral PFAS via fugitive emissions (i.e., migration through the waste layer) should also be examined, along with the role of landfill waste type, age, and temperature in neutral PFAS variability.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.4c00364.

> Acronyms, structures, and instrumental parameters for neutral PFAS via GC-HRMS and ionic PFAS via LC-MS/MS; detailed QC information and results; and fluorine mass balance/extrapolation details and assumptions (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Timothy G. Townsend − *Department of Environmental Engineering Sciences, College of Engineering, University of Florida, Gainesville, Florida 32611, United States;* orcid.org/0000-0002-1222-0954; Phone: 352-392-0846; Email: ttown@ufl.edu; Fax: 352-392-3076

Authors

Ashley M. Lin − *Department of Environmental Engineering Sciences, College of Engineering, University of Florida, Gainesville, Florida 32611, United States*

Jake T. Thompson − *Department of Environmental Engineering Sciences, College of Engineering, University of Florida, Gainesville, Florida 32611, United States*

Jeremy P. Koelmel − *School of Public Health, Department of Environmental Health Sciences, Yale University, New Haven, Connecticut 06511, United States*

Yalan Liu − *Department of Environmental Engineering Sciences, College of Engineering, University of Florida, Gainesville, Florida 32611, United States; Department of Civil, Environmental and Geomatics Engineering, Florida Atlantic University, Boca Raton, Florida 33431, United States*

John A. Bowden − *Department of Environmental Engineering Sciences, College of Engineering and Center for Environmental and Human Toxicology and Department of Physiological Sciences, College of Veterinary Medicine, University of Florida, Gainesville, Florida 32611, United States;* orcid.org/0000-0003-3135-4015

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.4c00364

Notes

The authors declare no competing financial interest.

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Review

A critical review of perfluoroalkyl and polyfluoroalkyl substances (PFAS) landfill disposal in the United States

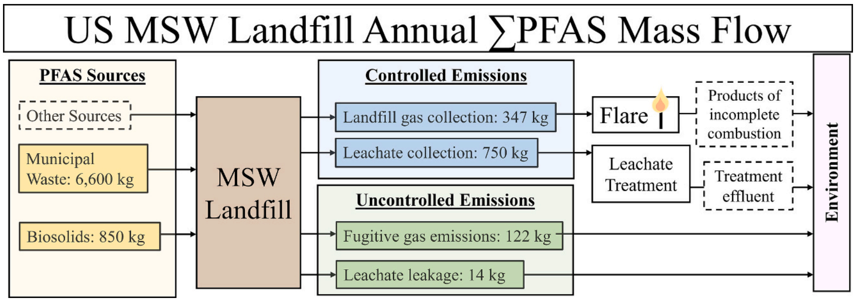
Thabet Tolaymat[a,*], Nicole Robey[b], Max Krause[a], Judd Larson[c], Keith Weitz[c], Sameer Parvathikar[c], Lara Phelps[d], William Linak[d], Susan Burden[e], Tom Speth[a], Jonathan Krug[d]

[a] The Center for Environmental Solutions and Emergency Management, Office of Research and Development, U.S. Environmental Protection Agency, Cincinnati, OH, USA
[b] Innovative Technical Solutions, Gainesville, FL, USA
[c] RTI International, Research Triangle Park, NC, USA
[d] The Center for Environmental Measurements and Modeling, Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC, USA
[e] Office of Science Advisor, Policy and Engagement, Office of Research and Development, U.S. Environmental Protection Agency, Washington, DC, USA

HIGHLIGHTS

- Solid waste management strategies impact PFAS emissions.
- PFOA has the highest ratio to its respective RSL in C&D and MSW landfill leachates.
- Unlined C&D landfills present a significant source of PFAS to the environment.
- An estimated 7.5 metric tons of PFAS enter MSW landfills annually.
- Annually, 460 kg of PFAS emitted via landfill gas, 750 kg via landfill leachate.

GRAPHICAL ABSTRACT



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ABSTRACT

Landfills manage materials containing per- and polyfluoroalkyl substances (PFAS) from municipal solid waste (MSW) and other waste streams. This manuscript summarizes state and federal initiatives and critically reviews peer-reviewed literature to define best practices for managing these wastes and identify data gaps to guide future

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Abbreviations: µg, Microgram; AFFF, Aqueous film-forming foams; C&D, Construction and demolition; diPAP, Polyfluoroalkyl phosphoric acid diesters; FASA, Perfluoroalkane sulfonamide; FASE, Perfluoroalkane sulfonamido ethanol; FTAc, Fluorotelomer acrylate; FTCA, Fluorotelomer carboxylic acid; FTO, Fluorotelomer olefin; FTOH, Fluorotelomer alcohol; H_2S, Hydrogen sulfide gas; HAL, Health Advisory Limit; kg, Kilogram; L, Liter; LFG, Landfill gas; MCL, Maximum Contaminent Level; MeFBSAA, Methyl-n-perfluorobutanesulfonamidoacetic acid; MeFOSAA, Methyl-n-perfluorooctanesulfonamidoacetic acid; mg, Milligram; Mg, Megagram (metric ton); MPCA, Minnesota Pollution Control Agency; MSW, Municipal solid waste; MSWI, Municipal solid waste incineration; NEBRA, North East Biosolids & Residuals Association; NWRA, National Waste & Recycling Association; PFAA, Perfluoroalkyl acids; PFAS, Per- and polyfluoroalkyl substance(s); PFBS, Perfluorobutane sulfonate; PFCA, Perfluoroalkyl carboxylic acids; PFHxA, Perfluorohexanoic acid; PFHxS, perfluorohexane sulfonate; PFOA, Perfluorooctanoic acid; PFOS, Perfluorooctane sulfonic acid; PFPeA, Perfluoropentanoic acid; PFSA, Perfluoroalkyl sulfonic acids; PIC, Product of incomplete combustion; RCRA, Resource Conservation and Recovery Act; RNG, Renewable natural gas; RO, Reverse osmosis; RSL, Regional Screening Limit; SI, Supplementary information; US EPA, United States Environmental Protection Agency; WWTP, Wastewater treatment plant.

[*] Corresponding author.

E-mail address: Tolaymat.Thabet@epa.gov (T. Tolaymat).

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research. The objective is to inform stakeholders about waste-derived PFAS disposed of in landfills, PFAS emissions, and the potential for related environmental impacts. Furthermore, this document highlights data gaps and uncertainties concerning the fate of PFAS during landfill disposal. Most studies on this topic measured PFAS in liquid landfill effluent (leachate); comparatively fewer have attempted to estimate PFAS loading in landfills or other effluent streams such as landfill gas (LFG). In all media, the reported total PFAS heavily depends on waste types and the number of PFAS included in the analytical method. Early studies which only measured a small number of PFAS, predominantly perfluoroalkyl acids (PFAAs), likely report a significant underestimation of total PFAS. Major findings include relationships between PFAS effluent and landfill conditions – biodegradable waste increases PFAS transformation and leaching. Based on the results of multiple studies, it is estimated that 84% of PFAS loading to MSW landfills (7.2 T total) remains in the waste mass, while 5% leaves via LFG and 11% via leachate on an annual basis. The environmental impact of landfill-derived PFAS has been well-documented. Additional research is needed on PFAS in landfilled construction and demolition debris, hazardous, and industrial waste in the US.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) include thousands of unique manufactured chemical compounds with a hydrophobic carbon-fluorine chain and a functional group that may be hydrophilic or hydrophobic. PFAS provides beneficial properties for many consumer products and industrial applications, mostly stick- and stain-resistance and surfactant qualities. PFAS's usefulness has led to a nearly ubiquitous presence in our lives, and PFAS's stability, due to the strength of carbon-fluorine bonds, result in long half-lives and the nickname "forever chemicals."

Human exposure to PFAS has been linked to detrimental health effects which impact all systems, including reproductive effects such as decreased fertility or increased high blood pressure in pregnant women, developmental effects or delays in children, including low birth weight, accelerated puberty, bone variations, behavioral changes, increased risk of some cancers, including prostate, kidney, and testicular cancers, reduced ability of the body's immune system to fight infections, including reduced vaccine response; interference with the body's natural hormones and increased cholesterol levels and risk of obesity (reviewed by Fenton et al., 2021). In response to the growing body of evidence identifying PFAS as a significant threat to human health and the environment, the United States Environmental Protection Agency (US EPA) is undertaking research to determine the impact of PFAS via a risk paradigm approach: (1) determine toxicity, (2) understand exposure, (3) assess risk, and (4) find and innovate effective treatment and remediation techniques and strategies. Because PFAS-containing products are disposed of at the end of their useful lives, significant PFAS quantities are managed with solid waste in the US and elsewhere. Properly managing solid waste via containment, treatment, and destruction is essential to protecting our environment and reducing the risk of harmful exposures.

Recognizing the impact of PFAS on human health and the environment, the US EPA released its first provisional Health Advisory Levels (HALs) for PFAS in drinking water in 2009. As analytical capabilities and scientific understanding of PFAS health impacts have improved, the Agency has promulgated additional guidance and risk-based thresholds. For the first time, in 2023, the US EPA proposed enforceable drinking water regulatory limits to reduce human exposure to PFAS (US EPA, 2022d). In April of 2021, the US EPA released the PFAS Strategic Roadmap, which outlines the EPA's commitments to action for 2021 through 2024. Information about US EPA PFAS initiatives is summarized in ***Table S1*** of the Supplementary information (SI), and applicable limits are included in Table 1. The US EPA has also proposed designating PFOA and PFOS as hazardous substances under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) and is considering adding certain PFAS to the Resource Conservation and Recovery Act (RCRA) list of hazardous constituents (US EPA, 2022b).

At the State level, all the US states except Arkansas, Louisiana, Mississippi, Nebraska, Texas, and Wyoming have dedicated websites providing PFAS-specific information. Some states have banned PFAS-containing products, as summarized in ***Table S2*** (SI). In contrast, others have initiated their own regulatory limits and advisory guidelines, as presented in ***Table S3*** (SI). Eight states have undertaken specific actions and introduced or passed bills targeting PFAS in solid waste (see ***Table S4***, SI). Notably, PFAS regulations are rapidly evolving, and any documentation of state-level PFAS initiatives will likely be outdated quickly.

Confronted with significant quantities of PFAS managed in landfills, the solid waste community struggles to understand the best means to manage PFAS-containing waste streams. Many studies have evaluated PFAS in landfills. However, there is a need for a critical review of the literature that would define the best methodologies for managing these wastes and identify data gaps to guide future research. This manuscript aims to inform the public and stakeholders from the solid waste industry

Table 1
Average concentrations (ng L^{-1}) of select PFAS in landfill leachate and US EPA risk-based thresholds.

Leachate matrix		PFOA		PFOS		PFNA		PFBS		PFHxS		PFHxA	5:3 FTCA
		Mean (n)	DF	Mean (n)	DF	Mean (n)	DF	Mean (n)	DF	Mean (n)	DF	Mean (n)	Mean (n)
MSW		*1400 (284)*	*23*	260 (284)	6.6	69 (234)	1.2	910 (234)	0.1	540 (234)	1.4	2800 (225)	3500 (86)
CDD		*1100 (17)*	*19*	660 (17)	17	50 (17)	0.8	530 (17)	0.1	2200 (17)	5.7	1600 (17)	1400 (17)
MSWI Ash		*800 (40)*	*13*	400 (40)	10	59 (40)	1.1	1400 (40)	0.2	510 (40)	1.3	1300 (40)	700 (40)
HW (Primary)		4900 (24)	81	*4100 (24)*	*102*	530 (24)	8.7	6500 (24)	1.1	12,000 (24)	32	12,000 (24)	NM
HW (Secondary)		*100 (5)*	*1.7*	14 (5)	0.4	40 (5)	0.7	57 (5)	0.01	86 (5)	0.2	440 (5)	NM
EPA limit (ng L^{-1})	Tapwater RSL (HQ = 1.0)	60		40		59		6000		390		n/a	n/a
	Lifetime HAL	0.004		0.020		n/a		2000		n/a		n/a	n/a
	Proposed MCL	4		4		n/a		n/a		n/a		n/a	n/a

(HAL = health advisory level; MCL = maximum contaminant level; RSL = regional screening level; HQ = hazard quotient; DF = average dilution factor required to meet RSL; NM = not measured).
Italicized values represent the controlling dilution factor.

about PFAS entering the waste stream and being disposed of in landfills, potential landfill PFAS emissions, and the related environmental impacts. Furthermore, this document highlights data gaps and uncertainties concerning the fate of PFAS during landfill disposal. Data were compiled and summarized, as described in the Methods section of the SI (***Section S2*** and ***Table S5***), to provide a concise critical review of this evolving research topic.

2. Solid waste management in the United States

A detailed discussion of solid waste management in the US is included in the SI (Section S3). Residents, businesses, and industries in the United States (US) generate significant amounts of solid waste; overall municipal solid waste (MSW) generation in 2018 was 265 million metric tons (US EPA, 2020b). In addition to MSW, significant amounts of construction and demolition (C&D) waste (545 million metric tons), wastewater treatment plant (WWTP) biosolids (2.5 million metric tons), and varied amounts of industrial waste and disaster debris enter the US solid waste management system every year (US EPA, 2020b). Over time, MSW generation in the US has increased. While the fraction of MSW which is landfilled has decreased from over 90% in 1960 to 50% in recent years, the mass of MSW disposed of in landfills reached its highest recorded level at 133 million metric tons in 2018 (see Fig. S1 in the SI). The US's landfill design, monitoring, and classification are identified and regulated according to the RCRA described in the SI's RCRA section. RCRA and its regulations provide requirements for landfill engineering controls based on the type of waste the landfill receives (MSW (Subtitle D), Hazardous (Subtitle C), industrial, construction, and demolition (C&D) debris) as outlined in the SI.

2.1. Sources of PFAS in solid waste

While extensive research has been undertaken to measure PFAS in effluent from waste management activities (particularly landfill leachate), fewer studies have attempted to estimate the PFAS load entering the waste management sector. Coffin et al. (2022) estimated an extractable $\sum$PFAS concentration in MSW of 50 μg kg^{-1} based on concentrations in MSW screenings reported by Liu et al. (2022a). Estimating PFAS loading to landfills is not only complicated by analytical challenges and the diversity of measurable PFAS, but also by the heterogeneity of MSW and other waste streams (e.g., household products, building materials, industrial waste, and "other wastes"). The following subsections focus on waste representing suspected high PFAS load or a significant fraction of the waste stream. Fig. 1 presents PFAS concentrations measured in various products and the environment compared to those measured in landfill leachate, compost, and biosolids from WWTPs.

2.1.1. Municipal solid waste

In the US, household waste is among the most significant fractions of MSW. Few studies measured the PFAS concentration of suspected PFAS-containing consumer products in the context of direct exposure during product use (Buck et al., 2011; Favreau et al., 2017; Glüge et al., 2020; Guo et al., 2009; Herzke et al., 2012; ITRC, 2022; Kotthoff et al., 2015; OECD, 2022; US FDA, 2022; Ye et al., 2015). These findings indicate a significant load of PFAS remaining in products at the end of their useful life. Household waste consists of two main categories: the biodegradable fraction and the non-biodegradable fraction. Both types of waste streams contain PFAS, but the fate of their PFAS may differ.

2.1.1.1. Biodegradable fraction. Paper and paperboard are the most

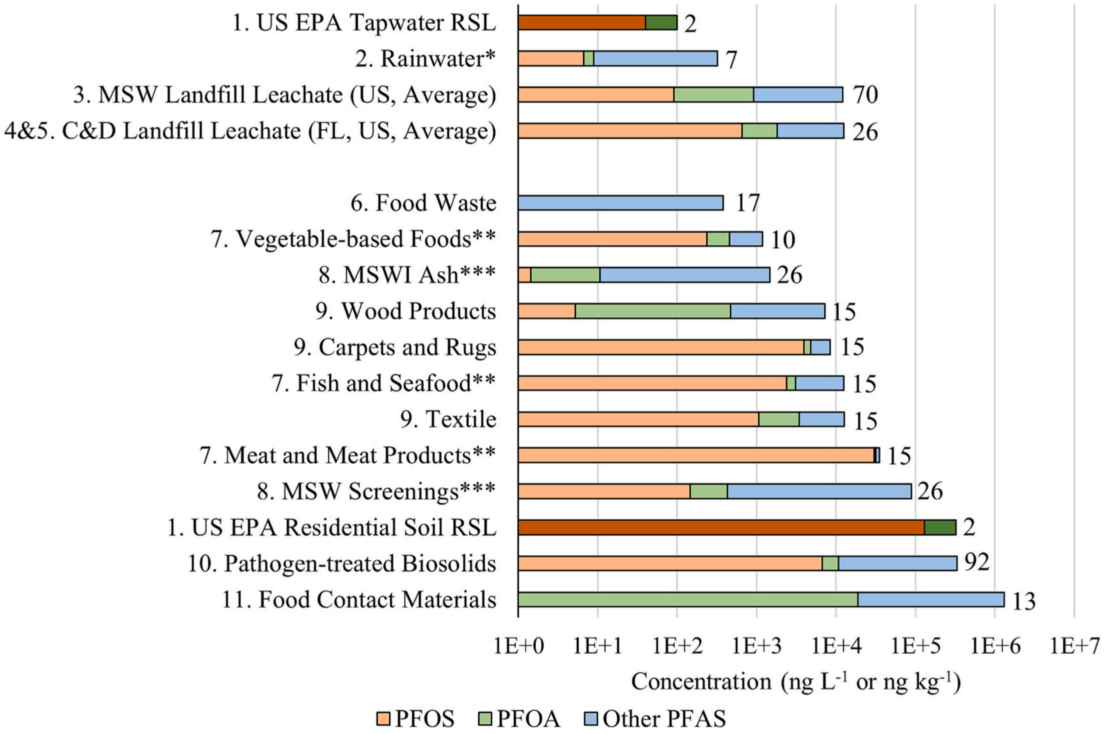

Fig. 1. PFAS concentrations and compositions measured in various products, wastes, and the environment compared to MSW landfill leachate.
* includes ultra-short chain PFAS, TFA.
** upper bound of the mean.
*** minimum total PFAS based on leachable fraction.
Note: numbers prior to matrix type refer to sources. Numbers to the right of the bars are the number of PFAS analytes. RSLs refer to risk-based screening levels, not enforceable regulatory limits. Sources: 1. US EPA (2022d) 2. Pike et al. (2021) 3. Lang et al. (2017) 4. Solo-Gabriele et al. (2020) 5. Chen et al. (2023) 6. Thakali et al. (2022) 7. European Food Safety Authority (2012) 8. Liu et al. (2022b) 9. Bečanová et al. (2016) 10. Thompson et al. (2023a, 2023b) 11. Siao et al. (2022).

abundant components of MSW, representing 23% of the US MSW generation in 2018 (US EPA, 2021a). PFAS are often added to paper products to improve stick and stain resistance, which results in paper products (including food packaging) consistently reported as a significant source of PFAS for human exposure and in the waste stream (Curtzwiler et al., 2021; D'eon et al., 2009; Ramírez Carnero et al., 2021; Robel et al., 2017; Seltenrich, 2020; Yuan et al., 2016; Zabaleta et al., 2016). In a review of studies that measured PFAS in food-contact materials, Siao et al. (2022) reported concentrations of $\sum_{13}$PFAS in food packaging as high as 8500 μg kg^{-1}; at these concentrations, paper and paper products likely contribute significantly to the overall PFAS loading in MSW, as well as contamination of food and food waste. Sapozhnikova et al. (2023) used targeted and total oxidizable precursor (TOP) assays to measure PFAS migration from food packaging into food products among 88 packaged food samples. TOP analysis identified a significant portion of total PFAS in packaging came from unknown precursor PFAS; average $\sum_8$PFAA was 28 μg kg^{-1} before oxidation and 380 μg kg^{-1} after oxidation. Migration from the packaging into food was found to increase over the course of the ten-day study. Unfortunately, many new products marketed as environmentally-friendly alternatives to plastic products have been found to contain PFAS (Timshina et al., 2021), and advocacy groups in the US and beyond have moved to revise compostable labeling to preclude PFAS-containing products (BioCycle, 2020). Disubstituted polyfluoroalkyl phosphates (diPAPs) have been found to represent a significant fraction of the PFAS used in paper products. However, most studies do not include diPAP as an analyte (D'eon et al., 2009; Thompson et al., 2023a). These findings suggest the concentration of $\sum$PFAS in paper products may be significantly higher than current estimates.

Another large fraction of biodegradable household waste is food waste, accounting for 22% of the MSW generated in the US in 2018 (US EPA, 2021a, 2021b, 2021c). Unlike paper products, PFAS are not intentionally added to food; contact with PFAS-containing equipment, packaging, water, feed, or soil amendments may result in residual PFAS. Several studies have been published describing the potential migration of PFAS from PFAS-impregnated food packaging (Ramírez Carnero et al., 2021). Up to 33% of extractable PFAS on the surface of food contact materials have been reported to migrate to simulated foods – the migration efficiency depends on the food type and PFAS class (Yuan et al., 2016). Additionally, some PFAS are known to bioaccumulate in the food chain. A European Food Safety Authority (2012) report lists seafood and meat as the food categories most frequently reported containing measurable concentrations of PFAS, with PFOS and PFOA quantified most commonly in 29% and 9% of samples, respectively. The same study estimated mean overall dietary exposure for PFOS and PFOA ranging from 0.07 to 32 ng kg^{-1} body weight per day, with lower exposure rates for 14 additional PFAS. Exposure was highest among toddlers and children due to higher food consumption for body size. Among 25 samples of food waste analyzed for PFAS by Thakali et al. (2022), 17 contained PFAS (mean $\sum_{17}\text{PFAS} = 0.38$ μg kg^{-1}); PFOS and PFOA were not detected in any of the samples.

Wood and yard trimmings represent approximately 18% of the US MSW generation (US EPA, 2020b). While natural wood and plant matter are unlikely to contain significant concentrations of PFAS (Thompson et al., 2023b), engineered wood building materials may be coated with PFAS to enhance performance. In a study of PFAS content in consumer and building materials, 100% of oriented strand board and wood products analyzed contained measurable PFAS concentrations, with median and maximum Σ_{15}PFAA of 5 and 18 μg kg^{-1}, respectively (Bečanová et al., 2016).

2.1.1.2. Non-biodegradable fraction. In the non-biodegradable category of household waste, carpets, and textiles have been consistently found to contain intentionally added PFAS that provide stick and stain resistance and waterproof properties (Kallee and Santen, 2012; Kim et al., 2015; Lang et al., 2016; Peaslee et al., 2020; van der Veen et al., 2022). A review of Σ_{15}PFAA in various household and consumer products found textiles, floor covering, and car interior materials represented the three highest maximum concentrations (78, 38, and 36 μg kg^{-1}, respectively); the highest non-biodegradable median PFAS concentration was from insulation (3.6 μg kg^{-1}) (Bečanová et al., 2016). PFAS and fluoropolymers are also used in non-stick cookware (Sajid and Ilyas, 2017) and electronics to provide smudge resistance, insulation, and other properties. An estimated 114 separate PFAS have been identified in electronic production (Garg et al., 2020). PFAS contamination and exposure through e-waste management have been the subject of several studies (Garg et al., 2020; Tansel, 2022; B. Zhang et al., 2020). Notably, the measurement of PFAS in e-waste itself (as opposed to through leachate, environmental contamination, or dust) is limited. A range of 0.07–0.43 μg kg^{-1} PFOS among all electronic products is provided by Garg et al. (2020). Σ_{15}PFAA reported by Bečanová et al. (2016) ranged as high as 11.7 μg kg^{-1} (median: 0.4 μg kg^{-1}) in electronic and electrical equipment (EEE) and as high as 2.2 μg kg^{-1} (median: 1.4 μg kg^{-1}) in waste EEE.

2.1.2. *Industrial waste*

Industrial processes generate waste and effluent in large volumes; processes that use PFAS, such as the leather tannery, chrome plating, and textile industries, represent a significant contribution of PFAS to the solid waste stream (ITRC, 2022) which are often disposed of in landfills. Other types of industrial processes which generate PFAS-containing waste involve the management of PFAS-contaminated materials, including the separation of wastewater biosolids as part of municipal wastewater treatment, the management of MSW incineration residuals (MSWI ash), and the disposal of PFAS-contaminated soils and other residuals generated as part of environmental cleanup processes.

2.1.2.1. Biosolids. WWTPs manage residential, commercial, and industrial wastewater, including landfill leachate, and have been the subject of many PFAS studies (Lenka et al., 2022). Biosolids account for a significant fraction of WWTP effluent (Fredriksson et al., 2022) and impact PFAS loading to the environment and landfills (Johnson, 2022; Thompson et al., 2023b). Reported PFAS concentrations in biosolids vary with the number of PFAS included in the analytical method. Gallen et al. (2018) reported mean $\sum_9$PFAS of 45 μg kg^{-1} of biosolids; Thompson et al. (2023a, 2023b) reported mean $\sum_{92}$PFAS of 500 μg kg (dry)$^{-1}$ in untreated biosludge and 330 μg kg(dry)$^{-1}$ in biosolids (biosludge treated for pathogen removal), indicating that early studies of PFAS in biosolids which measured fewer PFAS, and predominantly PFAAs, did not capture a significant portion of the total PFAS. Over 5.8 million dry metric tons of biosolids were managed in the US in 2018, of which 30% was managed in landfills, 15% was incinerated, and over 50% was used as a soil amendment (NEBRA, 2022).

2.1.2.2. MSW incineration ash. The incineration of MSW for energy recovery (MSWI) produces two solid waste streams – bottom ash, the material that does not burn, and fly ash, fine particulate matter collected in the air pollution control system. Approximately 13% of MSW in the US is managed through incineration (US EPA, 2021a, 2021b, 2021c), resulting in an estimated 7.5 million tons of MSWI ash (Liu et al., 2019). Few studies have measured PFAS in MSWI ash. Liu et al. (2021b) reported $\sum_{21}$PFAS in fly and bottom ash from three facilities in China, with concentrations ranging from 1.5 to 88 μg kg^{-1} in fly ash and from 3.1 to 77 μg kg^{-1} in bottom ash. Based on the concentrations of PFAS in a laboratory leaching study, the average minimum $\sum_{26}$PFAS in MSWI ash from a US facility was 1.5 μg kg^{-1} (Liu et al., 2022b); this represents a conservative estimate of total PFAS. These concentrations are in the same range as MSW. Incineration temperatures may not be sufficiently high to mineralize or destroy PFAS, and operational strategies likely play a significant role in the fate of PFAS during incineration. The impact of temperatures on PFAS leaching from MSWI is discussed in greater detail in the context of MSWI ash monofill leachates.

2.1.2.3. Manufacturing wastes. There is extensive use of PFAS in some industries, as PFAS is added intentionally to products (i.e., to produce stain-resistant properties in textiles and paper products) and as part of the manufacturing process (i.e., to facilitate demolding). This results in unintentionally contaminated materials through contact. The Interstate Technology Review Committee (ITRC) thoroughly lists PFAS uses in the industrial and manufacturing sectors (ITRC, 2022). PFAS-laden manufacturing waste is often sent to landfills for disposal across industries.

Among specific industries and industrial wastes which have been the subject of PFAS analysis, high-concentration effluents from electronic industries have been described in the literature; photolithographic effluent in Taiwan contained 130,000 ng L^{-1} each of PFHxS and PFOS (Lin et al., 2009); liquid effluent from television and circuit board manufacturing contained 1600 ng L^{-1} of $\sum_{11}$PFAS (Kim et al., 2016); sludge effluent collected from an electronics industry location in South Korea contained 91 μg kg^{-1} of $\sum_{11}$PFAAs (Kim et al., 2016). PFAS are used commonly in paper processing and treatment; a case study in Norway identified PFAS impacts downstream of a landfill used for paper factory waste disposal (Langberg et al., 2021). Chrome plating industry waste sludges are designated hazardous wastes (F006), which contain high concentrations of PFAS (ITRC, 2022) and are therefore managed in Subtitle C hazardous waste landfills. A study of chrome sludge in China identified PFOS as the most predominant PFAS at concentrations as high as 2435 μg kg^{-1} (Qu et al., 2020). The chrome plating industry consumes an estimated 6500 kg of PFOS annually (Garg et al., 2020).

2.1.2.4. PFAS remediation residuals. Sites with high levels of PFAS contamination from the historical use of PFAS-containing aqueous film-forming foams (AFFF) or other releases are frequently remediated, and the contaminated media is commonly disposed of in landfills (either with the waste or used as daily cover). Remediation approaches include mobilization of PFAS and collection of the leachate, sorption of PFAS using activated carbon or other sorbents, or soil excavation for landfill disposal (Bolan et al., 2021; Ross et al., 2018). Brusseau et al. (2020) reviewed PFAS concentrations measured in soils from contaminated sites, reporting median PFOA and PFOS concentrations of 83 and 8700 μg kg^{-1}, respectively, with concentrations as high as 50,000 μg kg^{-1} for PFOA and 460,000 μg kg^{-1} for PFOS.

3. Fate of PFAS in landfills

The fate of solid waste-derived PFAS within landfills is dominated by transformation and partitioning. Many PFAS species are persistent in the environment and PFAS that are degradable can transform into more recalcitrant, typically more environmentally mobile PFAS (Bolan et al., 2021). The partitioning behavior of PFAS are related to the chemical structure of individual species, both according to PFAS class, functional groups, and chain length among homologous species. In turn, the ongoing transformation will impact partitioning behavior (Robey et al., 2020; Zhang et al., 2020a, 2020b; Smallwood et al., 2023). In landfills, PFAS may partition to the liquid phase (leachate) and gaseous phase (landfill gas; LFG), remain sorbed to the waste, and/or interact with the engineering controls of the landfills (e.g., leachate collection systems, gas collection, and control systems). PFAS that are resistant to degradation and minimally soluble or volatile, such as certain polymeric PFAS, have historically been presumed to remain immobile and sequestered in landfills, although more recent studies have called this assumption into question (Lohmann et al., 2020).

3.1. PFAS transformation

Many studies observed the transformation of PFAS precursors into terminal species under abiotic and microbially active aerobic and anaerobic conditions. While this section briefly reviews these processes to provide context to PFAS in landfills, the aim is not to conduct an exhaustive review of the topic, which is available in other reviews (Lu et al., 2023).

3.1.1. Abiotic transformation

PFAS transformation pathways under abiotic conditions include oxidation, photolysis, and thermal degradation (ITRC, 2020; Washington and Jenkins, 2015). While the bulk of PFAS transformations in organic-rich landfills are likely a result of biodegradation, these abiotic processes play an essential role in solid waste management systems. PFAS such as fluorotelomer alcohols (FTOHs) can volatilize under temperatures typical in landfills (35–55 °C). Once in the atmosphere, FTOH can transform via photolysis or other chemical reactions into perfluorocarboxylic acids (PFCAs) which are then deposited on land and waterbodies (Esfahani et al., 2022; Martin et al., 2006). Other abiotic processes include thermal degradation. An increase in the temperature of waste may facilitate PFAS transformation. Wastes within landfills may be exposed to temperatures insufficient to mineralize or defluorinate PFAS but which may cause precursor transformations. Thompson et al. (2023a, 2023b) measured higher concentrations of diPAPs in biosolids that had undergone any form of heat treatment, including heat drying as well as higher temperature vector reduction treatment, indicating the presence and transformation of unidentified precursors.

3.1.2. Aerobic transformation

Aerobic environments exist at the early stages of landfill decomposition. The waste still contains atmospheric oxygen in its void space and likely contributes to the transformation of PFAS in waste. Thompson et al. (2023a, 2023b) observed a proportional increase in PFCAs after aerobic biosolids composting, especially short-chain (perfluoropentanoic acid, PFPeA, and perfluorohexanoic acid, PFHxA). Similarly, Li et al. (2022) found significant increases among short-chain PFAAs (including PFBS and PFOS) in aerobically treated anaerobic digestor sludge. These findings are significant because short-chain PFAS are more mobile in the environment, more likely to be uptaken by plants (Ghisi et al., 2019), and more challenging to treat (Ross et al., 2018). Multiple studies have shown that aerobic decomposition facilitates the transformation of precursor PFAS to shorter-chain terminal PFAS, such as PFOA and PFOS (Hamid et al., 2020; Lee et al., 2010a; Liu et al., 2010; Lott et al., 2023; Rhoads et al., 2008; Schultz et al., 2006; Wang et al., 2009, 2011; Zhao et al., 2013).

3.1.3. Anaerobic transformations

Comparatively, fewer studies have documented PFAS transformation under anaerobic or methanogenic conditions similar to landfills. Liu et al. (2021a) compared 52 PFAS in leachate from waste collection vehicles to anaerobic MSW landfill leachate and concluded the vehicle leachate contained proportionally more precursor PFAS and short-chain PFAAs compared to the landfill leachate as a result of the transformation in the anaerobic landfill environments. Studies of anaerobic precursor transformation identified FTCAs as the predominant by-product of FTOH degradation. Allred et al. (2015) reported increased MeFBSAA and FTCA leaching over abiotic reactors in biologically active landfill microcosm reactors, indicating that methanogenic biological transformation was responsible for the increase. Zhang et al. (2013) observed the accumulation of FTCAs in landfills over time, concluding that FTCAs are indicators of FTOH transformation, while Lang et al. (2016) and Weber et al. (2022) reported PFOA accumulation in leachate as a result of precursor transformation under anaerobic experimental conditions. Lang et al. (2016) attributed this to the longer experimental duration, with PFOA appearing as a significant degradation by-product only 200+

days into the 550-day experiment.

3.2. PFAS partitioning in landfills

3.2.1. PFAS partitioning to the liquid phase

PFAS concentrations in landfill leachate are a function of multiple factors, including the PFAS profile of the incoming waste stream and conditions within the landfill, which, in turn, correspond with waste composition, stage of decomposition, and environmental factors, especially rainfall precipitation. These factors also affect the physical-chemical aspects of the leachate quality, and any discussion of PFAS in leachate should also include matrix contextualization. The number of PFAS that can be detected and quantified in landfill leachate has grown. Early methods were able to quantify 24 PFAS compounds in three classes (Huset et al., 2011), but improvements have been made; more recent studies attempted to measure 92 PFAS and detected 53, as presented in Table 2.

3.2.1.1. PFAS in landfill leachate by type

3.2.1.1.1. MSW landfills. The vast majority of PFAS landfill leachate data are measured from MSW landfills (Allred et al., 2014; California Water Boards, 2023; Chen et al., 2022, 2023; Huset et al., 2011; Lang et al., 2017; Liu et al., 2022a; Masoner et al., 2020; NWRA, 2020; Solo-Gabriele et al., 2020). The ∑PFAS content of MSW landfill leachate in published US studies ranges from BDL - 125,000 ng L^{-1} with an average of 10,500 ng L^{-1} and a weighted average of 12,600 ng L^{-1}. The weighted average is notably similar to the estimated average ∑PFAS concentration reported by Lang et al. (2017) using Monte Carlo simulation. Often, the ∑PFAS content heavily depends on the number of unique PFAS measured in the study, which ranged from two to 70 for MSW landfill leachate (see SI ***Fig. S2***). For comparison among studies, we will focus on PFAS with corresponding US EPA tapwater Regional Screening Levels (RSL) (i.e., PFOA, PFNA, PFBS, PFHxS, PFOS, and Gen-X), as presented in Table 1. Except for Gen-X, which has only been quantified in a single sample of landfill leachate from a North Carolina MSW landfill with a history of accepting PFAS manufacturing wastes (NWRA, 2020), the remaining five PFAS are reliably quantified in all published landfill leachate studies. Other PFAS which reliably contribute significantly to ∑PFAS in landfill leachates, PFHxA and 5:3 FTCA, are also included in Table 1.

PFAS concentrations have also been reported for leachates from MSW landfills in other countries, including Australia (Gallen et al., 2016, 2017), Europe (Ahrens et al., 2011; Busch et al., 2010; Eggen et al., 2010; Fuertes et al., 2017; Kallenborn et al., 2004; Knutsen et al., 2019; Perkola and Sainio, 2013; Woldegiorgis et al., 2005), and Asia (Huang et al., 2022; Liu et al., 2022b; Yan et al., 2015; Yin et al., 2017; Zhang et al., 2014). International differences in waste composition, sample collection, and analytical processes can impact reported PFAS concentrations, making a direct comparison of the overall PFAS content challenging. Concentrations of PFOS and PFOA, which have been reliably measured in most or all studies, are included for eight countries in ***Table S6*** and described by Travar et al. (2020).

Table 2
Number of PFAS measured and ∑PFAS among published landfill leachate studies.

Matrix	Number of samples	Number of PFAS detected (in Method)	Average ∑PFAS (ng L^{-1})	∑PFAS range (ng L^{-1})	Country	Reference
MSW LL	1	38 (51)	9700	9700	USA	Liu et al. (2021a)
	1	32 (51)	9400	9400	USA	Robey et al. (2020)
	78	25 (26)	12,700	300–58,000	USA	Chen et al. (2023)
	4	10 (11)	17,200	15,000–18,000	USA	Solo-Gabriele et al. (2020)
	6	24 (24)	4700	2700–7400	USA	Huset et al. (2011)
	40	30 (70)	12,200	2000–29,000	USA	Lang et al. (2017)
	11	2 (2)	840	330–2600	USA	Clarke et al. (2015)
	19	28 (28)	5400	230–29,000	USA	Helmer et al. (2022)
	39	2 (2)	1500	47–3400	USA	EGLE (2019)
	9	22 (25)	24,300	1400–125,000	USA	NWRA (2020)
	131	31 (40)	17,500	BDL – 104,000	USA	California Water Boards (2023)
	17	14 (14)	3000	33–15,000	Australia	Gallen et al. (2016)
	94	9 (9)	6100	210–46,000	Australia	Gallen et al. (2017)
	22	15 (15)	7000	Not reported	Australia	Simmons (2019)
	6	25 (43)	6100	31–13,000	Germany	Busch et al. (2010)
	11	24 (24)	9800	2500–36,000	Canada	Benskin et al. (2012)
	31	16 (18)	2700	700–6400	Canada	Li (2009)
	10	2 (2)	*	50–2300	Canada	Gewurtz et al. (2013)
	2	16 (27)	4200	2200–6100	Norway	Eggen et al. (2010)
	5	7 (8)	770	200–1500	Norway	Kallenborn et al. (2004)
	2	4 (4)	400	210–610	Finland	Perkola and Sainio (2013)
	48	7 (10)	2400	14–17,500	Ireland	Harrad et al. (2019)
	4	8 (16)	1100	640–1400	Spain	Fuertes et al. (2017)
	12	28 (30)	1700	320–11,000	Norway	Knutsen et al. (2019)
	10	17 (26)	490	0.3–1300	Sweden	Gobelius et al. (2018)
	5	11 (14)	82,100	7300–290,000	China	Yan et al. (2015)
	9	33 (57)	42,900	3040–109,000	China	Liu et al. (2022b)
	6	17 (17)	14,200	1800–43,300	China	Huang et al. (2022)
	12	18 (18)	4060	1270–7660	Singapore	Yin et al. (2017)
	5	8 (9)	6000	4200–11,000	Australia	Gallen et al. (2017)
CDD LL	13	24 (26)	9500	270–30,500	USA	Chen et al. (2023)
	2	11 (11)	15,500	14,000–16,000	USA	Solo-Gabriele et al. (2020)
MSWIA LL	2	9 (11)	3100	2800–3400	USA	Solo-Gabriele et al. (2020)
	31	26 (26)	7300	39–54,500	USA	Chen et al. (2023)
MSW GC	21	26 (26)	12,200	199–80,900	USA	Chen et al. (2023)
	12	53 (92)	19,000	3000–50,000	USA	Smallwood et al. (2023)
HW LL (Primary)	24	17 (28)	68,000	570–377,000	USA	California Water Boards (2023)
HW LL (Secondary)	5	13 (24)	1800	25–3700	USA	California Water Boards (2023)

(LL = landfill leachate; MSW = municipal solid waste; CDD = construction and demolition debris; MSWIA = MSW incineration ash; GC = gas condensate).
[*] Gewurtz et al. (2013) do not provide detailed data to calculate average ∑PFAS.

3.2.1.1.2. C&D landfills. PFAS were detected in all C&D landfill leachate samples analyzed across three studies with $\sum$PFAS ranging from 270 to 30,500 ng L^{-1} (weighted average 10,300 ng L^{-1}). Solo-Gabriele et al. (2020) and Chen et al. (2023) found no significant difference in the total measured PFAS between leachate from MSW and C&D landfills. The analytical method used by Chen et al. (2023) included 18 terminal PFAS (PFAAs) and eight precursors (FASAs, FTCAs, and FTSs). The study, however, reported a significant difference in the fraction of $\sum$Terminal and $\sum$Precursor species between MSW and C&D landfill leachates. C&D leachate contained, on average, 86% terminal PFAS, while MSW leachate contained 64% terminal PFAS (Chen et al., 2023). This could be attributed to the different types of PFAS present in each waste stream and the type of biological activity prevalent in each landfill type. Because C&D landfills contain proportionally less food waste and more concrete and gypsum drywall, the prevailing landfill conditions result in higher pH leachate and proportionally more sulfate chemical species in the leachate as opposed to ammonia, which is typically at higher concentrations in MSW landfill leachate (Townsend et al., 1999). Further, due to those differences in leachate conditions, microbial differences result from presence of different carbon sources as well as electron donors and acceptors. Generally, sulfur-reducing bacteria are found in higher concentrations at C&D landfills due to higher amounts of sulfate, while methanogens are more prevalent at conventional landfills (Meyer-Dombard et al., 2020).

Fig. 2 includes the range of concentrations for PFAS with RSLs for MSW and C&D landfill leachate; average PFHxS concentrations were higher in C&D landfill leachate than in MSW landfill leachate, and PFBS concentrations were lower in C&D landfill leachate. Waste composition is highly variable between landfills as well as over time at an individual landfill, so, while limited studies may suggest potential sources of select PFAS in C&D debris (e.g., higher concentrations of PFHxS may be attributed to their use in carpeting and other building materials (Beesoon et al., 2012; Jin et al., 2011), generalizations about specific sources may not be appropriate. Gallen et al. (2017) measured nine terminal PFAS in Australian C&D landfill leachates (n = 5), reporting average $\sum_9$PFAS concentrations of 6000 ng L^{-1} (compared to 6100 ng L^{-1} in 94 MSW leachates from the same study).

Unlike MSW landfills, at the US federal level, C&D landfills do not require a bottom liner and leachate collection systems. This contributes to the lower number of studies describing PFAS in C&D relative to MSW landfill leachate and an increase in the probability of groundwater contamination from C&D compared to MSW landfills. Average concentrations of PFOA, PFOS, PFNA, PFBS, and PFHxS, along with corresponding US EPA risk-based thresholds (HALs, MCLs, and RSLs), are included in Table 1. PFOA poses the most significant challenge as its concentration in C&D landfill leachate would have to be diluted by 19 to meet the tapwater RSL or by 287 to meet the US EPA proposed MCL.

3.2.1.1.3. MSWI Ash monofills. Solo-Gabriele et al. (2020) and Chen et al. (2023) found leachate from MSWI ash monofills to have lower $\sum$PFAS concentrations than leachate from MSW landfills. Solo-Gabriele et al. (2020) reported $\sum_{11}$PFAS in MSWI ash monofill leachates ranging from 2800 to 3400 ng L^{-1} and inversely correlated with incineration temperature. $\sum_{11}$PFAS in leachate from MSWI ash that underwent incineration at 800 °C was almost three times higher than after incineration at 950 °C. The decrease indicates loss of measurable PFAS via mineralization (i.e., destruction), volatilization (i.e., air emission), or transformation to PFAS species which are not measured in standard analytical methods (e.g., products of incomplete combustion or PICs). Leachates from MSWI ash which had undergone incineration at 950 °C, still contained >2000 ng L^{-1} of PFAS, indicating PFAS are not fully mineralized at these operating conditions. Liu et al. (2021b) reported substantially higher $\sum_{21}$PFAS in MSWI ash leachate from three facilities in China, with concentrations ranging from 127,000–450,000 ng L^{-1}. The study did not report incineration temperatures or other operating conditions.

However, when MSWI ash was co-disposed with other wastes, such as MSW or biosolids, $\sum$PFAS concentration in the leachate was on par with that in MSW landfill leachate (Solo-Gabriele et al., 2020; Liu et al., 2022a). Liu et al. (2022a) found the co-disposal of a small fraction (e.g., 4%) of MSW, including biosolids, with MSWI ash resulted in leachate concentrations that were comparable to MSW landfill leachate, suggesting liquids are preferentially flowing through and leaching PFAS from the non-incinerated waste as opposed to the ash. While MSWI ash-derived leachates have lower concentrations of PFAS, these studies suggest care should be taken to dispose of MSW and MSWI ash separately, and more research is needed to understand the fate of PFAS during MSW incineration.

3.2.1.1.4. Industrial landfills. Unlined industrial landfills that received residuals from manufacturing PFAS and PFAS-containing products have been linked to contamination of local groundwater sources. Notable examples include the House Street landfill in Belmont, Michigan which received tannery waste (US EPA, 2022e); Crown Vantage landfills in Parchment, Michigan (MPART, 2020), that were used to dispose of paper mill waste from the production of laminated paper

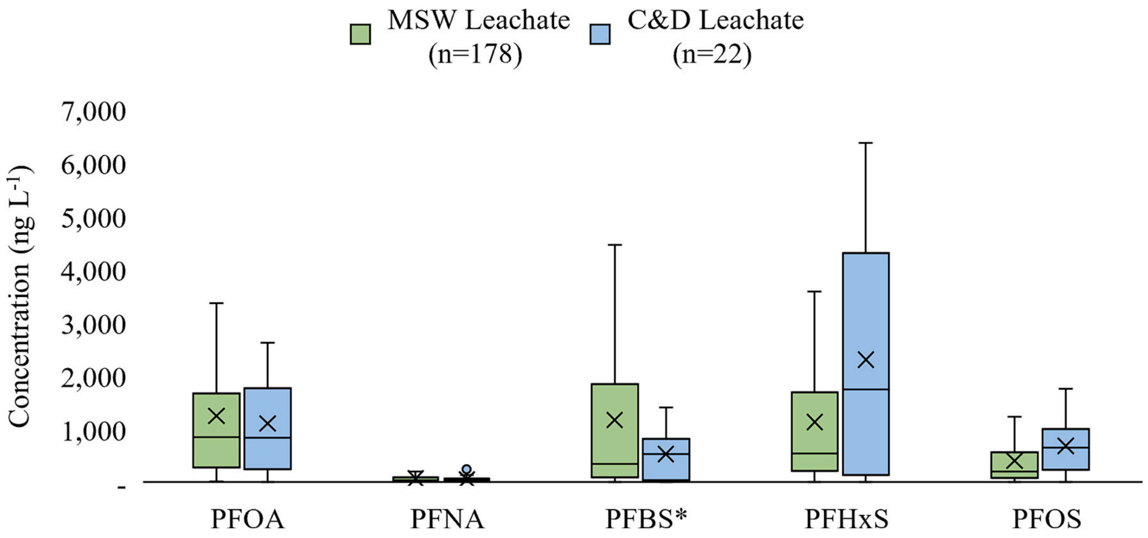

Fig. 2. Average concentrations of five PFAS with US EPA tapwater RSLs.
Data from Gallen et al. (2017), Solo-Gabriele et al. (2020), and Chen et al. (2023).
* Gallen et al. (2017) did not include PFBS analysis.

products; and the 3M Woodbury disposal site in Washington County, Minnesota, that was used to dispose of PFAS production waste. As part of this literature search, no leachate PFAS concentration data from industrial landfills in the US were located. However, Kameoka et al. (2022) measured PFAS in leachate from three industrial landfills in Japan; $\sum_{17}$PFAA concentrations averaged 45,000 ng L^{-1}.

3.2.1.1.5. Hazardous waste landfills. Although PFAS are not federally regulated as listed hazardous wastes, some solid wastes managed in Subtitle C hazardous waste landfills contain PFAS (as discussed in Section 2.1.2, e.g., chrome-plating sludge), while other PFAS-containing wastes may meet hazardous waste characteristic criteria (e.g., flammable, corrosive, etc.). Some hazardous waste landfills have also reported receipt of AFFF waste at their sites. No peer-reviewed studies have evaluated PFAS concentrations in leachate collected from hazardous waste landfills; however, California Water Boards have released PFAS concentrations for landfill leachate, including two hazardous waste landfills in California (California Water Boards, 2023). The data for these sites are included in the SI ***Table S7***. Among 29 samples from the two sites, $\sum_{24}$PFAS and $\sum_{28}$PFAS concentration was as high as 377,000 ng L^{-1} (average 68,000 ng L^{-1}), substantially higher than MSW, C&D debris, or MSWI ash landfill leachates (see Table 2). In the US, hazardous waste landfill disposal requires waste pre-treatment to minimize contaminant mobility – land disposal restrictions for hazardous waste are described in the Code of Federal Regulations (CFR; 40 CFR § 268). Leaching studies have shown minimal PFAS immobilization using traditional solidification techniques (Barth et al., 2021), which may explain elevated PFAS concentrations in the leachate.

US hazardous waste landfills must also use secondary leachate collection systems; California's database includes five samples of secondary hazardous waste landfill leachate from one site, with $\sum_{24}$PFAS averaging 1800 ng L^{-1} (see Table 2). Without exception, for all sampling locations with both primary and secondary leachate PFAS data, concentrations for individual and $\sum_{24}$PFAS were higher in the primary compared to the secondary leachate. While the absence of biological decomposition in hazardous waste landfills may minimize the microbially-mediated precursor transformation to PFAAs, waste treatment methods (e.g., lime treatment) may also impact transformation and partitioning, possibly oxidizing precursor PFAS. Hazardous waste pretreatment standards are designed to minimize traditional hazardous waste constituent leaching (e.g., lime treatment stabilizes metals and neutralizes acidic waste) and have not been optimized for PFAS stabilization; PFAS fate, transport, and transformations under hazardous waste pretreatment processes are not well understood. Because of the strict Subtitle C landfill operation requirements and the pre-treatment of wastes, leachate generation in these landfills is typically minimal, and any leachate which is produced is often managed as hazardous waste (i.e., not discharged to WWTP, as other landfill leachates often are).

3.2.1.2. *Other factors impacting PFAS concentrations in leachate*

3.2.1.2.1. Waste age. As waste degrades under the anaerobic conditions of biologically active landfills, the overall PFAS concentrations in the leachate and the ratio of the terminal to precursor species have been found to increase. Lang et al. (2017) reported leachate from waste older than ten years had significantly lower concentrations of PFNA, 8:2 FTCA, 5:3 FTCA, PFBS, MeFBSAA, and MeFOSAA than leachate from younger waste. These differences could be attributed to changes in the PFAS formulations in commercial products and/or the conversion of PFAA precursors. Liu et al. (2021a) measured PFAS in leachate from waste collection vehicles alongside leachate from the receiving MSW landfill. The study found significantly higher $\sum_{51}$PFAS concentrations in landfill leachate which had undergone further biological decomposition. Furthermore, Liu et al. (2021a) also reported a difference in PFAS profiles likely caused by the transformation of precursor PFAS in landfill environments.

3.2.1.2.2. Leachate quality. Although most PFAS behavior and solution chemistry studies focus on remediation technologies, generalizations regarding PFAS phase partitioning also apply to landfill leaching (Z. Du et al., 2014). Comparatively, fewer studies have explored PFAS partitioning in the context of leachate chemistry. In a landfill simulator study, Allred et al. (2015) observed increases in longer-chain PFCA and perfluoroalkyl sulfonic acids (PFSA) concentrations when biodegradation reached the methanogenic stage. At this stage, increased methanogenic and secondary fermentation and decreased volatile fatty acid concentrations from the acidogenic stage result In increased pH, more neutral pH, which is theorized to deprotonate waste surfaces, resulting in less sorption of PFAS to the degrading organic matter. This theory is supported by the results described by Solo-Gabriele et al. (2020), where a significant positive correlation was reported between PFAS concentrations and increasing leachate pH. This effect has also been observed in several previous landfill leachate sampling studies (Benskin et al., 2012; Gallen et al., 2017; Hamid et al., 2018; Yan et al., 2015).

In addition to partitioning behavior, the PFAS profile of landfill leachate is a function of PFAA precursor transformation resulting from biodegradation. Biological activity is catalyzed by landfill moisture, resulting in higher landfilled waste temperatures and more PFAS transformation. In a study of WWTP biosolids pathogen removal, precursor transformation and apparent increases in $\sum_{92}$PFAS, driven by increased PFAA content, resulted after aerobic composting and increased diPAP concentrations from heat treatment (Thompson et al., 2023b). Based on a nationwide study of 95 leachate samples collected from 18 landfills, leachate from MSW landfills in US regions with high annual precipitation showed significantly greater $\sum_{19}$PFAS than comparable landfills in arid locations (Lang et al., 2017); see Table 2 for all US-based studies included in this review. Further, leachate generation volume is significantly higher in regions that experience more precipitation. As a result, landfills in arid regions are estimated to contribute <1% of the nationwide landfill leachate PFAS mass load (Lang et al., 2017). When studies have evaluated the short-term impacts of precipitation on PFAS in landfill leachate, however, leachate PFAS concentration decreased within a day of a precipitation event due to dilution (Benskin et al., 2012; Gallen et al., 2017). Normalization of PFAS concentrations to bulk parameters such as chloride or total dissolved solids may be able to account for such dilution.

3.2.2. *PFAS partitioning to the gas phase*

MSW contains a proportionally more biodegradable organic matter which undergoes anaerobic decomposition in landfill environments compared to other waste streams (e.g., C&D). The decomposition of organic matter produces MSW LFG, which is, on average, about 50% methane (CH_4), and 50% carbon dioxide (CO_2), with a small fraction consisting of other gaseous and volatile constituents (Wang et al., 2021). LFG at MSW landfills is collected and managed according to the requirements of the US EPA New Source Performance Standards (US Clean Air Act, 40 CFR § 60). According to the US EPA's Landfill Methane Outreach Program (LMOP) August 2022 database, 1230 of the 2635 MSW landfills in the US have gas collection systems in place, and 1157 have flares in place (US EPA, 2022a). C&D LFG is rarely collected in the US, as C&D landfills contain less biodegradable organic matter and produce less LFG than MSW landfills. Additionally, C&D LFG contains proportionally more $H_2S_{(g)}$ produced by sulfur-reducing bacteria and the decomposition of gypsum disposed of as drywall.

Gas generation and composition at other landfill types has yet to be the subject of significant research. MSWI ash monofills are not expected to generate LFG because there is minimal biodegradable matter in the ash; however, the co-disposal of WWTP biosolids, MSW, or any degradable organic matter with MSWI ash will produce biogas as a result of decomposition. Gas generation at industrial landfill sites is primarily a function of the type of waste deposited. Organic waste like pulp and paper mill sludges will likely generate gas requiring management. In general, Subtitle C hazardous waste landfills in the US do not contain putrescible organic waste and do not generate biogas.

3.2.2.1.1. PFAS in MSW landfill gas. PFAS volatilization and release from MSW landfills within the gaseous phase is receiving an increased focus driven by advances in volatile PFAS measurement (Riedel et al., 2019) and an improved understanding of PFAS chemistry. The partitioning coefficients (e.g., Henry's constant) for ionizable PFAS are significantly lower than neutral PFAS (Abusallout et al., 2022), making ionizable PFAS less likely to volatilize under typical MSW landfill conditions. Experimental measurement of PFAS vapor pressures similarly suggests FTOHs (i.e., neutral PFAS) are more readily volatilized than PFCAs (i.e., ionizable PFAS) and that vapor pressure decreases logarithmically with carbon chain length in homologous species (M. Zhang et al., 2020). Measurement and data of PFAS in actual MSW LFG are still minimal.

In a 2007 analysis of landfills that accepted PFAS-containing industrial wastes, the MPCA detected several PFAS (12 PFAAs and perfluorooctane sulfonamide, PFOSA) in MSW LFG with $\sum_{13}$PFAS ranging from 4.1 to 18.7 ng m^{-3} (MPCA, 2010). Titaley et al. (2023) measured neutral PFAS in LFG of three active MSW landfills ($n = 12$ samples) and reported concentrations of four n:2 FTOHs ($n = 6, 8, 10,$ and 12), one fluorotelomer acrylate (6:2 FTAc), and one fluorotelomer olefins (12:2 FTO). Concentrations for individual PFAS range from 270 to 4900 ng m^{-3}, and the total measured neutral PFAS for each landfill was, on average, between 4600 and 14,000 ng m^{-3} (weighted average across all samples: 10,200 ng m^{-3}). Smallwood et al. (2023) reported FTOH in LFG condensate, which, when normalized to gas volume, was three orders of magnitude lower than the gaseous phase concentrations reported by Titaley et al. (see SI ***Table S8*** for calculations), indicating FTOHs preferentially partition to the gas phase; FTOHs may transform in the atmosphere into PFCAs, such as PFOA, which have known and suspected toxic effects.

3.2.2.1.2. PFAS in C&D landfill gas. While no data exist on the concentration of PFAS in C&D LFG, it can be conservatively assumed, based on data from MSW LFG measurements, that PFAS also leave C&D landfills via gas effluent. As previously described, PFAS-containing wastes are disposed of at C&D landfills, and it is highly likely C&D debris contains volatile PFAS, such as FTOHs, which readily transform into FTCAs and PFCAs as a result of biodegradation and environmental oxidation, respectively. Lower rates of biological activity in C&D landfills may result in slower biodegradation of PFAS like FTOH (and other volatile precursors) may persist longer in C&D compared to MSW landfills and therefore have more opportunity to volatilize and leave the landfill via LFG. This is likely offset by the lower volume of LFG generated overall at C&D landfills compared to MSW. Nonetheless, this read-across should be validated by experimental data.

3.3. Fate of PFAS in traditional landfill leachate and gas management systems

Most landfills compliant with New Source Performance Standards (Clean Air Act) and RCRA must capture gas effluent and leachate to minimize environmental impacts. Leachate is often intercepted using a low-permeability bottom liner made of high-density polyethylene, collected, and may be transported off-site to a WWTP, disposed of using deep well injection, or otherwise managed and treated on-site.

PFAS interactions with low-permeability landfill liners have been the subject of limited studies. Most landfill liners are constructed from polyethylene geomembranes. Laboratory studies of PFAS diffusion through linear low-density polyethylene report below detection diffusion rates (Di Battista et al., 2020). Diffusion through high-density polyethylene has yet to be reported but maybe even lower due to differences in material structure. Landfill liner integrity – the absence of flaws or holes – is the most critical factor in preventing PFAS transmission through geomembrane and composite liners (Di Battista et al., 2020). An analysis of landfill liner performance reported median leakage rates of 44 and 33 L ha^{-1} day^{-1} for geomembrane and composite liners, respectively, with an overall liner collection efficiency of 98% (Jain et al., 2023). Compacted clay liners, which are more common in older landfills and C&D landfills, do not adsorb PFAS, which are reported to pass through bentonite clay at the same rate as other mobile leachate constituents like chloride (Li et al., 2015). PFAS profile, leachate quality, and soil characteristics all play a role in soil interaction, and decisions should be made on a site-specific basis (Li et al., 2019; Gates et al., 2020; Mukhopadhyay et al., 2021).

Management of leachate in the US is dependent on climate – in dry regions, leachate generation is minimal, and many facilities use atmospheric evaporation. In contrast, in wet regions, leachate management presents a significant challenge (US EPA, 2021c). A nationwide survey found approximately 60% of US Subtitle D landfills conveyed their leachate to WWTPs for off-site treatment, 28% recirculated leachate or use other techniques resulting in no necessary leachate treatment, and 12% used on-site treatment (US EPA, 2021c). A breakdown of on-site leachate treatment strategies is included in SI ***Fig. S3***. Traditional leachate treatment typically targets non-PFAS leachate constituents of concern, such as ammonia and chemical oxygen demand (COD). The fate of PFAS in existing leachate treatment systems and wastewater treatment systems that manage leachate have been the subject of several studies and have been reviewed previously (Appleman et al., 2014; Lu et al., 2023; Meegoda et al., 2020; Travar et al., 2020; Zhang et al., 2022). To generalize, the treatment of ammonia and COD relies on chemical or biological oxidation, which do not effectively treat PFAS but often have the unintended effect of transforming precursor PFAS to terminal PFAS (US EPA, 2021b). Furthermore, during treatment, PFAS may partition into solids (e.g., biosolids) to a limited extent, which results in additional management challenges (Thompson et al., 2023b). Studies have recommended PFAS removal prior to such treatment (Lott et al., 2023). The targeted treatment of PFAS via removal or destruction in landfill leachate has been the subject of multiple reviews (Bandala et al., 2021; Berg et al., 2022; Lu et al., 2023; Ross et al., 2018; Travar et al., 2020) which have thoroughly discussed the effectiveness of different technologies and which are, here, summarized in Table 3.

PFAS separation technologies typically rely on adsorption over materials, such as activated carbons and ion exchange resins (US EPA, 2022c; Chow et al., 2022; Crone et al., 2019; Appleman et al., 2013), the use of high-pressure membrane separation (US EPA, 2022c; Lipp et al., 2010; Steinle-Darling and Reinhard, 2008), and newer technologies such as ozo- and foam-fractionation with the aim of concentrating the PFAS into a smaller volume of either a solid phase or concentrated liquid residual to either be disposed or destroyed via a subsequent high-energy destructive treatment method (Du et al., 2021; Labiadh et al., 2016). Several novel technologies are being investigated for the destructive treatment of landfill leachate – most require large amounts of energy in the form of chemical reactions or localized high temperatures to break the C—F bond. MSWI for energy recovery is not currently optimized to target PFAS destruction. Additional research is ongoing to define the conditions needed for PFAS destruction in MSWI and other incineration approaches, such as sewage sludge incineration.

Flaring and combustion are common LFG management techniques. Flaring is typically carried out in an open (candle) or enclosed flare. Combustion processes can generate energy on-site (e.g., a combustion engine) or off-site in a gas-fired power generation system. MSW LFG regulations target the destruction of nonmethane organic compounds (NMOCs), not PFAS. Flares generally operate at ~650 °C to 850 °C and temperatures in combustion engines or boiler systems could be lower (Wade, 2022). PFAS separation treatment has not been applied to LFG, however, laboratory-scale thermal PFAS destruction experiments indicate that temperatures higher than 1000 °C are necessary to achieve the mineralization of PFAS (Winchell et al., 2021). MSW LFG flare temperatures and the time that gaseous PFAS are in the presence of high

Table 3
Summary of treatment methods for PFAS in landfill leachate (Bandala et al., 2021; Berg et al., 2022; Lu et al., 2023; Ross et al., 2018; Travar et al., 2020; Wei et al., 2019).

	Technology	Pros	Cons	Matrix	References
Separation technologies	Activated carbon (GAC, PAC)	• High maturity level • Highly effective for long-chain PFAS	•Generates large quantities of spent sorbent that need additional treatment and disposal	GW	Busch et al. (2010); McCleaf et al. (2017), Pan et al. (2016), Ross et al. (2018); Bao et al. (2014), Pan et al. (2016); Malovanyy et al. (2023)
	Ion exchange resins	• High maturity level • Can remove compounds such as GenX	•Needs secondary treatment and disposal	GW, LL	Gao et al. (2015); Dixit et al. (2021); McCleaf et al. (2017); Ross et al. (2018); Boyer et al. (2021); Park et al. (2020); Ellis et al. (2022); Malovanyy et al. (2023)
	Membranes (RO, UF, NF)	• High maturity level and commonly practiced • 2-stage RO most effective on raw leachate	• Membrane fouling • Secondary stream with high PFAS concentrations and volume requires treatment • UF might not be effective	GW, LL	Das and Ronen (2022); Enzminger et al. (1987); Wei et al. (2019); Ross et al. (2018); Boo et al. (2018); Malovanyy et al. (2023)
	Foam/ ozofractionation	• High maturity level and commercially available pilot-scale technology • Potentially low cost	• Pretreatment of leachate might be required • Secondary treatment of concentrated PFAS required	GW, AFFF, LL	Smith et al. (2022); Robey et al. (2020); Malovanyy et al. (2023)
	Incineration	• Highly effective method • Can be used for regeneration of spent materials	•As a standalone method, not practical for large volumes of leachate		
Destruction technologies	Chemical Oxidation	•Controllable by varying pH and temperatures	• Uses additional chemicals for treatment • Low effectiveness of removal • Needs to be paired with other methods such as UV for higher effectiveness	LL	Abu Amr et al. (2013); Lin et al. (2012)
	Electrochemical	• 98–99.7% effectiveness demonstrated • Operates at ambient temperatures • No chemicals required • Lower energy consumption compared to incineration	• Expensive electrode materials • Perchlorates could be formed	LL	Labiadh et al. (2016); Du et al. (2021); Gomez-Ruiz et al. (2017); Witt et al. (2020); Krause et al. (2021)
	Photocatalysis	• 94–99% degradation reported • Can also potentially mineralize PFAS	• Slow kinetics • Lab-scale testing only • Difficult to scale for larger volumes		Esfahani et al. (2022)
	Sonolysis	• Can destroy short-chain and long-chain molecules • Effective for high concentration samples • Can be combined with chemical oxidation to lower costs	•High capital costs		Moriwaki et al. (2005); Vecitis et al. (2008); Babu et al. (2016)
	Microwaves	• Can be used to regenerate GACs • Catalytic microwave treatment could result in ~65–67% effectiveness	•Expensive for large-scale use		Gagliano et al. (2021); Lee et al. (2010b); Liu et al. (2020)
	Subcritical water oxidation	•Effective for short-chain PFAS	• Additional chemicals (e.g., zero-valent iron) needed for higher effectiveness • Slower kinetics		Hori et al. (2006)
	Supercritical water oxidation	• High maturity and close to commercialization • Low residence times required	• Full fluorine balance needed • High-pressure and temperature processes can be energy intensive	GW, AFF, LL	Pinkard et al. (2021); Hori et al. (2006); Krause et al. (2022)
	Wet Air Oxidation	•No demonstrated benefits for PFAS treatment	Converts FTOH precursors to PFCAs	LL	Travar et al. (2020)
	Biological processes	•Limited aerobic and anaerobic degradation of PFOS by bacteria and fungi reported	• Laboratory demonstrations only and thus low technology readiness level • Slow kinetics • Longer-chain PFAS converted to shorter-chain; no mineralization		Berhanu et al. (2023); Huang and Jaffé (2019)
	Constructed wetlands	•No demonstrated benefits for PFAS treatment	• Unlikely to be effective • Does not result in a concentrated PFAS stream that can be adequately managed • Environmental release of PFAS	LL	Yin et al. (2017, 2019); Awad et al. (2022); Lott et al. (2023)

(GAC = granular activated carbon; PAC = powder activated carbon; RO = reverse osmosis; UF = ultrafiltrations; NF = nanofiltration; GW = groundwater; LL = landfill leachate; AFFF = aqueous film-forming foam).

temperatures are too low to completely mineralize PFAS, but may result in the transformation of volatile PFAS into products of incomplete combustion (PICs). Notably, several PICs have been identified as significant greenhouse gases (Ahmed et al., 2020; Hong et al., 2013; Longendyke et al., 2022).

4. Environmental impact of PFAS emissions from landfills

Waste-derived PFAS may be emitted from landfills through multiple pathways, primarily in leachate or LFG effluent. While most RCRA-compliant landfills are operated to minimize environmental impacts, controls have yet to be designed to manage PFAS, and there is a subclass of small landfills in the US that are not required to install bottom liners as they are exempt from RCRA requirements (40 CFR § 258.1(f)(1)).

PFAS may be released into the atmosphere via fugitive gas emissions or gas flares. No data were found on PFAS concentrations in the ambient air surrounding C&D landfills, hazardous waste landfills, or industrial landfills; however, PFAS concentrations in the ambient air close to MSW landfills have been the subject of studies in the US, Germany, and China. Ahrens et al. (2011) reported average total FTOH concentrations of 2.6 and 26 ng m^{-3} at two US MSW landfills, representing 93% and 98% of total gas phase PFAS, with the remaining fraction consisting of perfluoroalkane sulfonamide (FASAs), perfluoroalkane sulfonamido ethanols (FASEs), and PFAAs. Weinberg et al. (2011) reported average total FTOH concentrations at two German landfill sites of 0.086 and 0.271 ng m^{-3}, representing 80% and 92% of total gas phase PFAS. Tian et al. (2018) measured PFAS in air sampled on-site at two landfills as well as downwind. The PFAS profile of the on-site air samples was more evenly split among classes. Total FTOHs were 0.61 and 2.1 ng m^{-3} at the two sites, representing 42% and 76% of $\sum_6$PFAS, with PFAAs representing the bulk of the remaining fraction. PFAS concentrations downwind of the two landfill sites were lower than on-site but elevated relative to control sites, indicating atmospheric transport of PFAS. Lower concentrations downwind may indicate dilution or deposition of volatile PFAS. Neutral PFAS readily transform in the environment – studies have shown the degradation of FTOHs into PFCAs via photooxidation (Esfahani et al., 2022; Martin et al., 2006). Tian et al. (2018) reported elevated neutral and ionizable PFAS in dry deposition samples on-site and downwind of landfills, driven primarily by PFBA and PFOA. Ahmadireskety et al. (2020) reported PFAS concentrations in landfill cover soils of approximately 8 μg kg^{-1}, similarly driven by PFCAs.

Deposition of PFAS from landfill-impacted air may also contribute to PFAS measured in surface water on landfill sites. Chen et al. (2023) reported $\sum_{26}$PFAS concentrations in stormwater at MSW landfill sites averaging 470 ng L^{-1}, significantly lower than leachate concentrations from the same study but significantly higher than groundwater samples, which averaged 140 ng L^{-1} of $\sum_{26}$PFAS. PFAS may be present in both surface and groundwater due to leachate contamination. At the same time, particulate transport from the working face or atmospheric transport and deposition of PFAS are more likely to impact surface water. The MPCA (2010) reported PFAS contamination in groundwater impacted by landfills accepting PFAS-laden industrial waste. Hepburn et al. (2019) measured PFAS and other landfill leachate indicators in groundwater impacted by legacy landfills in Australia, where PFOA represented >10% of total PFAAs, likely associated with legacy landfills.

Using the landfill liner collection efficiency reported by Jain et al. (2023) and overall leachate leakage rate of 1.9% with the leachate generation rate reported in Lang et al. (2017) (61 billion L $year^{-1}$), approximately 1.2 billion L of MSW landfill leachate enter the groundwater directly as a result of liner imperfections every year (14.3 kg of total PFAS using the average $\sum_{19}$PFAS from Lang et al. (2017)). This represents a conservative estimate, as Lang et al. (2017) note that most but not all landfills contributing to the total estimated leachate generation are lined. Although C&D leachate generation rates are not readily available, using leachate generation rates calculated for 17 MSW landfills in six US states, Jain et al. (2023) reported an average collection rate of 6900 L ha^{-1} day^{-1}. Assuming similar leachate generation rates for C&D landfills, this corresponds to approximately 2.5 million L of C&D leachate entering the groundwater per hectare of C&D landfill annually, representing a $\sum$PFAS mass of 26 g of PFAS per hectare of C&D landfill (see Table S9 in the SI for more calculation information). In 2012, the US EPA inventoried 1504 active C&D landfills (US EPA, 2012).

In the US, most landfill leachate generated from RCRA-permitted landfills is managed off-site (again, many C&D landfills are not required to collect leachate and thus operate without a bottom liner). This represents a significant flux of PFAS leaving the landfill. Multiple studies in the US and Australia have estimated the contribution of PFAS to municipal WWTP from landfill leachate and the environmental impact of PFAS in WWTP effluent. Masoner et al. (2020) estimated the PFAS load in landfill leachates and receiving WWTPs. They reported that landfill leachate while representing, on average, <2% of WWTP influent by volume across three sites, contributed 18% of influent PFAS. Gallen et al. (2017) reported similar contributions of PFAS to WWTPs from landfill leachates. PFAS are not effectively treated with traditional WWTP processes and are released to the environment via WWTP liquid effluent, land-applied biosolids, landfills, and possibly incineration of biosludge (Barisci and Suri, 2021; Coggan et al., 2019; Gallen et al., 2018; Helmer et al., 2022; Tavasoli et al., 2021).

5. Estimate of US MSW landfill PFAS mass balance

Estabrooks and Zemba (2019) evaluated landfill PFAS mass balance at an MSW landfill in Vermont, identifying the PFAS load from targeted waste types suspected to contain PFAS, not including residential MSW, and found that approximately 7% of the PFAS load entering landfills is emitted via leachate annually, and hypothesize the majority of PFAS remain in the waste mass within the landfill. Coffin et al. (2022) propose an estimated extractable $\sum$PFAS load in MSW entering landfills of 50 ng g^{-1} based on the findings in Liu et al. (2022a). This, combined with US EPA estimation of landfilled MSW in 2018 (the most recent year for which MSW generation data is available for the US), corresponds to 6600 kg of extractable PFAS entering MSW landfills in 2018 with MSW (US EPA, 2020b). Biosludge and biosolids also contribute a significant fraction of PFAS loading in MSW landfills. Using $\sum_{92}$PFAS in treated biosolids reported by Thompson et al. (2023b) and biosolids management statistics reported by NEBRA (2022), the 1.74 million dry metric tons of biosolids landfilled each year contribute an additional estimated 850 kg of PFAS to MSW landfills. Based on our calculations, a conservative estimate of 7480 kg of extractable PFAS entered US MSW landfills in 2018. This estimate does not include PFAS polymers.

As described earlier, PFAS can be emitted from landfills via the gaseous and liquid phase. MSW landfills in the US collect approximately 93.5 million m^3 of gas daily according to the US EPA LMOP database. This translates to nearly 1 kg of neutral PFAS emitted via MSW LFG per day (347 kg annually) based on the concentrations reported by Titaley et al. (2023). The US EPA estimates MSW LFG collection efficiency of approximately 75% (US EPA, 2020a), indicating an additional 31.2 million m^3 of LFG are released via fugitive emissions from MSW landfills annually. Leachate generation in the US, estimated by Lang et al. (2017), is 61.1 billion L $year^{-1}$ which corresponds to 750 kg of PFAS emitted from MSW landfills via leachate annually (using the weighted average $\sum$PFAS concentration of 12,300 ng L^{-1} calculated in this study). See Fig. 3 for a flowchart representing PFAS sources, controlled emissions, and uncontrolled emissions to the environment corresponding to MSW landfills, and Fig. 4 for a graphical presentation of the fraction of PFAS entering landfills from MSW and biosolids and corresponding emissions; the majority of PFAS entering landfills remain in the waste (84% annually) and significant a mass of PFAS have likely accumulated since PFAS use in consumer products began. Detailed calculations for Fig. 3 are included in the SI ***Table S10***.

One can estimate the total PFAS released via the gaseous phase per ton of MSW based on the potential methane generation capacity (L_o) of

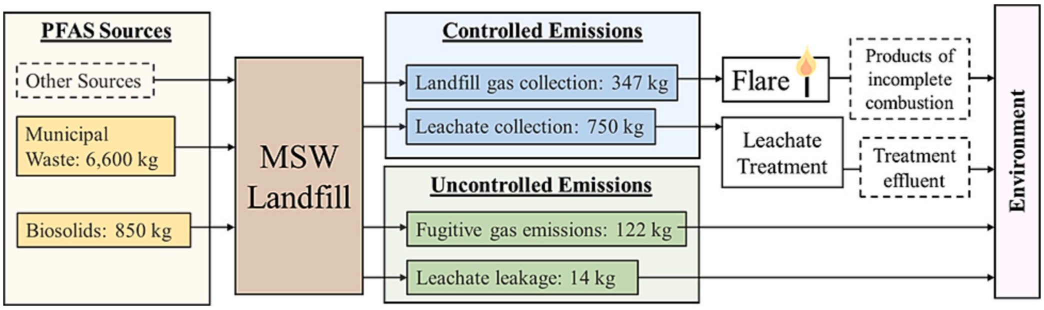

Fig. 3. Flowchart depiction of annual ∑PFAS loading and release at MSW and C&D debris landfills based on current understanding in the literature. Dashed lines represent PFAS streams which have not been quantified to any extent in the literature.

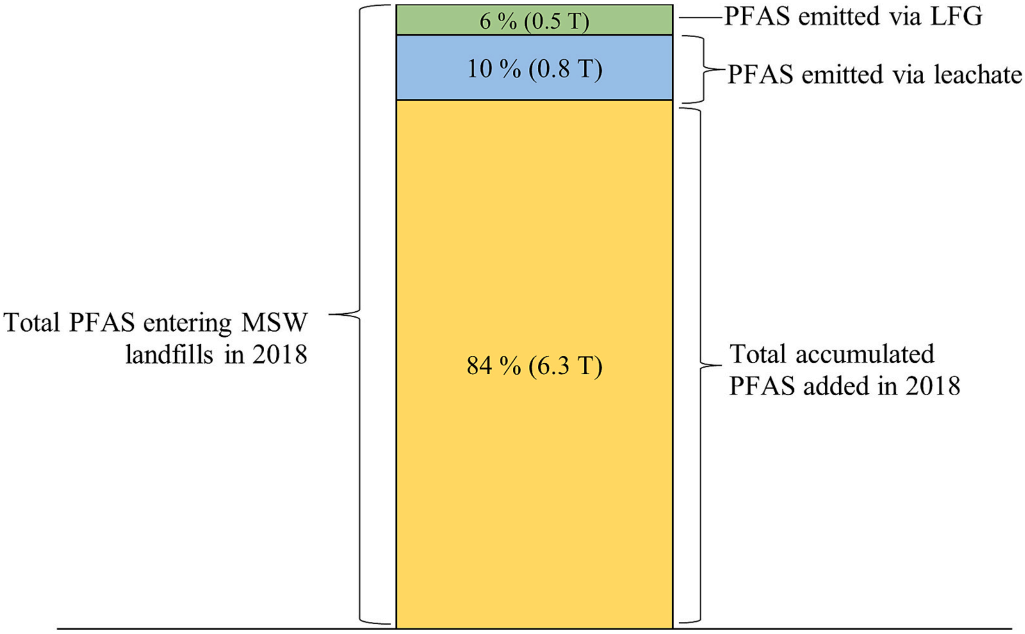

Fig. 4. Estimated PFAS mass balance for US MSW landfills.

MSW. Jain et al. (2021) estimated MSW methane emissions of 68 m^3 of methane per metric ton (Mg) of waste, or approximately 136 m^3 of LFG per Mg of waste; using these values and the Titaley et al. (2023) LFG PFAS concentrations suggest 1.38 mg of PFAS are released, cumulatively, via LFG for every Mg of MSW.

5.1. Limitations

The estimated PFAS mass loading and emissions presented here are based on multiple assumptions and, in some cases, limited data, resulting in significant uncertainty. We have not provided additional data quality assurance in this review process. A small number of studies have explored changes in landfill leachate PFAS profile over time, and no studies have looked for similar relationships in LFG; for this critical review, it was assumed that ∑PFAS reported in leachate and gas are representative of a range of waste ages and stages of decomposition and, overall, are expected to remain consistent over time. Even fewer studies have looked at C&D debris landfills in the US, and those studies are limited to Florida landfills. This critical review of previous analyses provides perspective, not precise values, which should be derived through additional empirical studies.

6. Conclusions and data gaps

The bulk of studies of PFAS in solid waste and landfills focus on MSW landfill leachate, with comparatively fewer studies estimating overall PFAS loading in other types of landfill leachate, in the solid waste itself, or gaseous effluent. Regardless of the type of landfill, in all studies across all locations, PFAS were quantified in all leachate samples. PFAS concentrations in leachates vary across studies, which may be a function of waste type, leachate qualities, climate, and the analytical method.

US MSW and C&D landfill leachates have similar ∑PFAS concentrations. However, C&D leachate contains proportionally more terminal PFAS. This is likely due to the PFAS present and the conditions within each landfill type. Concentrations of the five PFAS which have been the subject of proposed US EPA regulations (i.e., PFBS, PFHxS, PFOS, PFOA, and PFNA) consistently exceed US EPA tapwater RSLs in both MSW and C&D landfill leachates by a factor as high as 20 (PFOA) and in HW landfill leachates by a factor as high as 104 (PFOS); as presented in Table 1 and Fig. 2.

MSWI ash leachates have lower PFAS concentrations than other leachates, however, co-disposal of ash with other wastes results in disproportionately high PFAS concentrations in leachate. To minimize PFAS leaching from MSWI ash landfills, care should be taken to dispose of unburned waste which contains higher concentrations of PFAS separately from MSWI ash. No peer-reiviewed studies have reported

PFAS concentrations in effluent from hazardous waste (Subtitle C) landfills, although hazardous waste management methods are likely to reduce PFAS leaching. Since some hazardous waste landfills likely accept PFAS-laden industrial waste at relatively high levels, it would be helpful to have more information on effluent generated from these facilities.

Traditional leachate treatment methods that use oxidation (e.g., a treatment that targets ammonia, COD) are likely to increase the transformation rate of precursor PFAS to terminal PFAS, such as regulated PFAAs. Treatment that relies on volatilization, such as evaporation, likely contributes significant quantities of PFAS to the atmosphere and surrounding environment, increasing off-site transport. Separating PFAS from leachate prior to additional treatment would avoid these issues. Though there are many aqueous treatment technologies for the targeted removal or destruction of PFAS, few have been tested for effectiveness on landfill leachate. Those tested on leachate and have shown promise include supercritical water oxidation, electrochemical oxidation, reverse osmosis, and foam separation. Assuming treatment efficacy is comparable across PFAAs, reducing PFOA concentration to its limit (e.g., MCL) will reduce all other PFAS to below their respective limits. PFAS treatment of liquid wastes often produces a secondary residual waste requiring additional management.

PFAS are expected to be present in LFG as a product of volatilization and the anaerobic decomposition of biodegradable waste but have been quantified only in MSW LFG. PFAS have not been measured in C&D LFG. However, based on PFAS profiles in C&D landfill leachate, similar PFAS concentrations are likely present in C&D LFG, although LFG generation rates from C&D debris is lower. To reduce gaseous emissions of PFAS, biodegradable waste should be disposed of separately from other PFAS-containing waste. Data do not exist on the effectiveness of PFAS destruction from LFG combustion within flares and internal combustion engines or PFAS removal from LFG to RNG conversion processes. However, the temperatures reached in LFG flares are expected to transform volatile PFAS into terminal PFAS and possible PICs, with minimal mineralization.

Based on our estimate of the PFAS entering and leaving landfills, significant quantities of PFAS are emitted in both LFG and leachate; however, the bulk of PFAS remains within the waste mass on a per-year basis (see Fig. 3). This suggests landfills will be a source of PFAS emissions for the foreseeable future. Studies have demonstrated down-gradient impacts on groundwater from landfills. C&D landfills pose the highest risk of environmental contamination since they are not required (at the federal level) to install liners to collect leachate. Even among lined landfills, the average liner collection efficiency is approximately 98%, corresponding to an annual flux of 14.3 kg PFAS entering groundwater via liner imperfections.

Elevated PFAS concentrations were measured in ambient air at landfills across several studies. The highest concentrations were found among FTOHs, which transform into PFAAs in the environment. Atmospheric PFAS may deposit and contribute to soil and surface water concentrations. Even if LFG collection systems were equipped to operate at temperatures and residence times sufficient to destroy PFAS, current MSW LFG collection efficiency is only 75%, and landfills not required to collect LFG will continue to emit PFAS into the atmosphere. The fate of PFAS in LFG that passes through landfill cover soil should be analyzed in future studies.

This review has identified several data gaps for PFAS emissions from US landfills. Data are needed from hazardous waste landfill sites and relevant industrial waste landfills. Furthermore, US C&D landfill leachate data are limited to Florida landfills, and additional efforts should be made to collect information from other states. C&D waste streams may vary due to regional construction requirements. The measurement of PFAS in LFG and other gaseous emissions is an area of emerging study. More research is needed on both controlled and uncontrolled landfill gaseous emissions. A closer evaluation of the fate of PFAS during leachate treatment and LFG management is needed to help decision-makers guide the solid waste community. Geomembrane liners are the most effective tools for the protection from and collection of PFAS-containing liquids, such as landfill leachate. More research is needed to understand long-term interactions between PFAS and liner systems, especially in complex matrices such as landfill leachate. More research is needed to evaluate the long-term implications of PFAS in the landfill environment since the bulk of PFAS remains within the solid waste mass. This review focused on landfilling as a management option for solid waste; evaluation of PFAS fate during other solid waste management processes (e.g., anaerobic digestion, thermal treatment, composting, and recycling) is needed.

CRediT authorship contribution statement

Thabet Tolaymat: conceptualization, approach, methodology, writing-reviewing, editing, funding, **Nicole Robey**: data curation, methodology, calculations, writing-original draft, writing and editing. **Max Krause**: writing and editing, **Judd Larson, Keith Weitz, Sameer Parvathikar**: data curation, original draft preparation, editing, **Lara Phelps, William Linak, Susan Burden, Tom Speth, Jonathan Krug:** writing and editing

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167185.

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Review of the fate and transformation of per- and polyfluoroalkyl substances (PFASs) in landfills☆

Hanna Hamid [a], Loretta Y. Li [a, *], John R. Grace [b]

[a] Civil Engineering, University of British Columbia, 6250 Applied Science Lane, Vancouver, BC, V6T 1Z4, Canada
[b] Chemical and Biological Engineering, University of British Columbia, 2360 East Mall, Vancouver, BC, V6T 1Z3, Canada

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ABSTRACT

A critical review of existing publications is presented i) to summarize the occurrence of various classes of per- and polyfluoroalkyl substances (PFASs) and their sources in landfills, ii) to identify temporal and geographical trends of PFASs in landfills; iii) to delineate the factors affecting PFASs in landfills; and iv) to identify research gaps and future research directions. Studies have shown that perfluoroalkyl acids (PFAAs) are routinely detected in landfill leachate, with short chain (C4-C7) PFAAs being most abundant, possibly indicating their greater mobility, and reflecting the industrial shift towards shorter-chain compounds. Despite its restricted use, perfluorooctanoic acid (PFOA) remains one of the most abundant PFAAs in landfill leachates. Recent studies have also documented the presence of PFAA-precursors (e.g., saturated and unsaturated fluorotelomer carboxylic acids) in landfill leachates at concentrations comparable to, or higher than, the most frequently detected PFAAs. Landfill ambient air also contains elevated concentrations of PFASs, primarily semi-volatile precursors (e.g., fluorotelomer alcohols) compared to upwind control sites, suggesting that landfills are potential sources of atmospheric PFASs. The fate of PFASs inside landfills is controlled by a combination of biological and abiotic processes, with biodegradation releasing most of the PFASs from landfilled waste to leachate. Biodegradation in simulated anaerobic reactors has been found to be closely related to the methanogenic phase. The methane-yielding stage also results in higher pH (>7) of leachates, correlated with higher mobility of PFAAs. Little information exists regarding PFAA-precursors in landfills. To avoid significant underestimation of the total PFAS released from landfills, PFAA-precursors and their degradation products should be determined in future studies. Owing to the semi-volatile nature of some precursor compounds and their degradation products, future studies also need to include landfill gas to clarify degradation pathways and the overall fate of PFASs.

1. Introduction

Landfilling is one of the most common disposal methods for end-of-life consumer products (Renou et al., 2008). Engineered landfills are designed to contain solid waste and collect landfill leachate, while preventing migration of the contaminants to groundwater. Among the emerging contaminants, per- and polyfluoroalkyl substances (PFASs), detected in landfill leachate, are receiving attention due to their persistence, bioaccumulation potential and adverse effects on biota and humans (Houde et al., 2011). PFASs are a diverse group of aliphatic compounds containing one or more perfluoroalkyl moiety (C_nF_{2n+1}-). PFASs containing at least one perfluoroalkyl moiety are called polyfluoroalkyl substances (e.g., $CF_3CF_2CH_2COOH$). Perfluorinated substances are defined as aliphatic substances for which all of the H atoms attached to C atoms in the nonfluorinated substance from which they are notionally derived have been replaced by F atoms, except the H atoms present in any functional groups (Fig. S1 in supplemental information (SI)) (Buck et al., 2011).

Due to their unique surface-active properties and high chemical and thermal stability (Buck et al., 2011), PFASs are widely used in numerous consumer products (e.g. textiles, paper, non-stick cookware, carpets, cleaning agents) and industrial applications (e.g., metal plating, fire-fighting foams, electronics production,

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* Corresponding author.
E-mail address: lli@civil.ubc.ca (L.Y. Li).

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Abbreviations

DiPAP	Disubstituted fluorotelomer phosphate esters
EtFOSAA	Ethyl-perfluorooctane sulfonamide acetic acid
EtFOSE	Ethyl-perfluorooctane sulfonamidoethanol
FASA	Perfluoroalkane and *N*-alkyl perfluoroalkane sulfonamide acetic acid
FOSA	Perfluoroalkane and *N*-alkyl perfluoroalkane sulfonamide
FOSE	Perfluoroalkane and *N*-alkyl perfluoroalkane sulfonamidoethanols
FTCA	Fluorotelomer saturated carboxylic acid
FTI	Fluorotelomer iodide
FTOH	Fluorotelomer alcohol
FTP	Fluorotelomer polymer
FTSA	Fluorotelomer sulfonate
FTUCA	Fluorotelomer unsaturated carboxylic acid
MeFBSAA	Methyl-perfluorobutane sulfonamide acetic acid
MSW	Municipal solid waste
NF	Nanofiltration
PAP	Polyfluorinated phosphate ester
PEPE	Perfluoropolyether
PFAA	Perfluoroalkyl acid
PFAS	Per- and polyfluoroalkyl substance
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid
PFCA	Perfluoroalkyl carboxylic acid
PFDA	Perfluorodecanoic acid
PFHpA	Perfluoroheptanoic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonic acid
PFNA	Perfluorononaic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPA	Perfluoroalkyl phosphonic acids
PFPeA	Perfluoropentaonoic acid
PFPiA	Perfluoroalkyl phosphinic acids
PFPrA	Pentafluoropropionic acid
PFSA	Perfluoroalkyl sulfonic acid
POP	Persistent organic pollutant
POSF	Perfluorooctane sulfonyl fluoride
RO	Reverse osmosis
SI	Supplemental Information
TOC	Total organic carbon
UF	Ultrafiltration
WWTP	Wastewater treatment plant

photography) (Arvaniti et al., 2014; Kissa, 2001). Among the most commonly detected perfluoroalkyl acids (PFAAs) in the environment, pefluorooctane sulfonate (PFOS) has been listed under Annex B of the Stockholm Convention on persistent organic pollutants (POPs) since 2009, restricting its production and use, except for a few exemptions; perfluorooctanoic acid (PFOA) is currently under review by the POPs Review Committee of the Stockholm Convention (Stockholm Convention, 2017). While PFAAs may be directly released into the environment during production, usage and disposal, polyfluoroalkyl substances – the "PFAA-precursors" – can also be transformed abiotically or biologically into PFAAs (see Fig. S1 in SI).

A variety of consumer products (e.g., paper, textiles, carpets) and packaging containing PFAAs and their precursors are sent to municipal landfills at the end of their useful lives. In many municipalities, biosolids containing PFASs are also landfilled (Guerra et al., 2014; Arvaniti et al., 2012). Following disposal, PFASs are released from the waste through biological and abiotic leaching (e.g., desorption) (Allred et al., 2015), as shown in Fig. 1. Depending on their physio-chemical properties, some anionic, water soluble PFASs (e.g., PFAAs) can be released with the landfill leachate (Yan et al., 2015; Benskin et al., 2012); on the other hand, neutral PFASs with low water solubilities and relatively high vapor pressures (e.g., fluorotelomer alcohols (FTOHs)) partition with landfill gas and are subsequently released to the atmosphere, if not captured efficiently by a gas collection system (Fig. 1). Most often, leachate from lined landfills are collected and sent to wastewater treatment plants (WWTPs) for treatment before their final disposal in surface water bodies. However, WWTPs, already burdened with PFAS from wastewater, are not equipped to remove these classes of contaminants, instead are acting as secondary sources of PFASs in the aquatic environment (Allred et al., 2015; Eggen et al., 2010). Given that solid wastes have been, and will continue to be, landfilled, it is critical to investigate landfills as long-term point sources of PFASs in the environment.

As more and more studies are published regarding environmental occurrence, fate and degradation of PFASs, it is important to systematically review the published literature to critically evaluate the state of knowledge and identify research gaps. Recent reviews of PFASs have addressed environmental biodegradation (Liu and Avendano, 2013), fate and removal of PFASs in drinking water treatment plants (Rahman et al., 2014), and WWTPs (Merino et al., 2016; Arvaniti and Stasinakis, 2015). A comprehensive review on the fate and transformation of PFASs in landfills is needed. This study critically reviews existing publications i) to summarize the occurrence of various classes of per- and polyfluoroalkyl substances (PFASs) and their sources in landfills, ii) to identify temporal and geographical trends of PFASs in landfills; iii) to delineate factors affecting PFASs in landfill; and iv) to identify research gaps and key future research directions.

2. Methodology

Based on an online database search (Web of Science, ScienceDirect and Google Scholar) of peer-reviewed articles, 14 journal articles were identified that reported PFAS concentrations in landfill leachate. Two studies reporting PFAS concentrations in ambient landfill air, three investigating degradation and leaching of PFASs inside landfills and one reporting leaching of PFASs through sodium bentonite (landfill barrier material) were also uncovered. While the subsequent sections in this paper are heavily based on these 18 articles (published between 2004 and 2017), additional citations from peer-reviewed journals are also cited to contextualize and explain the observations of the selected articles. The reported concentrations of PFASs are compiled in Table S1 (PFAAs) and S2 (perfluoroalkane sulfonamide derivatives and polyfluoroalkyl compounds) of SI. Concentration ranges and, where possible, median and other statistical values were calculated for studies reporting concentrations from multiple samples (from one or more landfills). During data analysis, below-quantification-limit values were assumed to be zero.

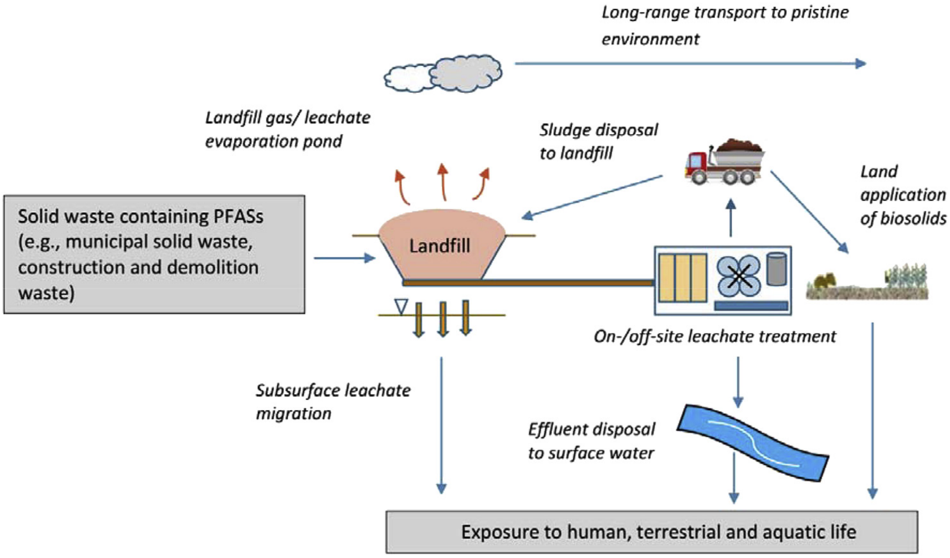

Fig. 1. Environmental pathways of per- and polyfluoroalkyl substances (PFASs) originating from solid wastes.

3. Per- and polyfluoroalkyl substances (PFASs) in landfills

3.1. Occurrence and sources of PFASs in leachate

3.1.1. Perfluoroalkyl acids (PFAAs)

Perfluoroalkyl carboxylic acids (PFCAs) and Perfluoroalkyl sulfonic acids (PFSAs), together known as PFAAs, are the most commonly studied PFASs in landfills. PFCAs with 4–14 carbon chain length and PFSA of mostly even chain length from C4-C10 have been reported in landfill leachate in the ng/L to µg/L range. Concentration ranges of PFAAs (C4-C10) in various countries are plotted in Fig. 2(a). Possible sources of PFAAs include consumer products (e.g., paper, textile, packaging, food contact paper, carpet) (Bečanová et al., 2016; Kotthoff et al., 2015; Ye et al., 2015), building materials (Bečanová et al., 2016), electronics (Bečanová et al., 2016) resulting from intentional addition of PFAAs during production and/or product use, and contamination with by-products impurities during production (Bečanová et al., 2016). Furthermore, PFAA precursors (e.g., FTOH, n:2 fluorotelomer carboxylic acids (n:2 FTCA) and n:2 unsaturated fluorotelomer carboxylic acids (n:2 FTUCAs)) present in the consumer products (Kotthoff et al., 2015; Ye et al., 2015) can degrade to PFAAs during product use and/or after disposal in the landfill (Allred et al., 2015; Lang et al., 2016).

3.1.2. Fluorotelomer-based substances

Fluorotelomer based compounds such as, n:2 FTCAs, n:2 FTUCAs, n:3 fluorotelomer carboxylic acids (n:3 FTCAs), fluorotelomer sulfonates (n:2 FTSAs) have been detected in landfill leachate (Lang et al., 2017; Allred et al., 2014; Benskin et al., 2012; Huset et al., 2011) and lab-scale landfill reactors (Lang et al., 2016; Allred et al., 2015) ranging from a few ng/L to µg/L (Fig. 2(b)). The FTCAs and FTUCAs are known degradation products of FTOHs (Buck et al., 2011), a major raw material of fluorotelomer polymers (FTPs), commonly used in textiles, upholstery, paper and carpets as surface protection agent (Rao and Baker, 1994). In addition to FTOH monomer released through biological (Washington et al., 2015; Rankin et al., 2014) and abiotic hydrolysis (Washington and Jenkins, 2015), residual FTOH present in FTPs (Dinglasan-Panlilio and Mabury, 2006) can biodegrade to FTCAs and FTUCAs, and subsequently to PFCAs in soil and activated sludge from WWTPs under both aerobic and anaerobic conditions (Liu and Avendano, 2013). The n:2 FTSAs can be released from consumer products applied with FTSA-containing surface protectors (Lang et al., 2016; Allred et al., 2015; Yang et al., 2014a), and by the degradation of complex fluorotelomer-based substances used in food packaging applications (Buck et al., 2011).

3.1.3. Perfluoroalkyl sulfonamide derivatives

Several unsubstituted, methyl- and ethyl-perfluoroalkane sulfonamide acetic acids (FASAAs) with C4-C8 carbon chain length have been reported in landfill leachates (Lang et al., 2017; Allred et al., 2014; Benskin et al., 2012; Huset et al., 2011) as shown in Fig. 2(b). Biodegradation of ethyl-perfluorooctane sulfonamidoethanol (EtFOSE), a major raw material of paper and packaging products (Buck et al., 2011), is said to form C8-based ethyl-perfluorooctane sulfonamide acetic acid (EtFOSAA) (Rhoads et al., 2008). Similar biodegradation pathways could be responsible for the shorter FASAA, MeFASAA and EtFASAA homologues resulting from methyl- and ethyl-perfluoroalkyl sulfonamidoethanols (FOSE) (Allred et al., 2014).

3.1.4. Polyfluoroalkyl phosphate esters (PAPs)

Detection of a few classes of polyfluoroalkyl phosphate esters (PAPs) (e.g., Di-substituted fluorotelomer phosphate esters (6:2–10:2 DiPAPs) and EtFOSE-based polyfluoroalkyl phosphate diester (DiSAmPAP)) have been reported in leachate (Allred et al., 2014; 2015, Lang et al., 2017) (see Table S2, in SI). PAPs are used in papers and synthetic fibers to impart oil and water repellency, in semiconductor materials and in personal care products (Liu and Liu, 2016). Microbial degradation of PAPs resulting in a mixture of FTCAs and PFCAs has been reported in activated sludge (Lee et al., 2010), and in aerobic soil (Liu and Liu, 2016; Lee et al., 2013), accounting for the infrequent detection of PAPs in leachate, despite their widespread use and high production volume (De Silva et al., 2012).

3.2. General observations and geographical trends of PFASs in leachate

Despite the high variabilities in PFAAs profiles and concentrations in landfill leachate reported in North America, Europe, China and Australia (Yan et al., 2015; Clarke et al., 2015; Allred et al., 2014; Bossi et al., 2008; Kallenborn et al., 2004), a few general trends emerge. For example, PFCAs are generally found to be the dominant

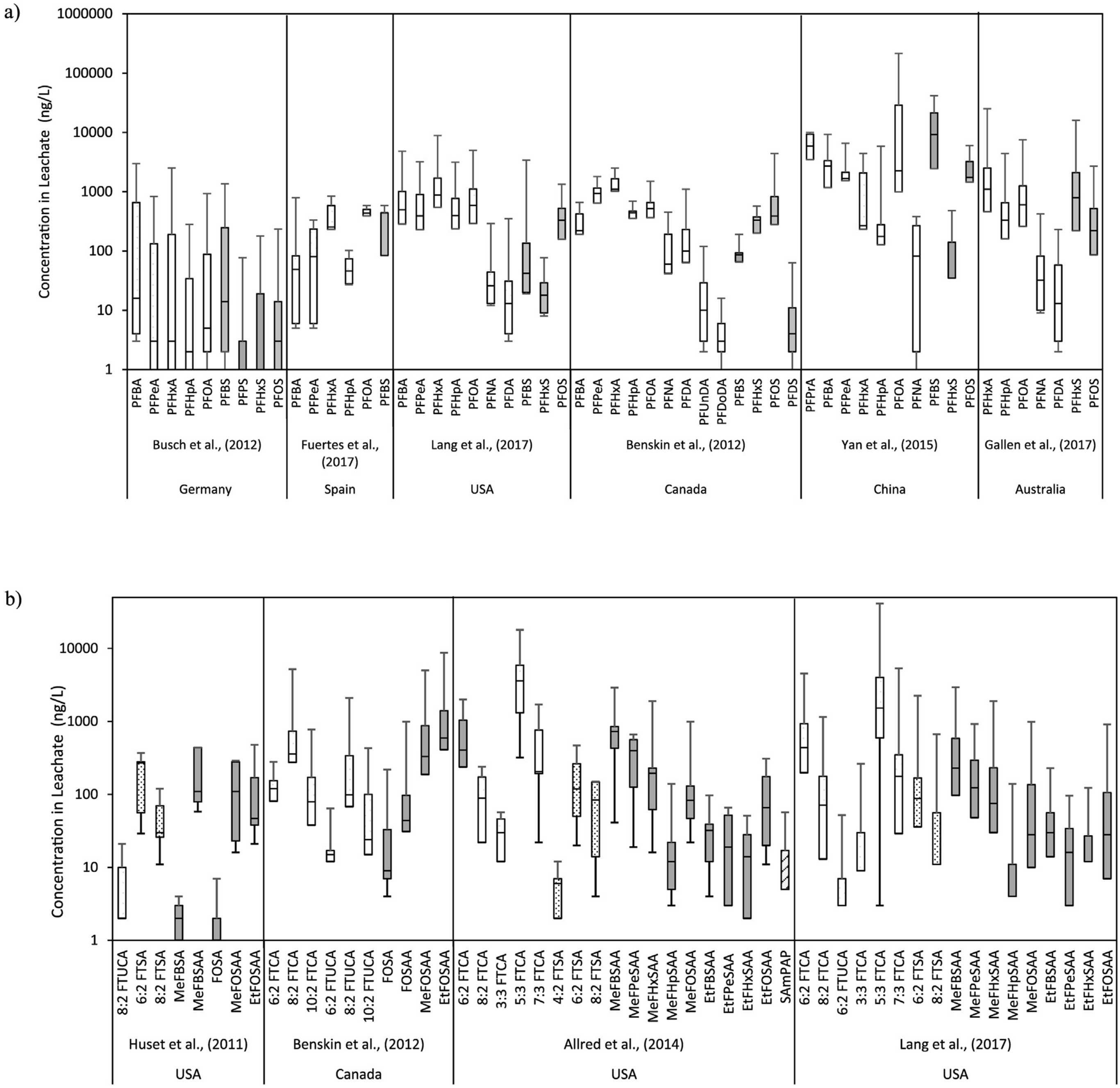

Fig. 2. Concentration ranges of (a) perfluoroalkyl carboxylic acids (PFCAs; plotted with white box) and perfluoroalkyl sulfonic acids (PFSAs; plotted with grey box); For countries with multiple studies, the study with highest number of landfill leachate samples are plotted in 2(a). Concentration ranges of (b) fluorotelomer carboxylic acids (plotted with white box), fluorotelomer sulfonic acids (black dotted box) and perfluoroalkane sulfonamide derivative substances (plotted with grey box) and polyfluorinated phosphate ester (plotted with slanted line) in landfill leachate in different countries. Analytes with ≥50% detection frequency has been plotted in 2(b). The box plots represent the 1st quartile, median and 3rd quartile values. The negative and positive error bars represent the difference between 1st quartile and minimum value, and maximum and 3rd quartile value, respectively. Note the logarithmic scale of Y-axis.
(Acronyms for 2(b): FTCA: fluorotelomer carboxylic acid, FTUCA: fluorotelomer unsaturated carboxylic acid, FTSA: fluorotelomer sulfonates, MeFASAA: methyl-perfluoroalkane sulfonamide acetic acid (C4-C8), EtFASAA: Ethyl-perfluoroalkane sulfonamide acetic acid (C4, C5, C6, C8), SAmPAP: 2 (*N*-ethylperfluorooctane-sulfonamido) ethyl phosphate).

PFASs (Fuertes et al., 2017; Allred et al., 2014; Li et al., 2012; Huset et al., 2011). Also, C4-C7 chain length PFAAs are more abundant than their longer-chain (≥C8) homologues (Fuertes et al., 2017; Li et al., 2012; Busch et al., 2010; Bossi et al., 2008; Kallenborn et al., 2004). Short-chain PFAAs are prone to preferential release and leaching from municipal solid waste (MSW), consistent with their higher aqueous solubilities and lower organic carbon-water partition coefficients relative to longer-chain PFAAs (Yan et al., 2015). In addition, the dominance of C4-C7 PFAAs could be related to the shift towards production of shorter-chain perfluorinated compounds since the early 2000's (Fig. 3). For example, 3M has commercialized surface treatment products containing C4-based side-chain fluorinated polymers since 2003 (Wang et al., 2013); 6:2 fluorotelomer-based side-chain fluorinated polymers have been registered in the Inventory of Effective Food Contact Substances Notifications of the United States Food and Drug

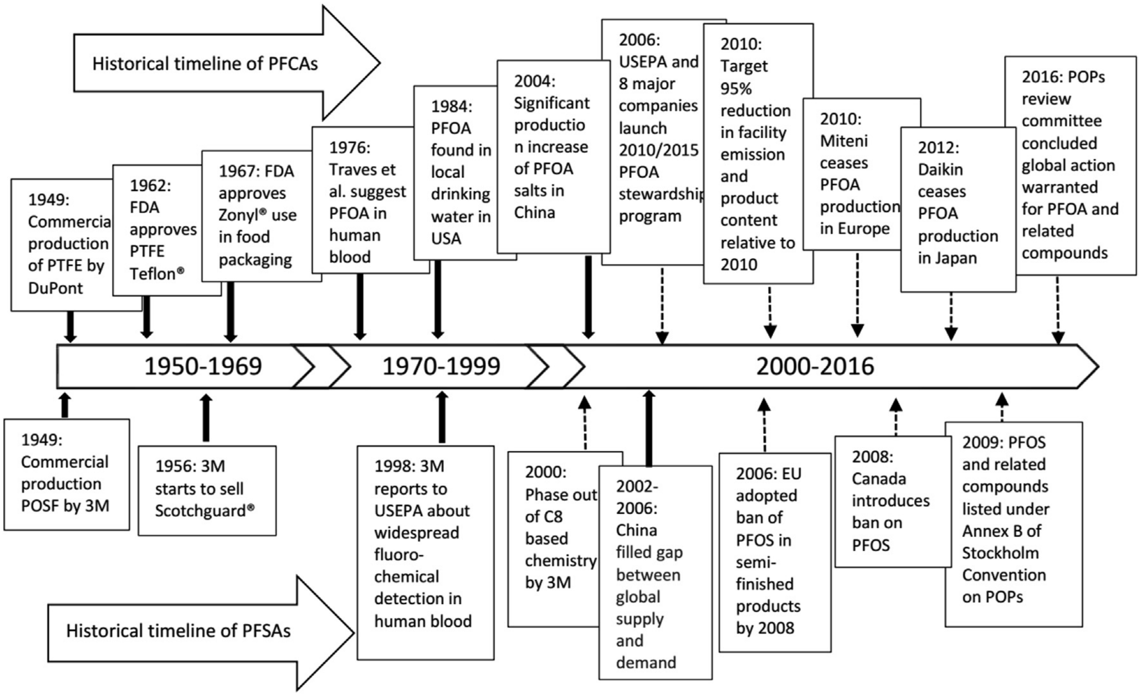

Fig. 3. Timeline of the production, commercialization and legislation of perfluoroalkyl carboxylic acids (PFCAs; at the top) and perfluoroalkyl sulfonic acids (PFSAs; at the bottom). Events and actions that may have resulted in increased concentrations in the environment and important findings are indicated by solid line arrow. Dotted arrows represent phase-outs and regulatory initiatives that may result in decreased concentrations in the environment. POSF (perfluorooctane sulfonyl fluoride) is the raw material of PFOS (perfluoroocatane sulfonic acid). Please note that the uneven time scale. Adapted from (Land et al., 2015; Lindstrom et al., 2011).

Administration since 2008 (Wang et al., 2013). Most of the landfill studies discussed here, involving sampling after 2010, likely reflect a product shift resulting from disposal of PFAS-containing consumer products with short residence life (e.g., food contact paper, packaging, other paper). For consumer products with longer residence times (e.g., carpet, upholstery, textiles), a time lag is expected before fluorinated alternatives used in these products reach measurable levels in leachate. The high frequency of PFOA detection in consumer products (Vestergren et al., 2015; Liu et al., 2014), together with its historical use in surface treatment agents of leather, textiles, paper and electronics (Wang et al., 2014a) explain the observed high detection frequencies and concentrations (often comparable to those of C4-C6 PFCAs) of PFOA in leachate. Studies have also reported that $\sum$PFAAs from landfill leachate (for a facility closed 2-4 decades ago), ranges from hundreds to a few thousands of ng/L (Gallen et al., 2016; Allred et al., 2014; Huset et al., 2011).

An increasing number of studies showing degradation of polyfluorinated compounds to PFAAs in the environment (Liu and Liu, 2016), along with increasing availability of chemical standards and improved analytical techniques, have led to recent studies (summarized in the next section) to investigate PFAA-precursors and their degradation products, as well as other classes of perfluorinated compounds (e.g., perfluoroalkyl sulfonamide derivatives) in landfill leachate. Some of the fluorotelomer-based (e.g., n:2 FTCA, FTUCAs) and *N*-alkyl FASAAs are frequently detected (Table S2) with concentration ranges (shown in Fig. 2(b)) that are comparable to and/or higher than those of PFCAs (>μg/L).

3.2.1. Concentration and trends in the USA and Canada

PFASs in landfill leachate in the USA have been studied by Huset et al. (2011), Allred et al. (2014) and Lang et al. (2017) (Table S1, in SI). PFCA contributed 20–90% of $\sum$PFASs (molar concentration basis), with concentrations from 10 to 8900 ng/L (Allred et al., 2014; Huset et al., 2011). While the median concentration reported by Allred et al. (2014) exceeded 1000 ng/L for all C4-C8 PFCAs, Huset et al. (2011) observed smaller concentrations (100–600 ng/L) for the same compounds. The difference in concentrations could result from variation in waste composition, climatic condition, age of the landfill and/or leachate management system (i.e., leachate recirculation vs flow-through), as indicated in Table 1 (see also section 3.5). PFSA concentrations in leachate have varied from 50 to 3200 ng/L in the USA, with median concentrations of a few hundred ng/L for perfluorobutane sulfonic acid (PFBS), perfluorohexane sulfonic acid (PFHxS) and PFOS (Allred et al., 2014; Huset et al., 2011), as shown in Fig. 2(a). While PFOS was detected in all leachate samples, its concentration was generally lower than that of PFBS and PFHxS (Allred et al., 2014; Huset et al., 2011). This dominance of shorter-chain PFSAs over historically used PFOS could be indicative of the transition towards C4-based chemistry after 2002, as shown in Fig. 3 (Vestergren et al., 2015; Huset et al., 2011; Lindstrom et al., 2011). Leachates from waste cells closed in 1993 or earlier also shows dominance of PFBS and PFHxS, indicating the role of C4-based chemistry even prior to 2002 (Wang et al., 2014b; Huset et al., 2011), in addition to the higher mobility of shorter chain PFSAs, leading to their release in aqueous phase (Higgins and Luthy, 2006).

PFCA concentrations in leachates collected from Canadian landfills have been in the range of tens to few thousands of ng/L (Li et al., 2012; Benskin et al., 2012). PFSAs, namely PFBS, PFHxS and PFOS, also varied within the same range, with median PFHxS concentration of 200 ng/L higher than for PFOS and PFBS in a cross-Canada study (Li et al., 2012). Landfill gas condensate was reported to contain C4-C8 PFAAs, with PFBS being the dominant compound at a concentration of 1000 ng/L (Li, 2011).

3.2.2. Concentration and trends in European Countries

PFAAs have been reported in several European countries, including Spain (Fuertes et al., 2017), Finland (Perkola and Sainio,

Table 1
Landfill locations and characteristics for leachate sampling sites.[a]

Reference	Woldegiorgis et al. (2006)	Busch et al. (2010)	Huset et al. (2011)	Benskin et al. (2012)	Perkola and Sainio (2013)	Allred et al. (2014)	Yan et al. (2015)	Gallen et al. (2016)	Gallen et al. (2017)	Lang et al. (2017)	Fuertes et al. (2017)
Location (Country)	Strandmossen, Djupdalen (Sweden)	(Germany)	Gulf Coast, Pacific Northwest, west coast, Mid-Atlantic states, Southeast (USA)	Pacific Northwest (Canada)	Espoo (Finland)	(USA)	Changzhou, Guangzhou, Nanjing, Shanghai, Suzhou (China)	(Australia)	(Australia)	Arid, temperate and wet climatic locations in USA	(Spain)
Landfill Sites	3	8 closed between 1979 and 2005, 14 active	4 active (since 1996), 2 closed (operated during 1982–1993)	2 active since 1960's	1 closed (1987–2007)	5 active (since 1990's), 1 closed (1975–1990)	4 active, 3 closed	6 active, 8 closed (operated between 1970's and 2010)	24 active, 3 closed (between 1993 and 2003)	18 active	2 active, 2 closed between 2015 and 2015
PFAS Analytes	13	43	24	24	4	70	14	14	9	70	16
Leachate System	not available	flow-through	recirculation, except one flow through system	flow-through and recirculated	not available	not available	flow through	flow through except one recirculated	recirculation and flow through system	recirculation	flow through
Sampling Year	November, 2005	not available	2006	February–June, 2010	October, 2009 and June, 2010	not available	Spring, 2013	February–April, 2014	August–December 2014	February 2013 to December 2014	March, 2015
Waste Type	not available	municipal and commercial	primarily municipal	primarily municipal	primarily municipal	municipal and commercial	municipal	primarily municipal and commercial	municipal, commercial, construction and demolition	primarily municipal	primarily municipal
Sampling Method	grab samples	not available	grab samples	grab sample	24-hr composite	grab samples	grab sample	grab sample	grab and composite samples	grab samples	grab sample
Leachate Treatment System	aerobic pond	biological and physical	not available	off-site at WWTP	not available	not available	off-site with two-stage process (MBR/RO or NF)	off-site at WWTP,	off-site WWTP, evaporation pond	Off-site WWTP	off-site with two-stage process (MBR/UF)
Estimated PFAS loading in leachate	not available	not available	not available	8-25 kg/y/landfill	not available	not available	3100 - 4000 kg/y (nationwide)	not available	not available	563 - 638 kg in 2013 (nationwide)	1 kg/y/landfill

[a] WWTP: wastewater treatment plant; MBR: membrane bioreactor; RO: reverse osmosis; UF: ultrafiltration; NF: nanofiltration; PFAS: per- and polyfluoroalkyl substance.

2013), Norway (Kallenborn et al., 2004), Sweden (Woldegiorgis et al., 2006), Denmark (Bossi et al., 2008), and Germany (Busch et al., 2010). Most of these studies (except for Fuertes et al., 2017; Busch et al., 2010) included less than 10 PFAAs, with concentrations reported to be in the range of <1–1800 ng/L. The median concentrations of all PFAAs were <550 ng/L, which is smaller than for the USA and Canada. Higher abundances of PFOS and PFOA compared to shorter chain PFAAs were also observed. However, more recent studies conducted in Spain (Fuertes et al., 2017) and Germany (Busch et al., 2010), reported higher abundances of shorter chain PFAAs (≤C7).

3.2.3. Concentration and trends in Australia

Gallen et al. (2016; 2017) reported the occurrence of PFAAs in leachates from a number of active and closed landfill sites in Australia, as shown in Table 1. The PFCA and PFSA concentrations of up to 5700 ng/L and 1900 ng/L, respectively, have been reported. While these ranges are slightly smaller than for the USA, the median concentrations were <550 ng/L for all PFAAs, except PFHxA (970 ng/L), similar to those reported in European Countries (Gallen et al., 2016).

3.2.4. Concentrations and trends in China

PFAAs have been reported in leachates from four active and three closed MSW landfills in China (Yan et al., 2015) with concentrations ranging from 70 to 214,000 ng/L for PFCAs (C4-C8) and 30 to 416,000 ng/L for PFSAs (Fig. 2(a)). While these ranges are orders of magnitude higher than for other countries, the median concentrations for most PFAAs (e.g., perfluorobutanoic acid (PFBA), PFOS, PFOA, PFBS, perfluoropentanoic acid (PFPeA)) in the range of several thousands ng/L were also higher than those reported in other countries. Observed high concentrations of PFAAs could be related to PFAS contents of the consumer products, and/or higher fractions of PFAS-containing wastes (e.g., construction and demolition waste, electronics, carpet, clothing) being disposed in these landfills. However, this could not be substantiated, as no survey of PFASs in consumer products in China is available to the best of our knowledge. Also, waste compositions of the Chinese landfills were not specified by Yan et al. (2015).

In contrast to other studies, PFOA (mean contribution 29%) was found to be the most abundant PFAA, followed by PFBS (26%) and pentafluoropropionic acid (PFPrA) (16%). Despite the high abundance of PFBS in leachate, suggesting a shift towards C4-based chemistry, high PFOS concentrations (1000–6000 ng/L) were reported in Chinese landfill leachates (Yan et al., 2015). It is noteworthy that, following the phase-out of PFOS by its largest manufacturer (3M) in the USA, production in China grew rapidly (see Fig. 3) from 50 tonnes/year in 2004 to current levels of 100–200 tonnes/y (Yang et al., 2014b; Xie et al., 2013). Since the addition of PFOS to Annex B of the Stockholm Convention in 2009, China has restricted its production and use, except for specific exemptions (Wang et al., 2016). Nonetheless, past manufacturing history could be a key factor underlying the observed high relative concentrations of PFOS. Other factors could be due to (1) long use lifetimes of major PFOS-containing products (e.g., carpets, textiles); (2) long residence of PFOS-containing MSW in landfill, and/or (3) on-going uses of PFOS-containing products in China (Yan et al., 2015).

3.3. Estimating PFASs mass discharged with landfill leachate

The mass of PFASs discharged with landfill leachate is a function of PFAS concentrations in leachate and leachate volume. As discussed in section 3.2, the total PFAS concentration in leachate is determined by the number/classes of PFAS analyzed and can be highly variable, depending on landfill- and waste-related factors (discussed in section 3.5 below). Similarly, leachate volume is highly dependent on climate (in particular, rainfall and subsequent infiltration into landfill), and may vary substantially from month to month, and from year to year (Gallen et al., 2017). A recent survey (Lang et al., 2017) of PFASs in landfill leachates in the USA (70 PFASs in 95 samples) estimated national release of 19 PFASs, with >50% of samples having quantifiable concentrations. A mass estimate was obtained by coupling waste age and climate-specific PFAS concentration estimates with climate-specific estimates of annual leachate volumes using Monte Carlo analysis. The estimates for the 19 PFASs ranged from 563 to 638 kg for the sampling year, 2013. PFCAs accounted for the majority of mass estimated (291 kg/yr), closely followed by FTCAs (285 kg/yr), with lower releases of PFSAs and their precursors (84 kg/yr) (Lang et al., 2017). This indicates that the estimated mass budget of PFASs, often based on measured PFAAs concentration only (Fuertes et al., 2017; Yan et al., 2015; Benskin et al., 2012; Busch et al., 2010; Oliaei et al., 2006), (<1–25 kg/yr per landfill as shown in Table 1) could grossly underestimate the total release of PFASs from landfills.

3.4. Occurrence of PFASs in landfill ambient air

While many of the PFAA-precursors and their degradation products (e.g., FTOHs, fluorotelomer iodides (FTIs), FOSEs) are semi-volatile in nature, the role of landfills as sources of PFAS gaseous emissions to the atmosphere has received little attention. Two studies (Ahrens et al., 2011; Weinberg et al., 2011) reported higher (2–30 times) PFAS concentrations in landfill ambient air compared to control sites that were presumably not contaminated with landfill emissions. Ambient landfill air predominantly contained FTOHs, with concentrations being >90% of total precursor compounds measured (see Table 2). While FOSAs and FOSEs were also detected, their concentrations were orders of magnitude lower than for the FTOHs (Ahrens et al., 2011; Weinberg et al., 2011). 8:2 FTOH was found to be the highest sole contributor (50–65% of the $\sum$FTOHs, FOSAs, FOSEs), followed by 6:2 FTOH (15–40%) FTOHs (Ahrens et al., 2011; Weinberg et al., 2011). Higher abundance of 8:2 FTOH compared with 6:2 FTOH has been reported (Jahnke et al., 2007; Shoeib et al., 2006) to be typical of urban air. This is also supported by a recent survey (Vestergren et al., 2015) of PFASs in consumer products in Norway imported from China, which showed that 6:2 and 8:2 FTOHs were the most abundant extractable PFASs measured.

PFAAs were also detected in the particulate phase (Weinberg et al., 2011) and gas phase (Ahrens et al., 2011) of ambient landfill air. PFBA, PFHxA, PFOA were detected most frequently and at higher concentrations compared to other PFAAs in the gas phase (Ahrens et al., 2011). This might indicate abundance of PFOA and shorter-chain PFCAs in landfill waste, or reduced availability of longer-chain PFCAs in air due to their higher affinity for solid particles (Arvaniti et al., 2012). This dominance of short and even chain length PFCAs is also consistent with the PFCA distribution in landfill leachates from 22 sites in Germany (Busch et al., 2010), indicating that this pattern is typical for landfill emissions. Although PFOS is frequently detected in landfill leachate, it exhibited very low air concentrations at the landfill sites (<5 pg/m^3), likely due to strong sorption of PFOS to landfill solids, efficient trapping of PFOS in landfill gas collection, and partitioning of PFOS to landfill leachate (Ahrens et al., 2011).

3.5. Factors affecting per- and polyfluoroalkyl substances (PFASs) in landfill

Following landfilling, PFASs undergo long-term leaching, as well

Table 2
Concentration ranges of various classes of per- and polyfluoroalkyl substances (PFASs) in ambient landfill air (pg/m^3).

∑PFAAs[a]	∑FTOHs[b]	∑FTAs[c]	∑FOSAs, FOSEs[d]	∑PFAS $_{\text{WWTP}}$/∑PFAS$_{\text{reference site}}$	Landfill status	Reference
130–320	2500–26,000	Not measured	60–120	5 to 30	active	(Ahrens et al., 2011)
5–10[e]	70–100	1–10	6–20	1.5 to 2.5	Closed for previous 6 years	(Weinberg et al., 2011)
<DL[f]-40[e]	120–660	<DL-20	7–20	1.5 to 3	active	(Weinberg et al., 2011)

[a] Perfluoroalkyl carboxylic and perfluoroalkyl sulfonic acids.
[b] Fluorotelomer alcohols.
[c] Fluorotelomer acids.
[d] Perfluoroalkane and *N*-alkyl perfluoroalkane sulfonamide and sulfonamidoethanols.
[e] Measured in particulate phase.
[f] Detection limit.

as degradation of precursor compounds, processes that are affected by the physio-chemical properties of the PFASs, as well as the landfill leachate (Yan et al., 2015). As the landfilled waste passes through successive stages of aerobic, acetogenic, and methanogenic stabilization stages, significant changes occur in the physio-chemical properties, such as pH and organic and inorganic constituents (Table S3 in SI) of the leachate (Renou et al., 2008), likely affecting the mobility and degradation of PFASs. In most cases leachate from various waste cells undergoing varying states of decomposition are collected together and subsequently sampled for PFAS analysis. This, along with the uncertainty surrounding waste input in various cells, makes it challenging to conduct any mass balance to better understand the PFAS release and transformation inside landfills. In addition, climatic factors (e.g., precipitation) affecting the moisture content inside landfills, and operating conditions (e.g., compaction of the waste, waste filling procedure, leachate recirculation) could also play an important role in determining the fate and transformation of PFASs in landfills.

3.5.1. *Effect of leachate physiochemical properties*

Several studies have reported increased mobility of PFAAs with increasing pH (Gallen et al., 2017; Yan et al., 2015; Benskin et al., 2012), possibly due to the altered electrostatic behavior of the sorbents (Higgins and Luthy, 2006). This observation is consistent with sorption studies of PFOS and PFOA to diverse adsorbents, which indicated decreased sorption with increasing solution pH due to protonation of the adsorbent surface, leading to fewer positive sites on the sorbent (Wang and Shih, 2011; Yu et al., 2009). Total organic carbon (TOC) has been weakly correlated with the PFAA concentration in leachate (Gallen et al., 2017; Benskin et al., 2012), possibly due to hydrophobic partitioning of the perfluorinated chain with organic matter. Electrical conductivity (a measure of ion concentration in solution) has been both positively (Benskin et al., 2012) and negatively (Yan et al., 2015) correlated with PFAA concentration in leachate. While earlier studies (You et al., 2010; Higgins and Luthy, 2006) indicated decreased mobility of PFAAs with increasing ionic strength, more recent work suggests that the effect of ionic strength of PFAA adsorption is quite complex and often ion-type and concentration specific. For example, multivalent cations can increase sorption by acting as bridges between anionic PFASs and negatively charged surfaces (Kim et al., 2015), whereas anions (e.g., Cl^-, SO_4^{2-} or $Cr_2O_7^{2-}$) have been reported to compete with anionic PFASs for adsorption sites, (in boehmite, chitosan and resins) leading to increased solubility of anionic PFASs (Du et al., 2014). This suggests that the observed seasonal variation of macro-constituents (e.g., Cl^-, Ca^{2+}, Mg^{2+}, SO_4^{2-}) in leachate (Kulikowska and Klimiuk, 2008) likely contributes to the observed variability and patterns of PFASs concentration in leachates. Sorption behavior is also affected by carbon chain length and the functional head group of the PFASs (Higgins and Luthy, 2006).

3.5.2. *Biological processes inside landfills*

Biodegradation is arguably one of the most important factors determining the fate of PFASs in landfills. Allred et al. (2015) and Lang et al. (2016) studied the evolution of PFASs into leachate using anaerobic landfill reactors (fed with MSW, carpets and clothing). While PFASs were released through a combination of biological (e.g., biodegradation) and abiotic (e.g., desorption) processes, the leachates from live bioreactors (producing methane) had on average 5 to 10 times higher ∑PFAS than the average for biologically inactive reactors (Lang et al., 2016; Allred et al., 2015). Following the onset of methanogenic conditions, concentrations of known biodegradation intermediates of PFAA precursors (i.e., methyl-perfluorobutane sulfonamide acetic acid (MeFBSAA), n:2 and n:3 FTCAs) increased steadily, with 5:3 FTCA becoming the single most concentrated PFAS (Allred et al., 2015). While the aforementioned studies have provided some valuable insights into the release of PFASs in landfills, further research including measurement of semi-volatile PFASs is needed to fully comprehend the transformation process.

Landfill operating conditions, such as leachate recirculation, could be important factors affecting biodegradation. For example, Benskin et al. (2012) reported order of magnitude lower ΣPFAS concentrations, (consisting entirely of PFAAs) in recirculated leachate, compared with flow-through leachate containing both PFAAs and their precursors (31–71% PFAAs and 29–69% PFAA-precursors). While it is possible that the PFAS profile observed in the single recirculated leachate sample was not representative of leachate from this site over the long term, another explanation could be that recirculating leachate back to the landfill facilitated more biodegradation of PFAA-precursors (Benskin et al., 2012). While Huset et al. (2011) observed slightly higher ∑PFAA-precursors in flow-through leachate, the few other studies which measured PFAA-precursors (Lang et al., 2017; Allred et al., 2014) did not identify the recirculated leachate sample. Therefore the observations of Benskin et al. (2012) need to be substantiated by other studies.

3.5.3. *Effect of climate*

Precipitation can dilute leachate, resulting in decreased PFAS concentration within a time frame of a single day (Gallen et al., 2017; Benskin et al., 2012). On the other hand, changes in the moisture content of wastes resulting from rainfall can affect hydrolysis reactions and bioactivity (through bacterial growth, mixing of leachate resulting in PFAS concentration change, etc.) inside landfill over longer periods (e.g., 2 weeks) (Benskin et al., 2012). However, Benskin et al. (2012) did not observe any significant correlations between any PFAS congener and cumulative 2-week precipitation for temporal leachate samples collected from a landfill. A more recent study (Lang et al., 2017) of 18 landfills located in different climatic conditions (wet, temperate and arid) observed that several PFAS compounds (perfluorononaic acid (PFNA), perfluorodecanoic acid (PFDA), PFBS, PFOS, 6:2 FTSA, and methyl-perfluorooctane sulfonamide acetic acid (MeFOSAA)) had significantly higher concentrations in leachates from wet climates, suggesting that leaching governed release of these compounds.

3.5.4. *Effect of waste age and waste composition*

Statistically significant decreases in the concentrations of several PFASs with increasing landfill age have been observed in studies of multiple landfills (both active and closed) (Gallen et al., 2017; Lang et al., 2017; Busch et al., 2010). This trend could result from industrial significant shift of PFASs used in consumer products (Gallen et al., 2017; Lang et al., 2017). For example, Lang et al. (2017) observed higher PFBS and PFNA concentration in leachate from young waste, possibly due to increasing manufacture of PFBS- and PFNA-based products as alternatives to PFOS- and PFOA-based products. Decreases in the concentrations of PFASs in wastes (e.g., due to biodegradation) could account for the observed decreasing trend of PFAS degradation intermediates (e.g., 8:2 FTCA, 5:3 FTCA, MeFBSAA and MeFOSA) with landfill age (Lang et al., 2017). PFASs with similar concentrations in old and young wastes, will likely continue to be released for many years (Lang et al., 2017). Collected leachate samples often contained mixtures of leachate which have undergone various stages of degradation from various waste cells. As a result, it is impossible to comment on the effect of any specific stabilization stage on the PFAS concentration. The above-mentioned studies considered the total period of operation (final year minus opening year) to calculate the average age of the waste.

Lab-scale anaerobic reactors treating various types of refuse, such as MSW (Allred et al., 2015), carpets and clothing (Lang et al., 2016), have demonstrated the abundance of various classes of PFAS, indicating the importance of waste type in determining the PFAS concentration. For example, Lang et al. (2017) observed that short carbon-chain ($\leq$C6) PFAS were dominant in reactors treating waste carpets, whereas reactors treating clothing accumulated short-chain PFCAs, PFOA, and 8:2 FTSA. High variability in total PFAS release was observed, even for reactors receiving similar types of waste (i.e., clothing) (Lang et al., 2016). Gallen et al. (2017) also observed that leachate from operating landfills accepting >50% construction and demolition waste generally had higher PFAA concentrations compared to landfills accepting >50% MSW. Understanding PFAS trends due to variation in waste type could be challenging considering the variation in PFAS contents in similar categories of waste (Lang et al., 2016), the heterogenous nature of the solid waste and uncertainties surrounding the input of solid wastes to landfills.

4. Fate of PFASs in leachate treatment systems

One of the most common waste management practices is to send leachate to an off-site domestic WWTP. The fate and occurrence of PFASs in WWTPs have recently been thoroughly reviewed by Arvaniti and Stasinakis (2015), and are outside the scope of this paper. Other leachate management options include on-site pre-treatment, followed by off-site discharge at a WWTP, and complete treatment and discharge on-site (Townsend et al., 2015). Leachate treatment options can be broadly categorized as either physio-chemical treatment (e.g., coagulation-flocculation, chemical precipitation, membrane filtration, activated carbon adsorption, chemical oxidation) or biological treatment (e.g., activated sludge system, aerated lagoon) (Foo and Hameed, 2009; Renou et al., 2008). Similar technologies are also used for groundwater and drinking water treatment, and their effectiveness in removing PFASs has been reviewed elsewhere (Merino et al., 2016; Rahman et al., 2014). Therefore, the focus of this section is to discuss removal/fate of PFASs specifically resulting from landfill leachate treatment.

Several studies (Fuertes et al., 2017; Yan et al., 2015; Busch et al., 2010) reported an overall increase in PFAA concentrations following on-site biological leachate treatment, which is consistent with their persistent nature and possible formation from PFAA-precursors. The extent of formation observed was analyte- and site-specific, ranging between 10 and 250% for individual PFAAs (Yan et al., 2015). A wet air oxidation process contacting with ozone to create OH-radicals to degrade contaminants, also showed slightly higher (~5%) $\sum$PFAAs concentrations in the effluent leachate, but the increase was less than for biological treatment (Busch et al., 2010). An adsorption technique using activated carbon was reported to be somewhat effective (removal efficiency ranges between 70 and 99%) in removing PFAAs from leachate (Busch et al., 2010). High-pressure membrane filtration techniques such as reverse osmosis (RO) and nanofiltration (NF) removed >95% PFAAs directly from leachate (Busch et al., 2010) and from biologically-treated leachate (e.g., membrane bioreactor, followed by RO or NF) (Yan et al., 2015). On the other hand, Fuertes et al. (2017) found that ultrafiltration (UF) integrated with membrane bioreactors showed little or no removal of PFAAs. Despite the success of high-pressure filtration systems, disposal of PFAS-rich concentrate remains a challenging issue in need of careful consideration (Rahman et al., 2014).

5. Future research directions

While the presence of various classes of PFASs in landfill leachate is well documented in North America, several Northern European countries, Australia and China, no data exist for South and Southeast Asia, Southern Europe and Africa. Considering the lack of regulations limiting the manufacture and use of C8-based PFASs and the lack of pollution abatement measures such as leachate collection systems and lining materials (Ismail and Manaf, 2013), landfills in these countries could be a significant source of PFASs in the environment. This could undermine regulatory initiatives in some parts of the world, due to the long-range transport and persistence of some PFASs (such as PFAAs).

Widespread application of FTPs to consumer products (e.g., paper, textiles, leather) (Rao and Baker, 1994) imply that most FTP products will ultimately be landfilled (Washington and Jenkins, 2015). Despite the decade-long debate on their stability, recent studies show that FTPs can undergo abiotic (Washington and Jenkins, 2015) and biological hydrolysis releasing FTOHs, which then biodegrade to PFCAs with an estimated half-life range of 8–100 years (Washington et al., 2015; Rankin et al., 2014). Therefore, degradation of FTPs under landfill conditions (e.g., anaerobic, $pH > 7$) needs to be studied, including the measurement of semi-volatile compounds (e.g., FTOHs). As indicated by Washington et al. (2014), concentrations of volatile degradation products of FTP measured by GC-MS could be much more than an order of magnitude higher compared to non-volatile PFASs measured by LC-MS/MS.

To avoid significant underestimation of the total PFAS released with landfill leachate, PFAA-precursors and their degradations products (e.g., n:3 FTCA, FSAA) should be included in monitoring studies. Phillips et al. (2007) reported a 100-times smaller toxicity threshold of FTCAs compared to PFCAs for freshwater microorganisms. PFAA-precursor concentrations in leachate would provide valuable information from a water quality perspective as well. Owing to the semi-volatile nature of some precursor compounds and their degradation products, studies need to include landfill gas to understand degradation pathways and the overall fate of PFASs. This would also enable more realistic assessments of the release of PFASs to the environment with landfill gas.

More than 3000 PFASs are available on the market, and the identity of many are unknown (Wang et al., 2017). In addition to the maximum number (70 compounds) of PFASs studied in landfills, there must be more PFASs present in landfill leachates. Application of non-targeted methods (e.g., total oxidable precursor assay, total fluorine analysis) could provide valuable information regarding the unknown PFASs in leachates. New analytical techniques including PFAS classes never studied in leachate could provide useful information. For example, among the legacy PFASs, mixtures of C6–C12 perfluoroalkyl phosphonic and phosphinic acids (PFPAs and PFPiAs) used as wetting agents in consumer products (Wang et al., 2016); FTI, fluorotelomer acrylate and fluorotelomer methacrylate (FTMAC) used in impregnating agents (and their degradation products) (Favreau et al., 2017) should be included in landfill studies. Emerging PFAS like perfluoropolyethers (PFPEs), used as fluorinated alternatives of PFOS and PFOA in fluoropolymer manufacture (e.g., ADONA from 3M/Dyneon ($CF_3OCF_2CF_2CF_2OCHFCF_2COO\text{-}NH_4^+$), GenX from DuPont ($CF_3CF_2CF_2OCF(CF_3)COO\text{-}NH_4^+$)), surface treatment food contact material (Wang et al., 2013), are now being increasingly detected in various environmental media, as outlined in a recent review (Xiao, 2017). Research gaps regarding fate and transport of the emerging PFASs (e.g., PEPEs) in the environment (including landfills) need to be addressed.

While the fate and release of PFASs in landfill have been studied to some extent, very limited research exists on the performance of current containment practices (e.g., landfill liners). Leaching cell tests with sand/bentonite mixture barriers showed partial retention of PFASs (10 PFCAs, 4 PFSAs, 1 FOSA and 3 FTUCAs), decreasing over time, indicating limited effectiveness of sodium bentonite liners in landfills in containing PFASs (Li et al., 2015). Therefore, more research is needed to evaluate the effectiveness of current containment practices and how they can be improved to reduce PFAS emissions from landfills.

6. Conclusions

This study reviews publications which have reported the occurrence and sources of PFASs in landfills, to identify temporal and geographical trends of PFASs in landfill leachate and to identify and discuss factors affecting PFASs in landfills. The need for further research has been discussed. Research over the past decade has shown that PFAAs are routinely detected in landfill leachate, with short chain (C4-C7) PFCAs being more abundant than longer-chain ones, possibly reflecting greater mobility and increasing application of shorter-chain compounds. Despite its restricted use, PFOA remains one of the most frequently detected and abundant PFCAs in landfill leachate. This indicates that, if not managed properly, landfills could act as secondary sources of PFOAs in the environment.

Recent studies also document the presence of PFAA-precursors and their degradation products in landfill leachate, at concentrations comparable to, or higher than, the most frequently detected PFAAs (e.g., PFBA, PFOA, PFOS). Landfill ambient air also contains elevated concentrations of PFASs, primarily semi-volatile precursor compounds (such as FTOHs), compared to upwind control sites, suggesting that landfills likely act as emission sources of atmospheric PFASs. The fate and transformation of PFASs inside landfills are complex, affected by combinations of external (e.g., climate, waste input) and internal (e.g., biodegradation, sorption) factors. Release of most of the PFASs from waste to leachate occurs as a result of biodegradation, closely associated with onset of the methanogenic phase. The methane yielding stage also results in higher pH (>7) of leachate, correlated with greater mobility of PFAAs.

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Appendix A. Supplementary data

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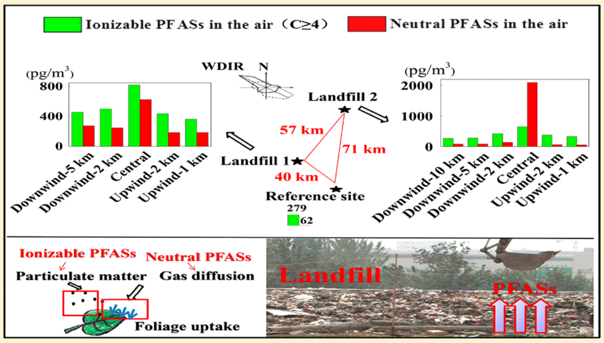
Occurrence and Phase Distribution of Neutral and Ionizable Per- and Polyfluoroalkyl Substances (PFASs) in the Atmosphere and Plant Leaves around Landfills: A Case Study in Tianjin, China

Ying Tian,[†] Yiming Yao,[†] Shuai Chang,[†] Zhen Zhao,[†] Yangyang Zhao,[†] Xiaojia Yuan,[†] Fengchang Wu,[‡] and Hongwen Sun*,[†]

[†]MOE Key Laboratory of Pollution Processes and Environmental Criteria, College of Environmental Science and Engineering, Nankai University, 300350 Tianjin, China

[‡]State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Science, 100012 Beijing, China

Supporting Information



ABSTRACT: A total of 23 per- and polyfluoroalkyl substances (PFASs) were investigated in the air, dry deposition, and plant leaves at two different landfills and one suburban reference site in Tianjin, China. The potential of landfills as sources of PFASs to the atmosphere and the phase distribution therein were evaluated. The maximum concentrations of $\sum$PFASs in the two landfills were up to 9.5 ng/m^3 in the air, 4.1 μg/g in dry deposition, and 48 μg/g lipid in leaves with trifluoroacetic acid and perfluoropropionic acid being dominant (71%−94%). Spatially, the distribution trend of ionizable and neutral PFASs in all three kinds of media consistently showed the central landfill > the downwind > the upwind > the reference sites, indicating that landfills are important sources to PFASs in the environment. Plant leaves were found effective in uptake of a variety of airborne PFASs including polyfluoroalkyl phosphoric acid diesters, thus capable of acting as a passive air sampling approach for air monitoring.

INTRODUCTION

Per- and polyfluoroalkyl substances (PFASs) have been widely used in industry and consumer products[1,2] due to their chemical and thermal stability together with their amphiphilic nature.[3] Perfluoroalkyl acids (PFAAs), are stable forms of PFASs being most frequently detected in the surface environment, especially for medium- and long-chain analogues (C7−C12), such as perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS).[4−7] Owing to their persistence, bioaccumulation, toxicity, and the high detection frequencies in the environmental and biota samples in remote regions,[8,9] PFOS and its salts were listed as persistent organic pollutants (POPs) under the Stockholm Convention in 2009, and the phase-out of PFOA has been implemented in many regions.[10] Meanwhile, short-chain analogues (C4−C6), which are recognized as less toxic and bioaccumulative to aquatic organisms and human beings, have been used as substitutes for long-chain PFASs.[11,12]

PFAAs can be directly released from products as well as derived from incomplete degradation of their precursors, which have greater production and wider applications. Fluorotelomer alcohols (FTOHs),[13,14] perfluorooctane sulfonamides (FOSAs),[15] and perfluorooctane sulfonamidoethanols (FOSEs)[16] are precursors of most concern, and their degradation in various media has been well investigated in laboratory studies, where PFAAs and other saturated and unsaturated polyfluorinated acids were proposed as stable metabolites.[17] Hence, the long-range atmospheric transport (LRAT) of precursors and their subsequent degradation are proposed as a dominant source of PFAAs detected in the remote environment.[13] For the past ten years, polyfluoroalkyl phosphoric acid diesters (diPAPs) that are ionizable and biodegradable to perfluoroalkyl carboxylic acids (PFCAs) have been detected in human specimens,[18,19] indoor dust[20,21] as well as seawater.[22] The high levels of up to 1.9 × 10^2 μg/g in indoor dust[20] have raised emerging concerns for their human exposure risk. More recently, diPAPs were found occurring in the particle phase of the oceanic atmosphere indicating a direct transport from nearby source regions.[23] Meanwhile, C2−C3 PFCA analogues (referred as ultrashort-chain PFCAs), especially trifluoroacetic acid (TFA), have been detected at high levels of 1.0 × 10^3 pg/m^3 to 2.1 × 10^3 pg/m^3 in the atmosphere of Beijing and up to 2.4 μg/L in Northern American

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precipitation.[24,25] Despite the fact that they may come from the breakdown of new refrigerants, such as HFC-134a, HCFC-123, and HCFC-124,[26] the degradation of PFAS precursors is suggested to also make an important contribution to these ultra-short-chain PFCAs in the environment.[13,15]

Most disposed products in landfills, can undergo both physical leaching by infiltration of precipitation and anaerobic/aerobic biodegradations. Hence, landfill is identified as an important source for various contaminants in the environment.[27] Dumped PFAS-treated products, such as carpets, textiles, and leathers, are potential sources of PFASs in landfills. Volatile and semi-volatile degradants in the waste may escape into the atmosphere from further degradation. However, only two studies have reported the fate of PFASs in the atmosphere of landfills.[28,29] One investigated the atmospheric concentrations of PFASs around one WWTP and two landfills in Ontario, Canada, revealing a comparable level of $\sum$PFASs over the landfills (2.8×10^3 pg/m^3 to 2.6×10^4 pg/m^3) with those over the WWTP (2.3×10^3 pg/m^3 to 2.4×10^4 pg/m^3).[28] The other only measured neutral PFASs (84 pg/m^3 to 7.1×10^2 pg/m^3) in the air over two landfills in Germany.[29] In China, the occurrence of PFASs in the atmosphere at and around landfills is yet to be reported, and their behaviors including ultra-short-chain PFCAs and diPAPs are of greater interest.

Wet and dry deposition brings atmospheric particulate matter to the land surface, which acts as a sink to the particle-associated contaminants in the atmosphere. The atmospheric partitioning of PFASs between gas and particle phases plays a key role on their LRAT potential and atmospheric fate.[30] Besides, particulate matter may provide sites for heterogeneous oxidation of volatile precursors to yield PFCAs.[31] Nevertheless, only a few studies have reported the levels and partitioning characteristics of PFASs to atmospheric particulate matter.[32−34] These studies suggested that hydrophobicity of the perfluoroalkyl tails and specific interactions between head groups and particulate components coinfluence the partitioning of ionizable PFASs, while that of neutral PFASs may mostly depend on their chain lengths. Although with rapidly increasing concentrations in the environment,[24,35] the occurrence of ultra-short-chain PFCAs in atmospheric particulate matter is less frequently reported.[36−38] Hence, the distribution of PFASs in particulate matter at and around landfills is to be clarified for their influence on atmospheric transport, particularly for ionizable PFASs including ultra-short-chain PFCAs.

Plant leaves are a dynamic environmental compartment that can take up various contaminants from the atmosphere. Since the 1960s, plant leaves have been used as a kind of rapid, easy, and economic bio-monitoring tool for polychlorinated biphenyls (PCBs),[39] polycyclic aromatic hydrocarbons,[40] and pesticides.[41] Among few studies that have reported PFASs in plant leaves,[42−44] only one investigated the distribution of TFA in pine needles.[45] These studies revealed that plant leaves can accumulate PFASs and can be used as a kind of passive monitoring tool. Plant leaves may accumulate contaminants from the atmosphere via two major pathways: the direct uptake of gas-phase chemicals from the ambient air and the uptake of the chemicals associated with atmospheric particulate matter via mass exchange.[46] The uptake of PFASs from the atmosphere in plant leaves depends largely on species of PFASs with different properties. Therefore, leaf uptake of particle-phase PFASs is probably a vital process especially for those with high affinity to atmospheric particulate matter.

Hence, the objectives of this study were (i) to determine the source potential of landfills to PFASs measured in the air, dry deposition, and leaves from surrounding environments; (ii) to investigate the partitioning of PFASs between air and plant leaves with potential contribution of dry deposition; and (iii) to examine the suitability of using plant leaves as a bio-monitoring tool for airborne PFASs. The levels of 7 neutral and 16 ionizable PFASs in air samples, dry deposition, and plant leaves collected from surroundings of two landfills and one reference site in Tianjin, China, were investigated. The distribution characteristics and correlations of typical ionizable and neutral PFASs between plant leaves, air, and dry deposition were discussed.

■ MATERIALS AND METHODS

Chemicals and Reagents. Seven neutral and 16 ionizable PFASs were analyzed. The neutral PFASs included 6:2, 8:2, and 10:2 FTOHs; *N*-methyl and *N*-ethyl (*N*-Me/Et) FOSAs; and *N*-methyl and *N*-ethyl (*N*-Me/Et) FOSEs. The ionizable PFASs included 6:2 and 8:2 diPAPs; C2−C12 PFCA analogues (TFA, PFPrA, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, and PFDoDA); C4, C6, and C8 perfluoroalkanesulfonic acid (PFSA) analogues (PFBS, PFHxS, and PFOS). The mass-labeled ionizable and neutral PFASs, including M−PFBA, M−PFOA, M−PFHxS, M−PFOS, M−6:2 diPAP, M−8:2 diPAP, M−6:2 FTOH, M−8:2 FTOH, M−*N*-EtFOSE, and M−*N*-EtFOSA), were used as internal standards in HPLC−MS/MS and GC−MS analysis. Methanol, ethyl acetate, and ammonia acetate of HPLC-grade were used. Milli-Q water was used throughout the study. The commercial sources of the target PFASs are listed in Table S1 in the Supporting Information (SI).

Sample Collection and Storage. Sampling campaigns were carried out at and around two landfills, and at one reference site from May 8th to June 9th 2016. The geographic map of the sampling sites is shown in Figure S1. Table S2 displays the geographic information on all the 12 sampling sites. The details of the two landfills, Shuangkou (SK) landfill and Baodi (BD) landfill, are described in the SI. JN, a suburban reference site far away from any known point sources of PFASs, is about 40 km and 71 km away from SK and BD landfills, respectively. Plant leaves (n = 72) and air samples (n = 12) were collected at all the 12 sites, and dry deposition were collected in duplicates (n = 10) at SK-Central, SK-Down-5 km, BD-Central, BD-Down-5 km, and JN. During the period of sampling campaign, the predominant wind direction was south or southeast with a speed of 11 km/h-28 km/h and the temperature varied between 16 °C and 29 °C.

Air samples were collected with a passive sampling technique using sorbent-impregnated polyurethane foam (SIP) disks. A rate of 4 m^3/d was used for calculation of the effective air volume during the 30-day sampling period as Ahrens et al. recommended.[47] The SIP disks were precleaned by the method described by Shoeib et al.[48] All passive samplers were assembled and loaded with SIP disks immediately on site to avoid contamination in transit. Three SIP disks were exposed to the atmosphere at SK, BD, and JN for about 8 h, respectively, and taken back to the laboratory for measurement of field blanks. Dry deposition samples were collected with pre-cleaned stainless steel cylinder buckets (40 cm in diameter ×70 cm in height). Plant leaves of local species were collected by precleaned scissors, including Chinese pine (*Pimus tabulaeformis Carr.*),Oriental plane (*Platanus orientalis Linn*), and Abele

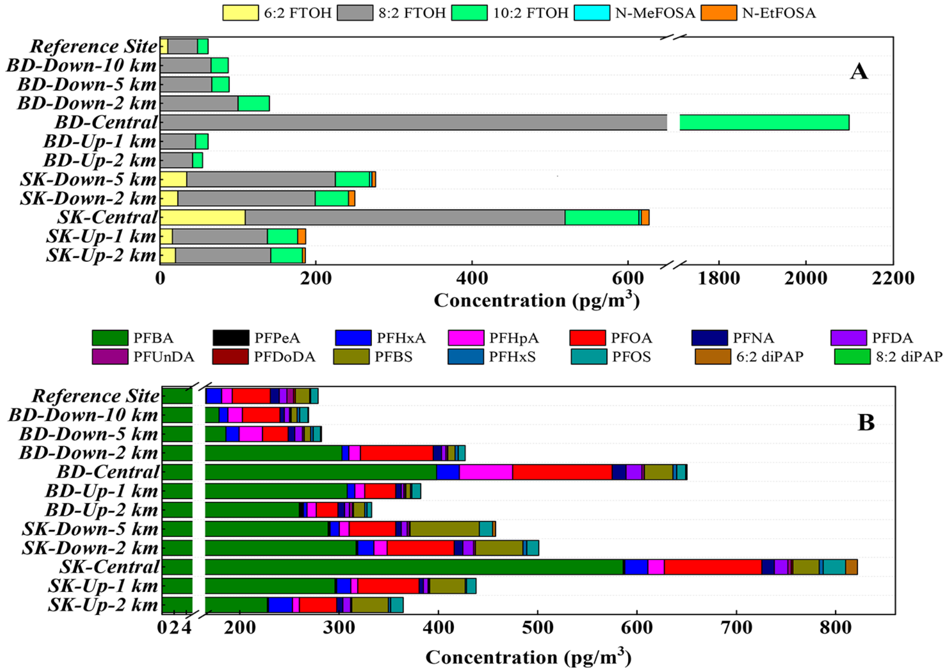

Figure 1. Concentrations and spatial distributions of neutral and ionizable PFASs in the air around two landfills (SK and BD) and the suburban reference site (JN) in Tianjin, China. SK-Central and BD-Central represents the central area of the two landfills, SK-Up and BD-Up represent upwind sites, SK-Down and BD-Down represent downwind sites. (A) neutral PFASs; (B) ionizable PFASs (C ≥ 4).

(*Populusalba*). At the end of the sampling campaign, all the samples were carefully wrapped in foil, put in pre-cleaned polypropylene bags, and kept at −20 °C before extraction.

Sample Pretreatment and Analysis. The 16 ionizable PFASs were analyzed using HPLC−MS/MS (Agilent Technologies, U.S.A.) and the 7 neutral PFASs were analyzed using GC−MS (Agilent Technologies, U.S.A.). Lipid contents of leaves were measured by weight difference before and after the extraction. The details of sample pretreatment and instrumental parameters are provided in the SI.

Quality Assurance and Statistical Analysis. Plant and dry deposition samples were numbered randomly and analyzed in duplicate. All the analyzed PFASs were normalized against the recovery of the corresponding mass-labeled internal standards spiked prior to extraction (Table S3). The limit of detection (LOD) and the limit of quantification (LOQ) were defined as a signal-to-noise of 3:1 and 10:1, respectively. The instrumental LOQs and matrix recoveries of the analyzed PFASs are shown in Tables S4 and S5, respectively. The final concentrations of the target analytes have been adjusted by field blank values, which were very low (0.04 pg/m^3 to 0.36 pg/m^3) as compared to levels in air samples. The method detection limit (MDL) was derived from three times the standard deviation of the field blank values. For analytes not detected in field blanks, MDLs were derived directly from three times of their corresponding LODs. All the data below MDLs were considered as not detected (n.d.). The MDLs and detection frequencies of the target analytes for air, dry deposition, and leaf samples are shown in Tables S4 and S5, respectively. For each batch of 10 samples, one procedural blank was processed to avoid background contamination. The SPSS 22 software was used for statistical analysis. Spearman rank and Pearson correlation analyses were used to examine possible correlations among various PFASs in different samples.

■ RESULTS AND DISCUSSION

Levels and Distribution of PFASs in the Air. As shown in Figure 1A, the highest concentrations of $\sum$FTOHs were 6.1 × 10^2 pg/m^3 and 2.1 × 10^3 pg/m^3 detected at SK-central and BD-central, respectively, whereas the concentration was 62 pg/m^3 at JN, the suburban reference site. The concentrations decreased evidently as distance from the central sites increased. Elevated mean levels of FTOHs were found at downwind sites as compared to the upwind sites for both SK (2.6 × 10^2 pg/m^3 vs 1.8 × 10^2 pg/m^3) and BD (89 pg/m^3 vs 58 pg/m^3) landfills (Figure S2). 8:2 FTOH was dominant with a concentration range of 42 pg/m^3 to 1.6 × 10^3 pg/m^3, followed by 10:2 FTOH (13 pg/m^3 to 5.4 × 10^2 pg/m^3). 6:2 FTOH was only detected at SK with a concentration range of 16 pg/m^3 to 1.1 × 10^2 pg/m^3 (Figure 1A). 6:2 FTOH may release much faster than 8:2 and 10:2 FTOHs in consumer products because of its higher vapor pressure.[49] This may lead to less residual 6:2 FTOH left in products disposed in landfills. After release, 6:2 FTOH is more susceptible to consumption by biodegradation in landfill leachate or soil.[50] In addition, it is only in recent years that the major fluorochemical manufacturers have been moving toward 6:2 FTOH as a principal raw material to manufacture FTOH-based products in an effort to eliminate PFOA and its precursors.[51] Due to a relatively short application time 6:2 FTOH-based products may have not been disposed to landfills yet. These reasons possibly accounted for the low concentrations or no detection of 6:2 FTOH in the two landfills. As reported in the other two studies 6 years ago, the levels of 6:2 FTOH were also consistently lower than those of 8:2 FTOH in the air over landfills.[28,29] Nevertheless, a previous study found that 6:2 FTOH was predominant in the air over WWTPs with a concentration of 9.0 × 10^2 pg/m^3 to 1.2 × 10^4 pg/m^3.[28] Our previous study also found an elevated air level of 6:2 FTOH over influent of a WWTP.[34] These results suggested either a delayed disposal to landfills or a faster release from products for 6:2 FTOH.

FOSAs were only detected at SK (3.6 pg/m^3 to 13 pg/m^3) with *N*-EtFOSA being dominant (3.6 pg/m^3 to 10 pg/m^3), while FOSEs were not detected at any sites. The concentrations of FOSAs were much lower than those of FTOHs, which was consistent with those reported in the literature.[28,29] This can be ascribed to the phase-out of perfluoro-1-octanesulfonyl fluoride (POSF) related products[52] and the increasing application of telomerisation products.[11] It is also possible that the release of FOSEs from landfill sites could be limited due to their higher logK_{OA} values as compared with FTOHs (6.6−7.1 vs 4.6−5.7).[53] In addition, the atmospheric lifetime of FOSEs was estimated for approximately 2 days based on smog chamber test, indicating a fast consumption of FOSEs in the atmosphere.[16] Photodegradation of FOSEs by hydroxyl radicals may produce FOSAs of much longer atmospheric lifetime (20−50 days)[15] and even further cleave the sulfur−carbon bond to yield persistent PFOA.[49] Therefore, the occurrence of ionizable PFASs in the atmosphere can be expected.

As shown in Figure 1B, the highest total concentrations of ionizable PFASs ($C \geq 4$) occurred at SK-Central (8.2×10^2 pg/m^3) and BD-Central (6.5×10^2 pg/m^3). The total concentrations of ionizable PFASs ($C \geq 4$) at the two landfills were all higher than that of the suburban reference site JN (2.8×10^2 pg/m^3 to 8.2×10^2 pg/m^3 vs 2.8×10^2 pg/m^3) except for the farthest site BD-Down-10km. Although not statistically significant ($p = 0.100 > 0.05$), the total mean concentration of ionizable PFASs ($C \geq 4$) at SK (5.2×10^2 pg/m^3) was higher than that at BD (4.0×10^2 pg/m^3). Specifically, the total concentrations of PFSAs were significantly higher at SK than at BD ($p < 0.01$) (Figure S2). These differences may be due to a larger landfill capacity for SK. However, it should be noted that FTOHs showed an opposite trend at landfill central sites. This may be ascribed to the longer time of operation for SK, which leads to different conditions inside the landfill and hence different degradation rates of contaminants. The total mean concentrations of ionizable PFASs ($C \geq 4$) were higher at the downwind sites than the upwind sites at SK (4.8×10^2 pg/m^3 vs 4.0×10^2 pg/m^3) while only BD-Down-2km was higher than those at upwind sites at BD (4.3×10^2 pg/m^3 vs 3.6×10^2 pg/m^3) (Figure S2). The trend was consistent with those of FTOHs. In general, the total concentrations of long−chain ($C \geq 8$) PFCAs (37 pg/m^3 to 1.3×10^2 pg/m^3) were significantly lower than those of short- and medium-chain (C4−C7) PFCAs (2.0×10^2 pg/m^3 to 6.3×10^2 pg/m^3) ($p < 0.01$). PFBA was the predominant PFCA with a concentration range of 1.8×10^2 pg/m^3 to 5.9×10^2 pg/m^3 (Figure 1B), which was higher than the levels at Canadian landfills (40 pg/m^3 to 1.0×10^2 pg/m^3).[26] The dominating level of PFBA may result from the increasing use of short-chain PFAS precursors[11,54] and their rapid release and degradation.[15] Among long-chain PFCAs, PFOA was dominant at most sites with a concentration range of 22 pg/m^3 to 1.0×10^2 pg/m^3 followed by PFHpA (7.0 pg/m^3 to 54 pg/m^3) and PFHxA (3.6 pg/m^3 to 24 pg/m^3). In China, materials treated with PFOA and their precursors have not been banned for use in various industrial and domestic applications.[55] This makes China one of the few remaining major producers and potentially the largest consumer of PFOA and their precursors thus far.[11] Therefore, the current air profiles of PFASs indicated that off-gassing of PFOA and their precursors from the consumer products disposed in landfills is important to consider.

The air concentrations of PFBS, PFHxS, and PFOS, were 4.8 pg/m^3 to 70 pg/m^3, < 0.02 pg/m^3 to 4.2 pg/m^3, and 4.4 pg/m^3 to 23 pg/m^3, respectively (Figure 1B). PFBS was the dominant PFSA but with concentrations much lower than PFBA. The levels of PFOS at landfills were comparable to those measured over a WWTP (11 pg/m^3 to 19 pg/m^3) in Tianjin, China.[34] PFBS to PFOS concentration ratios were calculated as 0.54−5.3 (mean 2.4) in the present study and values up to 3.0−14 have also been reported at WWTPs from some economically developed areas of China.[56] These results suggested an ongoing shift to short-chain alternatives after the ban of PFOS and related products.[54]

The air levels and detection frequencies (50%) of diPAPs were both lower than those of other PFASs at landfills, where their occurrence and distribution characteristics were reported for the first time. 6:2 diPAP was detected with a concentration range of <0.12 pg/m^3 to 12 pg/m^3 (mean 2.9 pg/m^3) and 8:2 diPAP was only detected at BD-Central with a concentration of 0.42 pg/m^3 (Figure 1B). diPAPs were not detected at the suburban reference site. The air levels of ΣdiPAPs at landfills were higher than those at local urban area, where diPAPs were detected only in particle phase at a mean level of 1.3 pg/m^3.[23] DiPAPs are less volatile[53] and susceptible to biodegradation as suggested in a study on anaerobic model landfill reactors.[57] In the atmosphere, diPAPs may also be consumed potentially by both gas-phase and heterogeneous reactions.[58,59] Even so, the highest concentrations of ΣdiPAPs were 12 pg/m^3 and 1.1 pg/m^3 detected at SK-Central and BD-Central, respectively. Spatially, diPAPs were barely detected at upwind sites while levels of ΣdiPAPs were found 14%−73% lower at distant downwind sites.

As compared with literature (Table S6), the air concentrations of PFASs at the landfills ($\sum$FTOHs 55 pg/m^3 to 2.1×10^3 pg/m^3, $\sum$PFCAs ($C \geq 4$) 2.5×10^2 pg/m^3 to 7.6×10^2 pg/m^3) in the present study were higher than those at the WWTPs investigated in the past five years in Tianjin, China ($\sum$FTOHs 94 pg/m^3 to 1.3×10^2 pg/m^3, $\sum$PFCAs 88 pg/m^3 to 2.3×10^2 pg/m^3)[34] and in Germany ($\sum$FTOHs 12 pg/m^3 to 5.1×10^2 pg/m^3, $\sum$PFCAs < MDL-13 pg/m^3),[60] as well as the levels in the ambient atmosphere,[5,61] while comparable to the levels at landfills reported by previous studies.[28,29] These results support that landfill is an important source of PFASs to its surrounding atmosphere.

Levels of PFASs in Dry Deposition. Dry deposition samples were collected at 5 sites, and the results are summarized in Figure 2. Similar to the air profiles, the concentration trends of neutral and ionizable PFASs both showed central landfill > the downwind > the suburban reference sites, and the total concentrations of PFASs were higher at SK than at BD (82 ng/g to 1.6×10^2 ng/g vs 45 ng/g to 70 ng/g) (Figure S3). It suggests that the spatial distribution of these PFASs was coinfluenced by the factors including prevailing wind direction, distance from the point source, and the landfill capacity. For neutral PFASs, 8:2 FTOH was dominant with a concentration range of 0.33 ng/g to 2.8 ng/g, followed by 10:2 FTOH (<0.21 ng/g to 1.7 ng/g). 6:2 FTOH, FOSAs, and FOSEs were not detected at any sites, which could be explained either by their low concentrations or no detection in the air samples. For short-chain PFCAs, PFBA was dominant with a concentration range of 11 ng/g to 55 ng/g, and those at landfill central sites (22 ng/g to 55 ng/g) were higher than the levels (<0.03 ng/g to 20 ng/g) in outdoor dust across mainland China ($p < 0.01$).[55] PFOA was the dominant long-chain PFCA

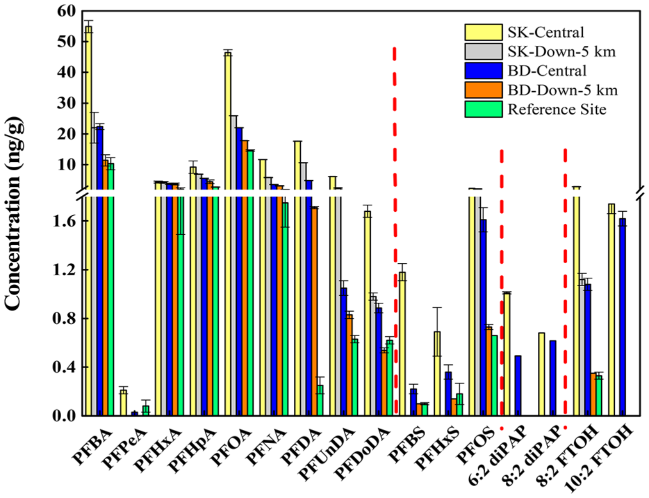

Figure 2. Mean concentration ($n = 2$) and spatial distributions of neutral and ionizable PFASs ($C \geq 4$) in dry deposition around two landfills (SK and BD) and the suburban reference site (JN) in Tianjin, China. SK-Central and BD-Central represents the central area of the two landfills. SK-Down-5 km and BD-Down-5 km represents downwind sites 5 km far from the central area. Error bars represent SD values.

with a concentration range of 15 ng/g to 46 ng/g, followed by PFNA (1.8 ng/g to 12 ng/g) and PFDA (0.25 ng/g to 18 ng/g). The total mean concentration of $\sum$PFSAs at the two landfills was 2.3 ng/g, which was about 2 orders of magnitude lower than that of $\sum$PFCAs (84 ng/g). PFOS was the dominant PFSA with a concentration range of 0.73 ng/g to 2.3 ng/g followed by PFBS (<0.03 ng/g to 1.2 ng/g). 6:2 and 8:2 diPAPs were only detected at SK-Central and BD-Central with a total concentration of 1.7 ng/g and 1.1 ng/g, respectively. Apart from reasons mentioned above, the low level of diPAPs was possibly a result of their rapid photochemical degradation.[62] In general, the total concentrations of ionizable PFASs ($C \geq 4$) in dry deposition at landfills (32 ng/g to 1.3×10^2 ng/g) were at the same level with those collected at local urban areas and at a WWTP (11 ng/g to 90 ng/g).[34]

Levels and Distribution of PFASs in Plant Leaves. The concentrations and spatial distributions of ionizable PFASs ($C \geq 4$) in the three kinds of leaves are shown in Figure 3. Enrichment of contaminants in plant leaves is influenced by many factors, such as physicochemical properties of the compound, meteorological conditions, and the composition of the plant growth.[63] The median dry-weight concentrations of ionizable PFASs ($C \geq 4$) in Chinese pine, abele, and oriental plane leaves at the two landfills were 23 ng/g dw, 17 ng/g dw, and 20 ng/g dw, which were positively associated with their measured lipid contents (13% in Chinese pine, 6.8% in abele, and 8.9% in oriental plane) ($r = 1.000$, $p < 0.01$). These results suggested that high leaf lipid content may favor the accumulation of ionizable PFASs in plant leaves. As a whole, the levels of ionizable PFASs normalized to lipid contents showed comparable levels between different leaf types and reflected consistent trends over distance (Figure 3). The total mean concentration of ionizable PFASs ($C \geq 4$) in the three kinds of leaves at SK (4.1×10^2 ng/g lipid) was higher than that at BD (2.2×10^2 ng/g lipid), and the total concentrations of ionizable PFCAs ($C \geq 4$) were higher at downwind than at upwind sites, which was statistically significant at SK. (Figure S4B) ($p < 0.01$). The total mean concentrations of medium- and long-chain PFASs ($C \geq 7$) at SK and BD (9.1 ng/g dw and 6.0 ng/g dw) were lower than those from an industrial center with condensed fluorochemical facilities in China, where the leaf concentrations of PFASs were 10 ng/g dw to 2.8×10^2 ng/g dw.[44] PFBA was a dominant PFCA with a mean

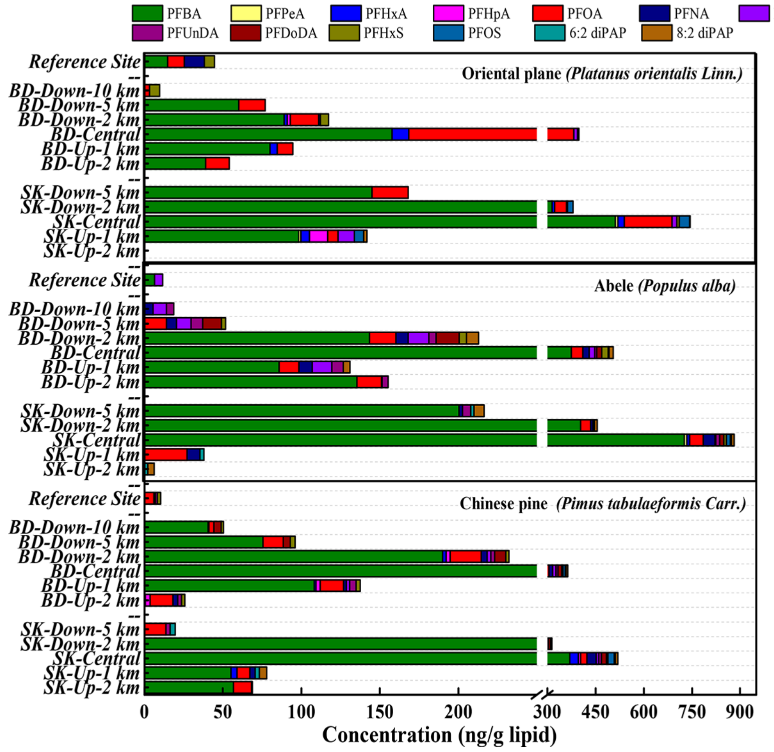

Figure 3. Mean concentrations ($n = 2$) and spatial distributions of ionizable PFASs ($C \geq 4$) in oriental plane, abele, and Chinese pine leaves.

concentration of 26 ng/g dw and 12 ng/g dw (2.9×10^2 ng/g lipid and 1.3×10^2 ng/g lipid) at SK and BD, which were comparable to those in pine needles from Norway (<MQL-23 ng/g).[43] Short-chain PFCAs such as PFBA with smaller molecular sizes may pass through the epidermis and enter into the internal part of the leaves more easily. *In addition, they have higher root-to-leaf translocation via the casparian strip than long-chain analogues from the soil.*[64] *These reasons may contribute to the dominating leaf concentrations of short-chain PFCAs.* Among medium- and long-chain PFCAs ($C \geq 6$), PFOA was most frequently detected with a concentration range of 3.3 ng/g lipid to 2.1×10^2 ng/g lipid, followed by PFNA (0.44 ng/g lipid to 37 ng/g lipid) and PFHxA (0.46 ng/g lipid to 27 ng/g lipid). In contrast, PFOS was detected with a concentrations range of 1.2 ng/g lipid-32 ng/g lipid, which were significantly lower than those of PFOA at central landfill sites ($p < 0.01$). Although hardly occurring in air samples, 8:2 diPAP was particularly detected in leaves with a concentration range of 2.2 ng/g lipid to 13 ng/g lipid, which was dominant over 6:2 diPAP (0.56 ng/g lipid to 3.5 ng/g lipid) ($p < 0.01$), which possibly suggested particular enrichment of 8:2 diPAP on plant leaves.

The spatial distribution pattern of neutral PFASs in plant leaves at all sites investigated was similar to that of ionizable PFASs (Figures S5A,B). 8:2 FTOH was dominant with a concentration range of <1.2 ng/g lipid-26 ng/g lipid (<0.15 ng/g dw to 3.4 ng/g dw) (Figure S4). However, 6:2 and 10:2 FTOHs were not detected at any sites. These results were consistent with the PFAS profiles in herbaceous plants, where 8:2 FTOH was dominant (<1.5 ng/g dw), and its concentrations were 10 times lower than those of PFOA.[42] *N*-Me and *N*-EtFOSAs were only detected at SK with concentration ranges of <2.1 ng/g lipid−9.6 ng/g lipid and <0.92 ng/g lipid−10 ng/g lipid, respectively. FOSEs were not detected at any sites. However, the total mean dry-weight concentrations of neutral PFASs (2.3 ng/g dw in Chinese pine, 1.6 ng/g dw in abele, and 0.60 ng/g dw in oriental plane) in the three kinds of leaves were not as positively associated with their lipid contents as those of ionizable PFASs ($r = 0.500$, $p = 0.667 > 0.05$) (Figure S4). It has been suggested that plant leaves may preferentially accumulate airborne semivolatile organic compounds with high $\log K_{OA}$ values (>8.0),[65,66] while those of FTOHs are mostly within the range of 4−7.[53,67] Gas-phase diffusion through stomata on leaf surface may be a preferential pathway for neutral PFASs to enter into leaves from air rather than adsorption by surface wax.[65] In this case, unlike ionizable PFASs, leaf uptake of neutral PFASs from the atmosphere is possibly less dependent on surface lipid. Besides, plant leaves are found to be able to metabolize FTOHs,[68] which may change the concentrations of both neutral and ionizable PFASs in leaves.

Among three tree species, relative standard deviations (RSDs) of dry-weight concentrations of PFASs ranged from 26% to 79%. After lipid normalization, RSDs of ionizable PFASs (C4−C8) and 8:2 FTOH particularly decreased by 3%−26%, while those of other shorter- or longer-chain analogues increased by 5%−28%. Besides surface lipids, lipids stored within cells possibly interact with ionizable PFASs in a way similar to fatty acids. Although interactions between PFASs with lipids in plants are rarely reported, PFBA in zebrafish liver cells may exert localized effects on lipid droplets.[69] This is likely a selective interaction between PFASs and intracellular lipid droplets, which are dynamic protein-embedded monolayer organelles that are also ubiquitously found in plants.[70]

In general, the detection frequencies of the target PFASs in leaves were comparatively lower than those in the atmosphere, but most PFASs in leaves followed a similar spatial distribution trend as those in the atmosphere, which suggested a good prediction of airborne PFASs by monitoring plant leaves. A higher contribution of FOSAs was also observed at SK as compared to those in air samples (ave. 51% vs 3.1%), which is partially because of their higher $\log K_{OA}$ values (Figures S2 and S4). Unlike the composition profiles in the atmosphere with dominance of FTOHs, in leaf profiles ionizable PFASs exceeded FTOHs in levels (Figure S5). This could be a result of root uptake and subsequent leaf translocation of ionizable PFASs but also likely due to the mass exchange between leaf surface and surface-attached particulate matter, which will be discussed thereafter.

C2−C3 PFCAs in the Environment. As shown in Figure S6, the concentrations of C2−C3 PFCAs in these three media were about 1 order of magnitude higher than those of medium- and long-chain PFCAs ($p < 0.01$). TFA was detected in all air samples with a predominant concentration range of 1.4 ng/m^3 to 3.0 ng/m^3, which contributed to 80% of the ΣPFCAs profiles. The air concentrations of TFA were comparable with those reported in Beijing, China (median 1.6 ng/m^3).[38] The air concentrations of PFPrA were 0.064 ng/m^3 to 0.36 ng/m^3, which were much lower than those of TFA but comparable to PFBA. In dry deposition, TFA also contributed to the majority of $\sum$PFCAs profiles with a concentration range of 1.1 ug/g to 3.7 ug/g, followed by PFPrA (78 ng/g to 7.4×10^2 ng/g), which were higher than those in outdoor dust across mainland China (1.8 ng/g to 1.2×10^2 ng/g).[71] In plant leaves, TFA was also dominant with a concentration range of 6.3 μg/g lipid to 42 μg/g lipid (0.56 μg/g dw to 3.0 μg/g dw) contributing to over 86% of $\sum$PFCAs profiles at each site. The concentrations of TFA in the present study were higher than that in pine needles (0.42 μg/g dw) from North America.[45] The overall spatial distribution of these ultrashort-chain PFCAs followed a similar trend to that of medium- and long-chain PFASs. Even so, their remarkably elevated levels suggested additional sources of these ultra-short-chain PFCAs, which were different from medium- and long-chain PFCAs. Indeed, it is well documented that C2−C3 PFCAs mainly come from the breakdown of new refrigerants, such as HFC-134a, HCFC-123, and HCFC-124.[26]

Particle-Gas Distribution of PFASs. The distribution of PFASs between particle and gas phases was roughly estimated by the ratio of their concentrations in dry deposition to those in the bulk air (K_{pa}). The ratio represents the readily precipitated portion of atmospheric particulate matter. For a conservative estimation, a moderate total suspended particulate (TSP) level of 200 μg/m^3 was used.[72]

The K_{pa} values for individual PFASs at the five sites varied broadly with structures of different PFASs, indicating that distribution based on field measurements were dramatically affected by multiple factors on both dynamic and thermodynamic aspects (Figure 4). Generally, ionizable PFASs possessed higher K_{pa} values than neutral PFASs of the same chain length (i.e., PFDA, $K_{pa} = 0.15$ and 8:2 FTOH, $K_{pa} = 0.03$, Table S7), which indicated that ionizable PFASs have higher affinity to particulate matter. For C4−C12 PFCAs, the K_{pa} values rise from 0.01 to 0.26 as carbon chain length increases, which is also predictable by the trend of their $\log K_{OA}$ values (6.0−8.4).[53] A greater $\log K_{OA}$ indicates that the compound has higher affinity to organic components in particulate matter.[73] Nevertheless, TFA and PFPrA showed higher K_{pa} values than C4−C8 PFCAs

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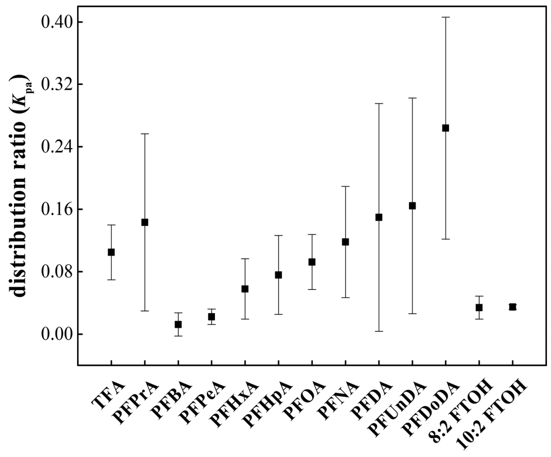

Figure 4. Particle−gas distribution ratios (K_{pa}) (n = 5) of PFASs derived from concentrations in dry deposition to those in bulk air (SIP passive sampler) at SK, BD, and JN in Tianjin, China. Error bars represent SD values.

(0.11−0.14 vs 0.01−0.09). PFCAs may have specific affinity to mineral components in particulate matter due to their polar carboxyl head groups.[32,74] This interaction could be more prominent for ultra−short−chain PFCAs due to their reduced molecular sizes. In addition, TFA and PFPrA in the atmosphere are mostly yielded from the breakdown of precursors,[15,16] which is likely to be associated with particle formation and enhanced heterogeneous degradation of their precursors on particles. The median K_{pa} value of PFOS was higher than PFOA (0.11 vs 0.09) (Table S7). This corresponds to relatively higher logK_{OA} value of PFOS as compared with PFOA (8.1 vs 7.2).[53] For other ionizable PFASs, no evident tendency was observed, which was possibly due to their low concentrations or low detection frequencies.

Correlations of PFASs in Leaves Air and Dry Deposition. Correlation analysis was carried out between the total concentrations of ultrashort-chain (C2−C3) PFCAs, ionizable PFASs (C ≥ 4), and 8:2 FTOH, in the three kinds of leaves (lipid correction) and those in air samples, as well as those in dry deposition (Figure 5). Ionizable PFASs (C ≥ 4) showed strong correlations between leaves and bulk air (Pearson's r = 0.909, $p < 0.01$) as well as between leaves and dry deposition (Pearson's r = 0.887, $p < 0.05$) (Figure 5C,D). Ultrashort-chain (C2−C3) PFCAs also showed a strong correlation between leaves and bulk air (Pearson's r = 0.768, $p < 0.01$) while they were also strongly correlated between leaves and deposition although not significantly (Pearson's r = 0.877, $p = 0.051 > 0.05$) (Figure 5A,B). However, the correlation was statistically significant for Spearman rank analysis (r = 0.900, $p < 0.05$). 8:2 FTOH was only found significantly correlated between leaves and bulk air (Pearson's r = 0.723, $p < 0.05$) and the correlation between leaves and dry deposition was weaker than those of ionizable PFASs (Pearson's r = 0.773, $p > 0.05$) (Figure 5E,F).

As suggested by the K_{pa} calculated in this study, dry deposition only accounted for less than 40% of ionizable PFASs measured in the bulk air (Figure 4). This indicates that most of ionizable PFASs in the atmosphere occurred in fine particulate matter,[34] which may as well contribute to the strong correlativity between leaves and bulk air observed in this study. In contrast, neutral PFASs that mainly occurred in the gas phase were directly absorbed by leaves via gas-phase diffusion. The similar process was also observed for PCBs.[41]

In addition to the uptake of airborne PFASs, the soil-plant transportation and plant transformation of PFAS precursors may also contribute to ionizable PFASs in leaves.[64,75] As consistently observed in hydroponic root exposure, leaf concentration factors (LCFs) of PFAAs for common crops sharply decreased as the carbon chain length increases and were

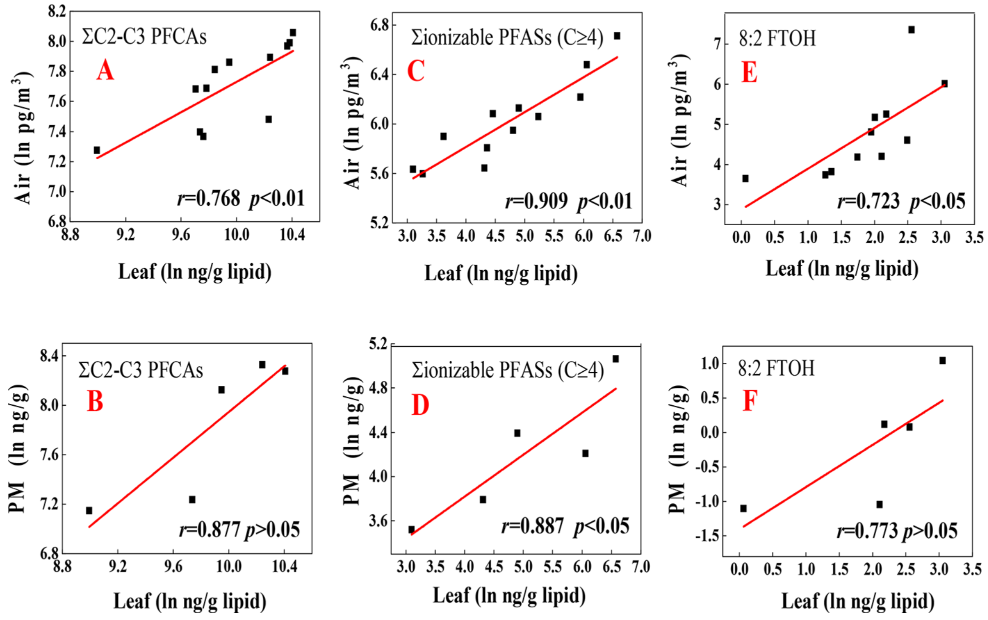

Figure 5. Correlations between natural logarithmic concentrations of PFASs in the three kinds of leaves (Chinese pine, Oriental plane, and Abele), bulk air (SIP passive sampler), and dry deposition (PM). Only the results of Pearson correlation are shown for linear fittings while those of Spearman rank correlation follow a same trend. It displays the correlations of ∑C2−C3 PFCAs between leaf and air (A) and between leaf and dry deposition (B); the correlations of ∑ionizable PFASs (C ≥ 4) between leaf and air (C) and between leaf and dry deposition (D); the correlation of 8:2 FTOH between leaf and air (E) and between leaf and dry deposition (F).

generally below 1 for PFOA or longer-chain analogues.[76,77] Although LCFs for trees are rarely reported, long-chain PFCAs (C > 8) were barely detected in tree leaves from a fire training facility, where soils and groundwater were contaminated with a suite of PFAAs including C9−C11 PFCAs.[75] In this study, long-chain PFCAs (C9−C12) were frequently detected in tree leaves (63%−68%) and their sum concentrations even exceeded those of short-chain PFCAs (C5−C7) by 1.2−50 times. Therefore, leaf uptake from the atmosphere probably remains significant for neutral and long-chain PFASs. Despite this, it is also possible that long-chain PFCAs may come from leaf metabolism of their precursors.[68] Further study is needed to deepen the understanding of foliage accumulation of PFASs.

Environmental Implication. The results suggest that direct release from the central landfill site is a significant source for PFASs in its surrounding environment. Mass emissions rates of PFASs from landfills were estimated previously.[29] Future research needs to further assess mass emission of PFASs at different stages of garbage disposal in order to fulfill lifecycle analysis. A consistent spatial distribution trend of PFAS concentrations in the three media was observed, indicating a coinfluence of the wind direction, distance from the landfills, and the treatment capacity of the landfills.

Ionizable PFASs showed higher K_{pa} values than neutral PFASs, and the values of PFCAs increased with an extending carbon chain length except for C2−C3 PFCAs, for which both K_{pa} values and concentrations were outstandingly high. The specific interaction between their carboxylic group and mineral components in particulate matter may have a greater impact as compared with other PFCA analogues.The concentrations of C2−C3 PFCAs were 1 order of magnitude higher than those of medium- and long-chain PFCAs ($C \geq 7$), while diPAPs were detected at 1 order of magnitude lower level than PFCAs.

Plant leaves were found effective in reflection of PFAS contamination, which coincided with levels of airborne PFASs, thus have a great capability for air monitoring as a passive sampling approach especially adjacent to potential point sources. Particularly for ionizable PFASs including diPAPs, the results indicated that mass exchange on leaf surface may involve dry deposition or even finer atmospheric particulate matter.

Soil-plant studies have shown that short-chain PFCAs are more effectively translocated from roots to leaves. Therefore, the source of short-chain ionizable PFASs in leaves may involve multiple processes. For ultrashort-chain PFCAs, fewer data were available on their soil-plant transportation and plant translocation, which needs further study. Even so, the uniformly high levels of TFA and PFPrA in the atmosphere, dry deposition, and leaves suggested that plant leaves remain a promising passive sampling approach for these ultrashort-chain PFCAs as well as their long-chain PFCAs ($C \geq 8$), which were low in foliage translocation.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b05385.

> Detailed information on sample pretreatment, instrumental analysis, and the landfill description. The commercial sources of the target analytes (Table S1); the geographic information on the sample sites (Table S2); the corresponding surrogates used to normalize each target analyte (Table S3); quantitative ion, instrumental limits of quantitation (LOQs), and method detection limits (MDLs) (Table S4); the matrix recoveries and detection frequencies of target analytes for leaf, air, and particulate matter samples (Table S5); the concentrations of PFASs in the air cited from the previous literature (Table S6); the particle-gas distribution ratio (K_{pa}) of PFASs (Table S7); the map of sampling sites (Figure S1); the spatial distributions of neutral and ionizable PFASs in the air (Figure S2); the total concentrations and spatial distributions of PFASs in dry deposition (Figure S3); the concentrations of neutral PFASs in leaves (Figure S4); the spatial distributions of neutral and ionizable PFASs in leaves (Figure S5); and the concentrations and spatial distributions of C2−C3 PFCAs in the environment (Figure S6) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: 86-22-23509241; e-mail: sunhongwen@nankai.edu.cn (H.S.).

ORCID

Fengchang Wu: 0000-0003-2615-2849
Hongwen Sun: 0000-0003-1948-5047

Notes

The authors declare no competing financial interest.

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JOURNAL ARTICLE

Per- and Polyfluoroalkyl Substance Toxicity and Human Health Review: Current State of Knowledge and Strategies for Informing Future Research

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Suzanne E. Fenton , Alan Ducatman , Alan Boobis , Jamie C. DeWitt , Christopher Lau , Carla Ng , James S. Smith , Stephen M. Roberts ✉

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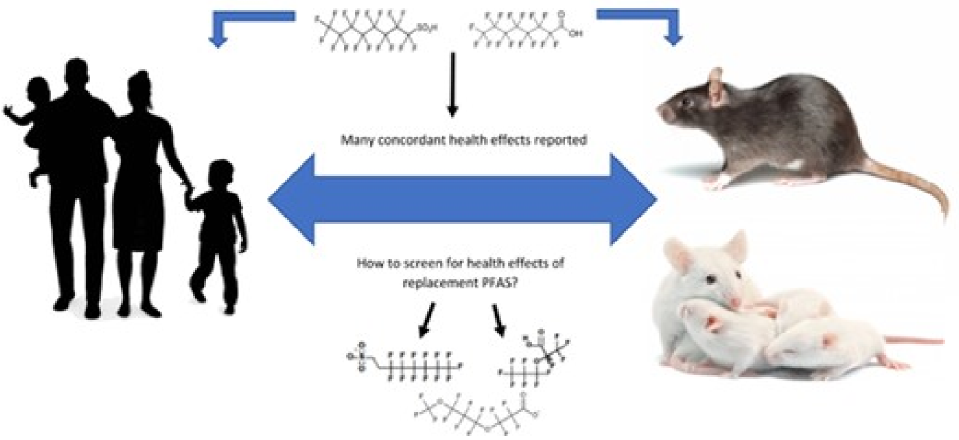
Abstract

Reports of environmental and human health impacts of per- and polyfluoroalkyl substances (PFAS) have greatly increased in the peer-reviewed literature. The goals of the present review are to assess the state of the science regarding toxicological effects of PFAS and to develop strategies for advancing knowledge on the health effects of this large family of chemicals. Currently, much of the toxicity data available for PFAS are for a handful of chemicals, primarily legacy PFAS such as perfluorooctanoic acid and perfluorooctane sulfonate. Epidemiological studies have revealed associations between exposure to specific PFAS and a variety of health effects, including altered immune and thyroid function, liver disease, lipid and insulin dysregulation, kidney disease, adverse reproductive and developmental outcomes, and cancer. Concordance with experimental animal data exists for many of these effects. However, information on modes of action and adverse outcome pathways must be expanded, and profound differences in PFAS toxicokinetic properties must be considered in understanding differences in responses between the sexes and among species and life stages. With many health effects noted for a relatively few example compounds and hundreds of other PFAS in commerce lacking toxicity data, more contemporary and high-throughput approaches such as read-across, molecular dynamics, and protein modeling are proposed to accelerate the development of toxicity information on emerging and legacy PFAS, individually and as mixtures. In addition, an appropriate degree of precaution, given what is already known from the PFAS examples noted, may be needed to protect human health. *Environ Toxicol Chem* 2021;40:606–630. © 2020 SETAC

Abstract

Many health effects have been reported in association with or due to per- and polyfluoroalkyl substance (PFAS) exposures in humans and toxicologic models. Species concordance of effects is evident for a handful of legacy PFAS. With hundreds of PFAS in commerce that lack exposure and health effects data, contemporary and novel methods must be implemented to inform exposed communities, risk assessors, and concerned citizens and prioritize those most likely to affect human health.



Keywords: Per- and polyfluoroalkyl substances, Perfluorooctane sulfonate, Perfluorooctanoic acid, Persistent compounds, Contaminants of emerging concern

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Review

Per- and polyfluoroalkyl substances (PFAS) in livestock and game species: A review

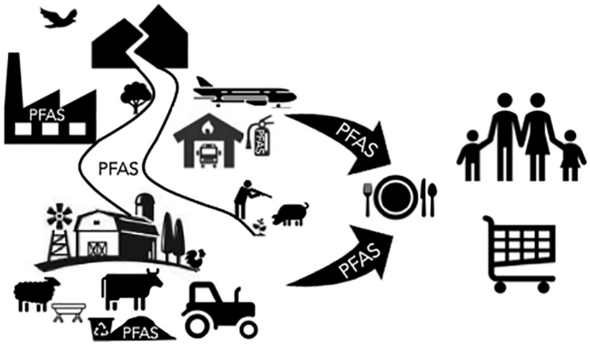
Clare Death [a,*], Cameron Bell [a], David Champness [a], Charles Milne [a], Suzie Reichman [b], Tarah Hagen [c]

[a] *Agriculture Victoria, 475 Mickleham Road, Attwood, Victoria 3049, Australia*
[b] *Centre for Anthropogenic Pollution Impact and Management (CAPIM), School of Biosciences, The University of Melbourne, Parkville, Victoria 3010, Australia*
[c] *SLR Consulting Australia Pty Ltd, Lvl 11, 176 Wellington Parade, East Melbourne, Victoria 3002, Australia*

HIGHLIGHTS

- There are a limited number of small studies investigating PFAS in livestock and game.
- Tissue distribution and elimination of PFAAs varies markedly between animal species.
- PFAA concentrations in meat samples are consistently lower than offal such as liver.
- PFAAs are transferred from exposed animals to offspring, milk and eggs.
- Concentrations of PFAAs in edible animal products decline after exposure ceases.

GRAPHICAL ABSTRACT



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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are synthetic, organic chemicals that resist environmental breakdown. The properties that made PFAS into an industrial success also led to their persistence and bioaccumulation. As PFAS were widely used for many decades their presence is evident globally, and their persistence and potential for toxicity create concern for human, animal and environmental health. Following the precautionary principle, a reduction in human exposure is generally recommended.

The most significant source of human exposure to PFAS is dietary intake (food and water) with additional exposure via dust. As PFAS concentrations have been more frequently studied in aquatic food sources, there is less understanding of exposure via terrestrial animals. To further define human exposure via animal products, it is necessary to determine PFAS concentrations and persistence in terrestrial livestock and game species. Studies assessing ambient concentrations of PFAS have noted that, aside from point sources of contamination, there is generally low input of PFAS into terrestrial agricultural food chains. However, livestock and game species may be exposed to PFAS via contaminated water, soil, substrate, air or food, and the contribution of these exposures to PFAS concentrations in food products is less well studied.

This review focuses on perfluoroalkyl substances (PFAAs) and compiles information from terrestrial livestock and game species as a source of dietary exposure in humans, and discusses toxicokinetics and health effects in animals, while identifying future focus areas. Publications describing the transfer of PFAAs to farmed and hunted animals are scarce, and demonstrate large variability in distribution and elimination. We outline several relatively small, short-term studies in cattle, sheep, pigs and poultry. While negative effects have not been noted, the poultry investigations were the only studies to explicitly assess health effects. Comparative information is presented on PFAA concentrations in livestock products and edible tissues of game animals.

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* Corresponding author.
E-mail address: clare@wildtox.com.au (C. Death).

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Contents

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a family of >4000 synthetic chemicals, used extensively throughout the world from the mid-twentieth century (Buck et al., 2011, Sunderland et al., 2019). PFAS are highly fluorinated aliphatic compounds, which differ in their functional groups and carbon chain length (Buck et al., 2011). Many PFAS can reduce surface tension, are resistant to degradation at high temperatures, and are water, oil and dirt repellent (OECD, 2013). These properties mean they have been widely used, for example in metal plating and coating agents, greases, lubricants, adhesives, paints, polishes, cleaning products, surfactants, photographic products, packaging, herbicides and insecticides, textile and leather products, and firefighting foams (OECD, 2013). PFAS resist biodegradation, photooxidation, and hydrolysis due to the strength of the carbon-fluorine bond (Sznajder-Katarzyńska et al., 2019). This review focuses on perfluoroalkyl acids (PFAAs) and, in particular, on the most frequently studied PFAAs: perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS). The majority of PFAS research and regulatory attention has been focused on PFOS and PFOA due to their frequent occurrence in the environment, known persistence, and bioaccumulative properties (CRC CARE, 2016).

PFOS and PFOA belong to the group of long perfluorocarbon chain PFAS and they can be the chemical breakdown product of other PFAS (Buck et al., 2011). There are less data available on the toxicity of PFAS other than PFOS and PFOA (ATSDR, 2018), nevertheless in the studies that are available the toxicity of shorter chain PFAS has been lower (e.g. Newsted et al., 2008; Lieder et al., 2009a, 2009b; Butenhoff et al., 2012a). Due to their widespread use, mobility, and persistence in the environment, PFAS are found in soil, surface water and groundwater in urban areas at low concentrations, and have been measured globally in a wide variety of marine and terrestrial animals, and humans (Buck et al., 2011, OECD, 2013). Globally, PFOS is the most prevalent PFAS found (Reiner and Place, 2015). Ecological studies in North America, Europe, Asia and remote polar regions have found PFOS in tissues of polar bears, river otters, albatrosses, bald eagles, fish, dolphins, penguins, and Arctic and Antarctic seals (e.g. Giesy and Kannan, 2001, Houde et al., 2011, Reiner and Place, 2015, Muir et al., 2019). Concentrations of PFOS in wild animals from relatively more populated and industrialised regions, such as the North American Great Lakes, Baltic Sea, and Mediterranean Sea, were greater than those in animals from remote marine locations (Giesy and Kannan, 2001). Honey samples originating from an industrial region of Poland showed 20% higher concentrations of perfluoroheptanoic acid (PFHpA) compared to those from non-industrial regions (Surma et al., 2016).

In 2009, PFOS and PFOA were listed under the *Stockholm Convention on Persistent Organic Pollutants* 'due to their demonstrated toxicity, bioaccumulation, persistence in the environment and ability to travel long distances from the point of release or application' (SC, 2019). This requires participating countries to eliminate or reduce the release of these chemicals into the environment. Manufacturing of other PFAS has continued, with a shift in manufacturing to short-chain PFAS (e.g GenX), which has led to more frequent measurement of these chemicals in the environment, some of which also appear to be environmentally persistent (Sunderland et al., 2019). However, human serum concentrations of PFOS and PFOA have shown a downward trend worldwide since the 2000's (Kato et al., 2015).

In a recent global survey of inhalation, dietary and drinking water sources of PFAS exposures, Jian et al. (2017) noted that food and drinking water are still the main routes of human exposure. This followed assessment of PFAS profiles in indoor air and dust samples collected from home, office, and vehicles, in addition to food (vegetables, dairy products, beverages, eggs, meat products, fish, and shellfish) and drinking water. Dietary exposure to PFAS in fish and shellfish remain the most significant dietary source (Domingo and Nadal, 2017). However, one Canadian study showed that beef contributed around 90% of the total dietary intake of PFOS, with fish being of lower importance (Tittlemier et al., 2007). In a 2016 Australian report, which recorded the occurrence of PFAS in animal products sourced near contaminated sites, PFOS concentrations were highest in rabbit meat, finfish livers, cattle meat and mammalian offal (FSANZ, 2016). PFOA was below reporting limits in most samples, but where measurable it was highest in molluscs and freshwater fish (FSANZ, 2016). There are currently no specific regulatory limits for PFAS in food in any country (FSANZ, 2017), but Tolerable Weekly Intakes (TWI) have been set in Europe (EFSA, 2020), there are guidelines for selected PFAS in drinking water (US EPA, 2016; US EPA, 2017; NHMRC, 2018), and some countries like Australia have also set 'trigger levels' for investigation of PFAS in food products and environmental media (FSANZ, 2017; HEPA, 2020).

PFAS have the potential to bioaccumulate and biomagnify in food webs (Kelly et al., 2009; Houde et al., 2011; Reiner and Place, 2015; Xiao, 2017). The rate of bioaccumulation has been shown to increase as the carbon chain length increases (Houde et al., 2011), supported by studies where bioaccumulation of PFOA is lower than for PFOS (Conder et al., 2008). For example in sheep (Kowalczyk et al., 2012), similar to rats (Cui et al., 2010), PFOA had a higher elimination rate than PFOS. In addition, it has been shown that perfluoroalkyl sulfonic acids (PFSAs) (as compared to perfluoroalkyl carboxylic acids, PFCAs, of the same perfluorinated chain length (see **Table S1** for description of chemical grouping)) accumulate in organisms to a higher degree due to their differing functional groups (Zhao et al., 2012; Conder et al., 2008). Müller et al. (2011) studied a lichen-caribou-wolf terrestrial food web and found that trophic magnification factors were highest for PFAS with nine to eleven carbons, that PFOA did not significantly biomagnify, and that magnification factors were around two times lower than in the marine environment.

Studies have shown large variability in PFAS elimination half-lives between species and chemical type (Kudo, 2015). As these compounds are proteinophilic, PFAS concentrations are generally highest in blood and liver, followed by varying concentrations in bile, kidney, lung, skin, muscle, fat and brain, which vary with species and chemical (Pizzurro et al., 2019; Kudo, 2015). Some PFAS are transferred via the placenta, milk (Kato et al., 2015; Pizzurro et al., 2019) and eggs (Wilson et al., 2020), which reflect direct pathways of PFAS into animal products for human consumption.

While this review includes publicly available PFAS data from terrestrial livestock and game species, most studies have focused exclusively on PFAAs. The term PFAS is used here to be inclusive of PFAAs and other *per-* and polyfluoroalkyl substances. The available studies are outlined by species, and a summary of PFAS concentrations in retail samples of livestock products or edible tissues of game animals (e.g. wild boar, deer, quail and ducks) is provided.

2. Sources of PFAS in livestock and game species

2.1. Contaminated air and water

PFAS can be transported long distances in dust (OECD, 2002; US EPA, 2017). In general, concentrations of PFAS in indoor air and dust samples significantly exceed those found outdoors when an industrial point source is absent, for example home and office PFOS concentrations in Europe, South Korea and North America ranged between <1.0–400 pg/m^3 and outdoor concentrations ranged between <1.0–150 pg/m^3 (Goosey and Harrad, 2012). Nonetheless, studies in China have shown that total PFAS concentrations in monkey blood samples near urbanised areas (i.e. zoo animals) were one order of magnitude higher than the concentrations in wild monkeys (i.e. nature reserve animals) and that tree leaves accounted for the highest percentage of daily intake (Cui et al., 2019), likely due to airborne deposition. This suggests that food-producing animals in peri-urban areas may also have higher airborne PFAS exposure when compared to those from remote areas, due to a higher density of sources such as landfills and industry.

In relation to exposure via water, reviews of the range of ambient PFOS concentrations in drinking water in Japan (Guruge et al., 2008), Australia (Thompson et al., 2011) and Europe (EFSA, 2018) have reported concentrations varying from <0.1–51 ng/L. Global drinking water PFAS concentrations have been reviewed recently by Rahman et al. (2014) and Domingo and Nadal (2019). PFAS have been measured in sediments in bays, lakes and rivers, and in effluents from sewage treatment plants (OECD, 2002; Ahrens et al., 2009; Möller et al., 2010). An example of surface water PFOS concentrations downstream from wastewater treatment facilities includes concentrations in Japan up to 157 ng/L (Guruge et al., 2008). Increased water concentrations occur downstream of PFAS production sites (Boiteux et al., 2017) and sites where historical use of aqueous film forming foams (AFFF) occurred for fire-fighting purposes (D'Agostino and Mabury, 2017), as well as downstream of landfills (for example, concentrations in leachate from an Australian study were PFOA max. 48 ng/L, and PFOS max. 240 ng/L; Gallen et al., 2017). PFOS and PFOA have surface water half-lives of 41 years and 92 years respectively (DoER, 2016) and nanofiltration may be required to remove PFAS from treated water (Domingo and Nadal, 2019). In Australia, the principal source of PFAS contamination in livestock has been due to stock drinking water contaminated via stormwater runoff from historic use of AFFF at neighbouring sites (Australian Government, 2020).

2.2. Contaminated soil, pasture or substrate

Globally, many regions are continuing to discover PFAS contaminated sites from AFFF use, particularly next to airports, fire training areas and military bases (Sunderland et al., 2019; DoD, 2020). Background PFAS concentrations in soil versus concentrations seen at contaminated sites globally have recently been reviewed by Brusseau et al. (2020). Concentrations reported for PFAS-contaminated sites were generally orders-of-magnitude greater than background concentrations, particularly for PFOS, which ranged upwards of several hundred mg/kg (Brusseau et al., 2020). Among other PFAS, PFOA and PFOS are found in sludge or biosolids from wastewater treatment plants with 80–100% detection frequency, thus application to pastures raises concern about accumulation in the edible tissues of food animals (Lupton et al., 2011, 2014, Venkatesan and Halden, 2013). PFAS concentrations in biosolids have been found to range from 1 to 244 μg/kg dry weight (dw) PFOA and 5–3120 μg/kg dw PFOS in the USA (Lupton et al., 2011, 2014), from 4.3 to 89 μg/kg dw PFOS in Italy (Brambilla et al., 2016) and between 4.7 and 86 μg/kg dw PFOS in Australia (Sleep and Juhasz, 2020). Concentrations of PFOS in soils of biosolid-amended fields have been measured up to 483 μg/kg dw (Sepulvado et al., 2011).

Animals consume soil while they are grazing and plants also uptake PFAS from soils; multiple studies have demonstrated that the amount of PFAS in plants is directly proportional to soil concentrations (e.g. Stahl et al., 2009), but uptake occurs to different extents according to their concentrations, chain lengths, functional group, plant species and variety, growth media (hydroponics vs. soil), and soil characteristics, primarily soil organic matter (Ghisi et al., 2019). Example concentrations in potential livestock feed products grown in soil spiked with 1 mg/kg of each compound include rye grass: PFOA 408–7250 μg/kg dw and PFOS 92–470 μg/kg dw, and wheat straw: PFOA 1900 and PFOS 270 μg/kg dw (Stahl et al., 2009). In forage grown on biosolid-amended soil, concentrations of PFOS and PFOA have been measured at 1–20 μg/kg and 10–1200 μg/kg dw respectively (Yoo et al., 2011). Fernandes et al. (2019) found evidence of PFAS uptake into pig liver from biosolids exposure, and also demonstrated that PFAS concentrations in eggs showed evidence of uptake from chicken exposure to recycled cardboard, dried paper pulp and wood shavings used as substrate/bedding. This indicates that long-term exposure to PFAS from soil and forage is important to consider in livestock grazed on pastures with ongoing application of biosolids (Lupton et al., 2014), and that care must be taken with use of recycled materials as substrates in the animal industries (Fernandes et al., 2019).

In 2018, the European Food Safety Authority (EFSA) reported modelling where forages represented 78% of PFAS exposure in ruminants, while soil accounted for >80% in outdoor poultry/eggs and pigs (Brambilla et al., 2015). This proportionality would clearly change depending on the PFAS concentrations in each exposure medium (water, soil, grass) at a particular location of interest, as well as with housing and rotation practices of livestock, and feeding habits. As an example of dietary PFAS exposure from processed animal feed other than fishmeal, a Turkish study of PFAS concentrations in commercial feed for layer hens, cattle and sheep found PFOA and PFOS concentrations up to 7.55 μg/kg ww and 0.88 μg/kg ww respectively (Onel et al., 2018).

2.3. High-energy rations and fishmeal

Some PFAS have been shown to bioconcentrate in fish (US EPA, 2017). Thus, mixed feeds for animals originating from fish may contribute to higher PFAS exposure in some farm animal species (Guruge et al., 2008). A study from Japan suggested that use of feeds with animal and fish products for fattening could explain accumulation of PFOS in beef cattle (Guruge et al., 2008). Li et al. (2019a) collected 92 commercial fishmeal samples from leading fishmeal-producing countries and found that the sum concentration of 16 common PFAAs ranged from 0.65 to 85.5 μg/kg (mean: 18.2 μg/kg, 12% moisture). PFOS predominated, with high detection of perfluoroundecanoic acid (PFUnDA), and wide occurrence of short-chain PFAS (e.g. PFBA, PFBS) (Li et al., 2019a). The total concentrations of PFAA in fishmeal were significantly higher in products originating from fish in the northern compared to the southern hemisphere (Li et al., 2019a), which correlates with modelled ocean PFAA concentrations (Muir et al., 2019).

2.4. Transplacental and lactational transfer

Studies of humans and laboratory animals demonstrate that both PFOA and PFOS in maternal plasma can cross the placenta and can also enter breast milk (Kato et al., 2015; Pizzurro et al., 2019; ATSDR, 2018) and that the ratio of PFOS concentrations between maternal serum:milk:cord blood are comparable across species (van Asselt et al., 2013). Guruge et al. (2008) found high concentrations of PFOS in cattle foetal livers, indicating that PFOS crosses the placental barrier to enter foetal circulation. Following birth, lactational transfer has been shown to occur in humans and mice (Lau, 2015) and studies in PFAS exposed cattle and sheep have demonstrated PFAS transfer from feed to milk (Kowalczyk et al., 2012, 2013), so this is likely to be the same in livestock and game species.

3. PFAS toxicokinetics

PFAAs are generally well absorbed from the gastrointestinal tract (e.g. >90% absorption of PFOA and PFOS in rats) and are not metabolised (Kudo, 2015, Pizzurro et al., 2019). As they are non-volatile and metabolically inert, body clearance of PFAAs depends on elimination into urine and faeces, and the enterohepatic circulation that occurs following excretion into bile may extend half-life (Kudo, 2015). In contrast to neutral hydrophobic organic contaminants (e.g. polychlorinated biphenyls or PCBs), which are primarily accumulated in adipose tissue (Conder et al., 2008), PFAAs are distributed mainly to the serum, liver, and kidneys (ATSDR, 2018, Kudo, 2015, Pizzurro et al., 2019) and their bioaccumulation potential cannot be predicted by traditional approaches (Conder et al., 2008). Toxicokinetic studies in rats demonstrated that different homologues of the two major groups of PFAA, PFCAs and PFSAs, have different urinary clearance rates (Ohmori et al., 2003). Renal elimination is the most critical process in determining total body clearance and biological half-lives of PFAS (Kudo, 2015).

Large differences in elimination rates of PFAS have been observed within and between different species (Lau, 2015, see Table 1), with the longest half-lives seen in humans (Pizzurro et al., 2019). Half-lives vary with chemical, and are longer for the eight-carbon versus four-carbon PFAS, and longer for the sulfonates compared to the carboxylates (Pizzurro et al., 2019). Human, mouse, rat and monkey studies show that half-lives decrease in the order of PFHxS > PFOS > PFOA > PFBS > PFBA but the longer half-life estimate for PFHxS may be related to uncertainty in the assumptions used to derive the estimate (Pizzurro et al., 2019). There have been relatively few studies in livestock. The variation in half-lives between species is large and ranges from hours e.g. PFOA in rabbits (Hundley et al., 2006), to days or weeks e.g. PFOS and PFOA in mice (Chang et al., 2012; Lou et al., 2009) and birds (Newsted et al., 2007; Wilson et al., 2020; Tarazona et al., 2015), then up to several months e.g. PFOS in monkeys (Chang et al., 2012; Seacat et al., 2002), and finally years for PFOS in pigs (Numata et al., 2014) and humans (Spliethoff et al., 2008; Glynn et al., 2012). Studies of PFAS in rats have also demonstrated differences in elimination half-lives between sexes (e.g. Chang et al., 2012) however further work is required to explain these gender differences.

Tissue to serum partition coefficients of PFAS vary by chemical type, and between species (**Table S2**). For example while PFOS and longer chain PFAS bioaccumulate and persist in protein-rich compartments (liver, blood) (Kelly et al., 2009), partitioning between the blood and the liver is highly specific to animal species (Guruge et al., 2008). The evidence in humans and experimental mammals suggests that PFOA, PFOS, and PFBS preferentially distribute to the liver, whereas PFBA and PFHxS appear to preferentially distribute to the serum (Pizzurro et al., 2019). While PFAS are proteinophilic and their concentrations exhibit positive correlation with protein content of tissues, rather than fat content (Kelly et al., 2009), PFAS are found in fat tissue to varying extents and cannot all be thought of as being lipophobic (Numata et al., 2014). In humans, the long half-lives of PFOS and PFOA appear to arise from the processes of enterohepatic recycling and of saturable resorption from the kidney (Roberts, 2016). Overall, studies demonstrate varying levels of excretion and accumulation of PFAS, dependent on both the chemical and the animal species. Several studies have demonstrated that short chain PFAS compounds (such as PFBS), which show different elimination kinetics due to smaller molecular size, higher water solubility and reduced protein binding, have demonstrably shorter half-lives (e.g. Kowalczyk et al., 2013). Riebe et al. (2016) proposed that the low PFOA concentration in herbivorous and carnivorous species in their study, compared with the mean PFOA concentrations seen in wild

Table 1

Serum/plasma elimination half-lives of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) across experimental, game and livestock species.
Modified from Lau (2015) and ToxConsult (2016 a,b).

	PFOS		PFOA	
	Female	Male	Female	Male
Rabbit	88 days[a]		7 h[b]	5.5 h[b]
Rat	14–83 days[c,d]	7.5–82 days[e,d]	2–16 h[f]	1.3–21 days[g,d]
Mallard duck		13.6 days[h]		
Quail		20.7 days[h]		
Chicken	3.5–160 days[i,j]			4.6 days[k]
Mouse	31–38 days[k]	36–43 days[l]	16 days[m]	22 days[m]
Dog			8–13 days[n]	20–30 days[n]
Cattle	39–106 days[o]	120 days[p]	1.3 days[o]	19.2 h[q]
Sheep	No data	No data	No data	No data
Monkey	110–200 days[k,r]	132–200 days[l,r]	33 days[s]	21 days[s]
Pig	1.7 years[t]		236 days[t]	

[a] Tarazona et al. (2016). [b] Hundley et al. (2006). [c] Wang et al. (2010a, 2010b). [d] De Silva et al. (2009). [e] Johnson and Ober (1979). [f] Kemper (2003). [g] Benskin et al. (2009). [h] Newsted et al. (2007). [i] Wilson et al. (2020) (laying hens). [j] Tarazona et al. (2015) (8-wk old males). [k] Yoo et al. (2009). [l] Chang et al. (2012). [m] Lou et al. (2009). [n] Hanhijärvi et al. (1988). [o] Vestergren et al. (2013). [p] Lupton et al. (2015). [q] Lupton et al. (2011). [r] Seacat et al. (2002). [s] Butenhoff et al. (2004). [t] Numata et al. (2014).

boar, suggests an omnivorous diet could play an important role in the uptake of PFAS, which may be a result of the affinity of PFAS for protein and therefore animal tissues.

4. Health effects of PFAS in animals

Although concern has been raised over the toxic effects seen in experimental animal studies at high PFAS doses, it is presently unclear whether these findings are relevant in livestock and game species at environmentally relevant exposure concentrations. The primary effects observed in laboratory animals exposed to perfluoroalkyl compounds (generally at doses higher than human exposure concentrations) are liver toxicity, developmental toxicity and immune toxicity (ATSDR, 2018). Experimental exposure to PFOS results in accumulation mostly in the serum and liver (OECD, 2002), with evidence of liver toxicity at high oral doses of PFOS (e.g. 0.75 mg/kg in monkeys) (Seacat et al., 2002) and PFOA (e.g. 5 mg/kg in mice) (Crebelli et al., 2019). Studies in monkeys and rats have shown a decrease in serum cholesterol concentrations, weight loss, and detrimental effects on glycogen metabolism (Seacat et al., 2002; Jiang et al., 2015). Also in rats, carcinogenicity (e.g. testicular, liver and pancreas tumours) has been demonstrated in some sub-acute and chronic PFOS and PFOA studies at high oral doses (e.g. 5-20 mg/kg in feed) (Biegel et al., 2001; Butenhoff et al., 2012b; NTP, 2020; Thomford, 2002). There is evidence of reproductive (Lu et al., 2019) and developmental toxicity in rats, mice and rabbits (OECD, 2002; Lau et al., 2003; Thibodeaux et al., 2003; Luebker et al., 2005a, 2005b; Wang et al., 2010a; Li et al., 2019b), and altered thyroid hormone concentrations in monkeys (Seacat et al., 2002). These types of health effects have not been investigated or reported in livestock or game species, and the variability seen in experimental animal species suggests a wide range of potential effects, over a range of exposure doses and time frames, need to be considered in future PFAS studies in food-producing animals. Due to the large differences in PFAS toxicokinetics between species, blood serum (or plasma) PFAS concentrations are a better indicator than external dose for comparing adverse effects between different species (Pizzurro et al., 2019).

From many experimental studies, it appears that animals can tolerate extremely high concentrations of PFAS. Recorded PFOS 'No Observable Adverse Effect Levels' (NOAELs) in experimental animals include blood concentrations of 67 mg/L in monkeys (Seacat et al., 2002) and 40 mg/L in rats (Luebker et al., 2005a, 2005b; ToxConsult, 2016a). In livestock, detrimental health effects were not reported, and are therefore assumed not to have been observed, in multiple short-term experimental studies at similarly high maximum observed blood plasma PFOS concentrations, including up to 0.24 mg/L in sheep (Kowalczyk et al., 2012), 0.25 mg/L in pigs (Kowalczyk, 2014) and between 2.46 mg/L (Kowalczyk et al., 2013) and 76.3 mg/L (Lupton et al., 2015) in cattle. For context, the average human blood PFOS concentration from various studies in the United States between 2005 and 2009 ranged from 13.2 to 17.1 ng/mL (Kato et al., 2015).

Several studies have discussed health effects in birds. Although bird eggs have been used in biomonitoring studies on PFAS, and there are some indications of eggshell thinning and reduced hatching success in wild birds, effects of environmental PFAS concentrations on avian reproduction remain largely unknown (Custer et al., 2014; Groffen et al., 2019). Some avian studies have demonstrated higher plasma oxidative damage and lowered plasma antioxidant defences secondary to longer-chain PFAS exposure, which has the potential to affect key fitness traits such as reproduction (Costantini et al., 2019). The acute and chronic effects of PFOS (Newsted et al., 2006, 2007) and PFBS (Newsted et al., 2008) in mallard ducks and northern bobwhite quail have been studied using high doses in experimental settings, in which adult health, body and liver weight, feed consumption, gross morphology and histology of body organs, and reproduction were examined (for half-life results see Table 1). These studies show that high doses can be tolerated in these birds, with effects not reported below the 10 mg/kg of PFOS in feed dose (Newsted et al., 2007, 2008). Exposure concentrations of ≥10 mg/kg in feed showed decreased survivorship of quail offspring, a greater incidence of small testes length (Newsted et al., 2008) and an increase in quail, but not mallard, liver weight (Newsted et al., 2008). In the acute and chronic PFBS studies, the only detrimental effect was reduced body weight gains at doses >5620 mg/kg feed (Newsted et al., 2008). These studies appear to support the hypothesis that shorter chain PFAS, like PFBS, are of lower toxicity.

There were no published studies found that were specifically designed to investigate health effects in terrestrial livestock species; experimental studies to date have focused on chemical uptake and clearance after relatively short repeat exposures (approx. 3 weeks to 3 months e.g. Kowalczyk et al., 2012, 2013, Kowalczyk, 2014) or high bolus doses (e.g. Lupton et al., 2014, 2015), so the reported tissue concentrations, elimination rates and health observations are more relevant to acute rather than chronic exposure situations. The only livestock studies that have actively assessed health effects, in addition to reporting tissue PFAS concentrations and elimination times, are the three main studies discussed below in poultry (Yeung et al., 2009; Yoo et al., 2009; Wilson et al., 2020) in which no adverse effects were reported. In one of the two studies assessing domestic pigs reviewed below (Numata et al., 2014), general health was assessed by daily observation and there were no reports of adverse effects. There was no discussion of health effects in the cattle and sheep studies reviewed (Kowalczyk et al., 2012, 2013; Lupton et al., 2014, 2015; Vestergren et al., 2013), and the assumption is that no overt adverse effects were observed.

5. PFAS studies in livestock and game species

5.1. Overview

There have been relatively few investigations of PFAS in livestock and game species, which complicates management of farm animal health and food safety regulation. PFOS, PFHxS and PFOA are the predominant PFAS that have been described in livestock and game studies. Some studies have assessed livestock tissue concentrations due to ambient exposure to various PFAS (e.g. Guruge et al., 2008; Vestergren et al., 2013). The absolute concentrations of PFOS seen in Swedish cattle (Vestergren et al., 2013) were substantially lower compared with Japan (Guruge et al., 2008), emphasising regional differences in the magnitude of ambient PFAS exposure for farmed animals. Guruge et al. (2008) collected blood and liver samples from multiple farm animal species. PFOS was measurable in all samples and was the most prominent PFAS found in farm animals, with chicken livers containing the highest mean PFOS concentration, followed by livers from pigs and cattle (Guruge et al., 2008). The presence of other PFAS in livestock (such as PFOA, PFNA, PFDA, PFUnDA and PFDoDA) was insignificant (Guruge et al., 2008).

Some of the studies in livestock described below have taken advantage of "naturally contaminated" forage, for example, following a case of illegal mixing of industrial waste with fertiliser. The contaminated forage was subsequently used for two seminal pilot studies in cattle and sheep (Kowalczyk et al., 2012, 2013), with doses estimated to be 10- to 100-fold less than those used for toxicokinteic studies in chickens and rats. The majority of experimental studies are, however, less reflective of ambient exposure and have looked instead at tissue concentrations and elimination half-lives after a single, large bolus dose of PFAS (e.g. Lupton et al., 2014, 2015), with some investigating exposure over several months (e.g. Zafeiraki et al., 2016b; Wilson et al., 2020).

5.2. Cattle studies

Cattle have been the subject of more PFAS studies than other livestock species, and this work has demonstrated that PFAS concentrations in exposed animals will be highest in blood, liver and kidney, with some accumulation in fat and muscle, and potential transfer to milk. Enterohepatic circulation of PFOS results in its prolonged presence in plasma, whereas PFOA is almost fully excreted (Lupton et al., 2014). In general, the shorter-chain PFAS like PFHxS and PFBS have lower accumulation potential in cattle compared to the long-chain compounds like PFOS (Lupton et al., 2011, 2014).

Vestergren et al. (2013) studied the bioaccumulation of PFAS in five dairy cows (<24 months of age) receiving feed and drinking water in Sweden with ambient concentrations of PFAS. Despite feed and water concentrations of PFOA being approximately double those of PFOS, tissue concentrations of PFOS were an order of magnitude higher (Vestergren et al., 2013). Mean PFOS concentrations were highest in whole blood, followed by liver and then muscle, and the authors concluded that long-chain PFAS have a relatively high potential for transfer to milk and beef from the diet of dairy cows (Vestergren et al., 2013).

Kowalczyk et al. (2013) investigated the transfer of perfluorobutanesulfonate (PFBS), PFHxS, PFOS and PFOA into tissue and milk of 6 Holstein dairy cows fed contaminated hay and silage. After 28 days, three cows were slaughtered while three underwent a 21-day depuration period prior to slaughter. Overall, PFBS, PFHxS, PFOS, and PFOA showed different kinetics, and different milk elimination patterns. In plasma, concentrations of PFBS and PFOA remained low, whereas PFHxS and PFOS continuously increased during the feeding period (Kowalczyk et al., 2013). After the dietary exposure had stopped, PFOS concentrations continued to increase in muscle, kidney, plasma and liver, whereas PFHxS decreased linearly during the depuration time (Kowalczyk et al., 2013). The highest PFHxS concentrations were detected in liver and kidney, with lowest total PFAS concentrations seen in muscle. At the end of the feeding study, cumulative secretion in milk was determined for PFOS (14 ± 3.6%) and PFHxS (2.5 ± 0.2%), whereas PBFS and PFOA were barely secreted into milk (Kowalczyk et al., 2013). Overall, the kinetics of PFOA were similar to those of PFBS and substantially differed from that of PFHxS and PFOS (Kowalczyk et al., 2013). The very low concentration of PFBS in plasma and milk, the relatively high urinary excretion, and only traces of PFBS in liver and kidney suggest that PFBS does not accumulate in the body of dairy cows (Kowalczyk et al., 2013). This study showed that the longer the carbon chain, the lower the elimination rate via urine and milk, corresponding to higher accumulation in tissue samples, however the kinetics of PFOA were more similar to the short chain PFBS, compared to PFOS and PFHxS (Kowalczyk et al., 2013).

van Asselt et al. (2013) developed a physiologically based pharmacokinetic (PBPK) model for transfer of PFOS from feed to milk using the data described above from Kowalczyck et al. (2013), and estimated the half-life of PFOS in milk (56 days). There was a high correlation between PFOS concentrations in blood and milk, and model calculations showed that once steady state is reached almost all ingested PFOS is excreted through the cows' milk, although parameter estimation was complicated as the experiments were shorter than the half-life (van Asselt et al., 2013).

Lupton et al. (2011, 2014, 2015) have performed multiple small trials in cattle using various doses of PFOS and PFOA. Lupton et al. (2011) studied the elimination of a single, high oral dose (1 mg/kg) of PFOA in four Angus steers in the USA and found that it was fully excreted in the urine within 9 days of dosing, with a plasma elimination half-life of <20 h. Although PFOA was rapidly absorbed, it was also rapidly excreted and did not persist in edible tissues (Lupton et al., 2011). This finding was supported by Kowalczyk et al. (2013), where negligible amounts of PFOA were observed in all tissue samples.

Following a single, high, oral dose (approx. 8 mg/kg) of PFOS in three Angus steers, Lupton et al. (2014) found that the major route for excretion was via faeces (11 ± 1.3%), with minimal excretion via urine (0.5 ± 0.07%) and tissue concentrations decreased in the order: liver > back fat > kidney > intraperitoneal fat > lung > spleen > muscle. The high PFOS concentrations in liver and bile, and the prolonged presence of PFOS in plasma, indicated the important role that enterohepatic circulation plays in PFOS fate and distribution (Lupton et al., 2014). Fat samples had consistently higher concentrations than muscle (Lupton et al., 2014), but these results are not typical, as other animal studies have observed low accumulation of PFOS in fat tissues (Yoo et al., 2009; Bogdanska et al., 2011). A large concentration of the initial dose was still in blood at 28 days (36%), followed by the carcass remainder (6%) and muscle tissue (4%) (Lupton et al., 2014). Approximately 39% of the dose was not accounted for in the mass balance, therefore they hypothesized that large compartments such as skin and bone could be pools where PFOS is distributed in cattle (Lupton et al., 2014). Bogdanska et al. (2011) analyzed the skin and bone of mice and observed concentrations similar to those observed in blood, indicating that distribution to these tissues could be a source of the unaccounted PFOS, which is supported by a finding that concentrations of PFOS in duck skin were higher than those in muscle (Senversa, 2018).

Two Angus steers given a single oral bolus dose (capsule; 0.098 mg/kg bw) of PFOS and slaughtered on day 343 showed similar average PFOS concentrations in liver and plasma (0.15 μg/g ww and 0.15 μg/mL), whereas muscle concentrations (0.005 μg/g ww) were much lower (Lupton et al., 2015). Heifers in the same study were given a much higher oral bolus dose of PFOS (9.1 mg/kg bw) and tissue concentrations on Day 343 ($n = 2$) remained highest in plasma (8.3 μg/mL), followed by liver (4.7 μg/g) and then muscle (0.28 μg/g ww) (Lupton et al., 2015). Plasma depletion half-lives for the steers and heifers were 120 ± 4.1 and 106 ± 23.1 d, respectively (Lupton et al., 2015).

Guruge et al. (2008) found a positive correlation between serum PFOS concentration and age in lactating Holstein cows. In Australia, serum samples from cattle that resided adjacent to a site with historical use of firefighting foam showed much higher serum PFOS concentrations in bulls that had been resident for a longer period (i.e. several years) compared to the cows and calves, but PFHxS concentrations between the groups did not differ (Senversa, 2018).

5.3. Sheep studies

A pilot study by Kowalczyk et al. (2012) in Germany demonstrated the transfer of PFOS from contaminated feed (corn silage cultivated on cropland where illegal waste had contaminated farmland) into sheep milk and meat. Two East Friesian sheep were fed PFAS (PFOS: 90 μg/kg dry matter, PFOA: 33 μg/kg dry matter) contaminated corn silage for 21 days and PFOS was excreted in milk at higher concentrations (0.2–19.2 μg/L) than PFOA (<0.2–1.3 μg/L) (Kowalczyk et al., 2012). Although the transfer was low over this short period, PFOS could be measured in milk, liver, kidney and muscle tissue (Kowalczyk et al., 2012). PFOA was excreted in the urine, but PFOS excretion was primarily via the faeces (Kowalczyk et al., 2012).

Zafeiraki et al. (2016a) sampled livers from sheep fed with grass obtained from a river floodplain in the Netherlands, with PFOS concentrations up to 0.5 μg/kg, compared to those fed clean grass. Some sheep were fed for 112 days and PFOS concentrations in liver reached 10.9 ng/g *w*/w, whereas animals switched to clean grass after 56 days of exposure showed a decrease in liver concentrations from 9.2 to 4.7 ng/g w/w after 64 and 112 days respectively (Zafeiraki et al., 2016a). The percentage of PFOS ingested and retained in the liver was estimated to be 12% at day 56, which reduced to 6% after another 56 days on clean grass (Zafeiraki et al., 2016a).

5.4. Poultry studies

Yeung et al. (2009) dosed juvenile male domestic chickens with 0, 0.1 or 1 mg/kg combined PFOS, PFOA and perfluorodecanoate (PFDA) via gavage three times a week for three weeks. After three weeks of exposure, half of the chicks were sacrificed, and the other half underwent depuration for a further three weeks. No dose-dependent statistically significant differences in body/organ weights were observed among treatment and control groups for the duration of the study, and histological and plasma biochemical parameters did not differ. The half-lives at the higher dose rate were 17 days for PFOS, 16 days for PFDA, and 3.9 days for PFOA (Yeung et al., 2009). The liver was the main target during exposure, and the blood was the main reservoir during depuration.

Yoo et al. (2009) exposed groups of six male chickens to two levels of PFOA (0.1 or 0.5 mg/mL) or PFOS (0.02 or 0.1 mg/mL) via subcutaneous osmotic pump for four weeks and then allowed them to depurate for an additional four weeks. This administration route is unusual and unlikely to reflect environmental exposures, however, these exposures did not cause any statistically significant changes in body index, clinical biochemistry or histology among treatments relative to the controls ($p = 0.05$), except that concentrations of total cholesterol and phospholipids were less in chickens exposed to PFOS. The elimination rate constant for PFOA was approximately six-fold greater than that of PFOS, and the greatest concentrations of PFOA and PFOS were found in kidney and liver, respectively (Yoo et al., 2009). In summary, in broilers, PFOS concentrations were higher in liver and blood compared to kidney, whereas PFOA concentrations were highest in kidney and it was eliminated faster (Yeung et al., 2009; Yoo et al., 2009).

A study in 119 × 30-week old layers dosed for two months with PFOS, PFOA, PFHxS and PFHxA (perfluorohexanoic acid) at up to 300 μg/L water did not demonstrate any negative health or productivity effects (Wilson et al., 2020). There was a linear correlation between the PFAS concentrations in the drinking water of hens and those detected in the egg (Wilson et al., 2020). The PFAS elimination half-life in eggs measured over the study period (collected every second day), calculated as the rate at which the PFAS concentrations in eggs decreased after PFAS exposure to the hen via drinking water ceased, also referred as the "clearance phase", was 7 d for PFHxS, 5.4 d for PFOA, 3.5 d for PFOS and 2 d for PFHxA (Wilson et al., 2020).

Some egg injection studies have shown that PFOS is embryotoxic to domestic chickens (*Gallus gallus domesticus*). In a study by O'Brien et al. (2009), in which chicken eggs were directly injected with 0.1, 5, or 100 μg PFOS/egg into the air cell prior to incubation, the embryo median lethal dose (LD50) was 93 μg/g, however egg injection studies have limited applicability to how embryos are exposed in the environment. Briels et al. (2018) demonstrated that when PFOS was injected into chicken eggs prior to incubation, embryonic survival was not affected, nor were there any effects detected on hatchling weight or oxidative stress parameters.

The poultry studies described above have explicitly assessed and reported health effects in exposed animals, in addition to PFAS tissue concentrations, and are the only livestock studies to have taken this approach.

5.5. Domestic pig studies

PFAS elimination half-lives in pigs are longer than in most animals reported in the literature. Numata et al. (2014) investigated the transfer of a mixture of PFAS from contaminated feed in Germany (hay and barley grown in contaminated soil; range 10–137 μg/kg of feed; fed for 21 d) into the edible tissues of 24 fattening pigs. As percentages of unexcreted PFAS, the substances accumulated in plasma (up to 51%), fat, and muscle tissues (collectively, meat 40–49%), liver (under 7%), and kidney (under 2%) for most substances; an exception was PFOS, with lower affinity for plasma (23%) and higher for liver (35%) (Numata et al., 2014). The authors developed a toxicokinetic model to quantify the absorption, distribution, and excretion of PFAS and to calculate elimination half-lives. PFHxA had the shortest half-life at 4.1 days, whereas the half-life for PFOS was 634 days (Numata et al., 2014). The elimination half-life was influenced by the end functional group, with sulfonic versus carboxylic acid end groups leading to much longer half-lives (irrespective of carbon chain length) (Numata et al., 2014).

Guruge et al. (2016) found the blood half-life after a single oral dose of a mixture of ten PFAS (3 mg/kg bw of each of 10 PFAS) in minipigs ranged from 1.6 to 86.6 days. The liver was the greatest site of accumulation of PFOS and longer chain PFAS such as perfluorodecanoic acid (PFD(*e*)A), perfluoroundecanoic acid (PFU(*nD*)A) and perfluorododecanoic acid (PFDoDA). The study authors observed an increasing accumulation trend of PFAS associated with the fluorinated carbon chain length and perfluorononanoic acid (PFNA), a 9-carbon PFAS, showed the highest body burden of the administered PFAS (Guruge et al., 2016).

A recent study investigated the tissue distribution of 8:2 fluorotelomer alcohol (8:2 FTOH) of which a primary metabolite is PFOA, after dosing 30 pigs (70 days old) at 5 mg/kg bw for one week (Xie et al., 2020). The parent compound was not detected in tissues 3 d after exposure cessation but the absolute half-life of PFOA in the kidney was 61.4 days, which was marginally longer than liver half-life, following a peak concentration in kidney and liver of 42 ± 7.0 μg/kg and 50 ± 4.8 μg/kg respectively (ww, SD) (Xie et al., 2020). PFOA concentrations in fat, lung and heart were similar after 21 days, and an order of magnitude higher than muscle concentrations (Xie et al., 2020).

5.6. Game bird studies

The dietary and migratory habits of game birds are important considerations for PFAS exposure, for example ducks that feed in the sediment layer are likely to have higher exposure, and determination of the origin of exposure is complex due to movement (Larson et al., 2018).

Work in free-ranging waterfowl is limited, with some studies in wild birds showing relatively low PFAS concentrations, such as mallard and pintail (*Anas acuta*) ducks in Japan (**Table S3**) (Taniyasu et al., 2003). High concentrations of PFAS have been found in liver and muscle samples of ducks hunted in contaminated Australian wetlands (Sharp et al., 2020). Pacific Black (*Anas superciliosa*) and Grey Teal (*Anas gracilis*) ducks from a wetland near a military base had PFOS and PFHxS detected in all liver and muscle samples, with PFOS concentrations approximately 100 times higher than PFHxS (**Table S3**; Senversa, 2018). These concentrations are similar to those seen in liver and serum from great tits (*Parus major*) roosting near a fluorochemical plant in Belgium (Dauwe et al., 2007). Almost all concentrations of PFAS other than PFOS and PFHxS (including PFOA) were below the LOR (0.0002 mg/kg) (Senversa, 2018).

A subsequent state-wide survey of 4 species of duck (Pacific Black duck, Grey Teal, Chestnut Teal (*Anas castanea*) and Pink-eared duck (*Malacorhynchus membraneaceus*)) was conducted across 19 wetlands in Victoria, Australia in 2018 and showed relatively lower tissue concentrations (**Table S3**), but resulted in an advisory notice to avoid or minimise consumption of ducks hunted at three wetlands (EPA, 2019; Sharp et al., 2020). PFOS + PFHxS were detected in 95% of liver samples, whereas PFOA was detected in 29% (Sharp et al., 2020). Relatively higher PFAS concentrations were observed in water and sediments at wetlands near sources of contamination, and even though PFAS concentrations in duck tissues did not necessarily correlate with the environmental samples at all sites, only ducks inhabiting wetlands near local sources of PFAS were deemed likely to pose a risk to consumers (Sharp et al., 2020). This study demonstrates the limitations of using abiotic PFAS criteria to assess risk to biota, due to the complexities of bioaccumulation, movement of animals and spatiotemporal variation (Sharp et al., 2020), which supports the need to sample animal tissues to assess exposure and effects.

5.7. Mammalian game species

Relatively high PFAS concentrations have been reported in European wild boar, with liver as the main site of accumulation. Wild boar feeding behaviour, which includes rooting in soil and access to dumpsites that often contain municipal waste, is likely to influence exposure (EFSA, 2018). Wild pigs ($n = 13$) sampled near an open waste dumping site in India showed an average PFOS concentration of 71 ± 70 (SD) μg/kg ww in adult female liver ($n = 7$) compared to concentrations at a reference site of 19 ± 8.6 (SD) μg/kg ww ($n = 4$) (Watanabe et al., 2010).

In 2014, PFOA and PFOS in the liver of German wild boars showed an average ratio of PFOS:PFOA concentration in liver of 21:1 (**Table S3**) (Kowalczyk et al., 2018). Stahl et al. (2012) also reported maximum liver PFOA and PFOS concentrations in German wild boar that were at least an order of magnitude higher than muscle concentrations (**Table S3**). In 2019, PFOS was detected in 25% of muscle samples from wild boar collected in North West Italy at concentrations lower than those reported from Germany (**Table S3**) (Arioli et al., 2019).

Müller et al. (2011) found that tissue distribution of PFAS in caribou/reindeer (*Rangifer tarandus*) from the Canadian Arctic was similar to that observed in dairy cows. Total PFAS concentrations in muscle and kidney were both approximately one order of magnitude lower than liver (Müller et al., 2011).

A retrospective German study demonstrated a reduction in the mean PFOS concentration in roe deer (*Capreolus capreolus*) liver from 9.2 μg/kg in 2000 to 1.8 μg/kg ww in 2010 (Falk et al., 2012). The reduction of PFOA was less conspicuous than that of PFOS, but was still significant (Falk et al., 2012). Livers from chamois (*Rupicapra rupicapra*) in Austria in 2016 showed similar mean PFOS concentrations to the 2010 German roe deer samples, with a detection rate of 91% (Riebe et al., 2016). In contrast in 2019, PFOS in muscle samples from 12 roe deer, 24 chamois and 23 red-deer (*Cervus elaphus*) hunted in North West Italy were below the LOQ of 150 pg/g ww (Arioli et al., 2019).

5.8. Synthesis of findings from studies in livestock

In livestock, detrimental health effects were not reported, and are therefore assumed not to have been observed, even at high maximum observed blood plasma PFOS concentrations, including up to 0.24 mg/L in sheep (Kowalczyk et al., 2012), 0.25 mg/L in pigs (Kowalczyk, 2014) and between 2.46 mg/L (Kowalczyk et al., 2013) and 76.3 mg/L (Lupton et al., 2015) in cattle.

While the studies outlined above showed similarities in overall PFAS tissue distribution between species, there are also notable differences that vary by species, to some extent by dose and, to a lesser extent, by sex. For example, in cattle, the shorter-chain PFAS like PFHxS and PFBS have lower accumulation potential compared to the long-chain compounds like PFOS (Lupton et al., 2011, 2014), whereas in chicken eggs PFHxS showed greater accumulation potential than PFOS (Wilson et al., 2020) and ongoing exposure is likely to offer partial explanation for some observed differences. Livestock studies consistently demonstrated that following experimental exposure in cattle (Kowalczyk et al., 2013; Lupton et al., 2014; Lupton et al., 2015), pigs (Numata et al., 2014; Guruge et al., 2016) and chickens (Guruge et al., 2008; Yeung et al., 2009; Yoo et al., 2009), PFAS concentrations in muscle, likely the most commonly consumed animal product, were consistently lower than those measured in offal (primarily liver and blood, but also kidney).

The variation in elimination half-lives within and between livestock and game species is also large and studies showed that elimination generally occurs more rapidly for PFOA compared to PFOS (Table 1). PFAS elimination half-lives in pigs are longer (236 days to 1.7 years, see Table 1) than in most animals reported in the literature (Numata et al., 2014, Guruge et al., 2016). Studies in cattle showed large variation (19.2 h to 3 months, see Table 1) depending on the PFAS chemical and dose, and the age and sex of the study subjects (Vestergren et al., 2013; Lupton et al., 2015; Lupton et al., 2011). Avian elimination of PFAS is more rapid (3.5–160 days, see Table 1) than mammals and in laying hens, PFAS transfer to eggs is almost exclusively as PFOS in the yolk (Wilson et al., 2020).

Experiments in sheep, chickens and steers showed an initial increase of PFOS concentrations in plasma after dosing stopped (Kowalczyk et al., 2012; Lupton et al., 2011; Yeung et al., 2009), indicating slow excretion, enterohepatic circulation, and continued release of accumulated chemical from other organs prior to steady state being achieved and the commencement of depuration. However, PFAS concentrations in these species began (i.e. within days (Yeung et al., 2009) to weeks (Kowalczyk et al., 2012)) relatively predictable declines soon after exposure ceased, providing a management opportunity for depuration with uncontaminated feed and water.

Excretion of some PFAS into eggs was demonstrated to be a major excretory pathway for laying hens, explaining the differences between elimination half-lives in female (Wilson et al., 2020) and male chickens (Tarazona et al., 2015). However, after PFAS exposure ceased in laying hens, concentrations in eggs declined to undetectable concentrations within several weeks (Wilson et al., 2020). While PFAS excretion in cattle and sheep milk has been demonstrated, PFAS is not preferentially excreted or concentrated in milk (van Asselt et al., 2013; Kowalczyk et al., 2012; Kowalczyk et al., 2013). For example, during the 21-day feeding period in two sheep, a total PFOS transfer into milk of ≤2% of the estimated intake dose was calculated (Kowalczyk et al., 2012).

6. PFAS in animal products - the food chain

Product sampling studies have concurred with the livestock experimental data in showing that muscle concentrations are lower than those measured in offal (see **Table S3**). While offal and blood are not very popular diet components, the propensity for PFAS to accumulate in these protein rich tissues warrants particular attention (Sznajder-Katarzyńska et al., 2019). A study by Hlouskova et al. (2013) of PFAS in a small sample of various food animals from four European countries showed that the PFAS concentrations in the analyzed food commodities from terrestrial species decreased in the following order: pig/bovine liver > egg > meat > dairy products (butter). Unfortunately, most animal product testing has occurred without any knowledge of antemortem PFAS exposure concentrations.

6.1. Liver

One of the reasons for the preferential accumulation of PFAS in liver is that it is the main site of plasma albumin synthesis, and PFAS bind very effectively to plasma albumin (Lau, 2015). In a recent Chinese study, beef liver showed mean total PFAS concentrations that were over 60-fold higher than beef muscle (**Table S3**) (Wang et al., 2017). In chickens, PFOS first accumulates in the liver, after which it is redistributed into the blood and kidney for elimination (Yoo et al., 2009; Yeung et al., 2009) and elimination via eggs is also very important (Wilson et al., 2020). A comparative study of Japanese farm animals exposed to ambient concentrations of PFAS showed that chicken liver contained the highest PFOS concentrations compared to cattle, pig and goat liver (Guruge et al., 2008). In pigs, PFAS that were not measurable in other tissues were still measurable in liver, so Numata et al. (2014) suggest sampling liver in porcine monitoring programs. A Chinese study from 2010 showed the total PFAS concentrations were 30 times higher in liver from pigs compared to chickens (**Table S3**) (Wang et al., 2010b). The relative proportions of PFOA and PFOS in pig liver samples have been different across studies, with Chen et al. (2018) finding much higher concentrations of PFOA than PFOS in Taiwanese liver samples, and Wang et al. (2010b) finding the opposite in Chinese retail samples.

Recently, EFSA (2018) reported that PFOS and PFOA concentrations were particularly high for liver samples of game mammals, especially wild boar, with the maximum PFOS concentrations being two orders of magnitude higher than PFOA concentrations (**Table S3**), and PFOA concentrations in the edible offal of European farmed animals were much lower than wild boar liver (EFSA, 2018). Zafeiraki et al. (2016a) sampled livers from farmed (i.e. ambient exposure only) sheep, horses, cows, pigs and chickens collected from the Dutch market. PFOS was the only measurable PFAS and its concentration was higher in free ranging animals like cows and sheep, nevertheless measured concentrations of

PFOS in the liver samples were very low (**Table S3**) (Zafeiraki et al., 2016a).

6.2. Blood

The two tissues with the higest PFAS concentrations are the liver and the blood (Kudo, 2015), and studies outlined above in livestock have shown this proportion varies bewteen species. Experimental studies in cattle have shown PFOS to have the highest relative plasma concentrations of studied PFAS compounds (Lupton et al., 2015). In a Japanese study, mean sera concentrations of PFOS were seen in descending order in chickens > cattle > and pigs (**Table S3**) (Guruge et al., 2008). Numata et al. (2014) showed that in pigs, compared to cows and sheep, PFAS had an even higher affinity for blood over other tissues. Male chickens, when compared to dairy cows, showed a greater tendency to accumulate PFOA and PFOS in the blood, and a more rapid elimination of PFOA from blood (Kowalczyk, 2014). The affinity of PFAS compounds for blood indicates that dietary practices where blood contributes a significant portion should be evaluated as part of determining human exposure risk.

6.3. Kidney

Across experimental animal species, concentrations of PFAS are generally lower in the kidney compared to the blood and liver (Roberts, 2016), but PFHxS concentrations in kidney were relatively higher than other tissues in one dairy cattle study (Kowalczyk et al., 2013). When comparing concentrations of PFAS in different livestock tissues, primarily in pigs (Wang et al., 2010b; Xie et al., 2020) and poultry (Yoo et al., 2009), there is a pattern of preferential binding of PFOA in renal tissues, compared to blood or liver. In 20 Chinese retail samples of pork kidney in 2010, the maximum PFOA concentration was an order of magnitude higher than the maximum PFOS concentration (**Table S3**) (Wang et al., 2010b).

6.4. Eggs

Newsted et al. (2007) noted from their studies in mallard ducks (*Anas platyrhynchos*) and northern bobwhite quail (*Colinus virginianus*) that due to transfer to eggs in female birds, concentrations of PFOS measured in the liver and blood at study termination were greater in male birds compared to female birds. Zafeiraki et al. (2016b) found that PFAS concentrations in yolk were higher in home produced eggs from the Netherlands and Greece compared to supermarket eggs, which they hypothesized was due to access to soil and kitchen waste. To assess the risk of PFAS exposure to consumers from eggs laid by backyard chickens, Wilson et al. (2020) studied 119, 30-week old layers dosed with up to 300 μg/L of PFAS in water. The study showed that the vast majority of the PFAS (PFOS, PFOA, PFHxS and PFHxA) excretion in eggs is via the yolk and that once chicken PFAS exposure ceases, PFAS concentrations in the eggs laid by those chickens progressively reduce to below the Laboratory Limit of Reporting (LOR) within two to three weeks (Wilson et al., 2020). Wang et al. (2010b) also reported that close to 100% of the PFOS in the egg was distributed in egg yolk.

6.5. Milk

PFAS transfer from feed to milk has been confirmed in cattle (van Asselt et al., 2013; Kowalczyk et al., 2013) and sheep (Kowalczyk et al., 2012) and these studies showed species-specific differences in the transfer of PFAS from serum to milk. For PFOA, Kowalczyk et al. (2012, 2013) state the milk/serum concentration ratio was higher in dairy cows compared with sheep. For PFOS, the ratio in sheep was approximately 0.06, compared to 0.013 in cattle (Kowalczyk et al., 2012, 2013). Different PFAS also showed quite different milk elimination patterns in both cattle and sheep; PFOS and PFHxS were excreted in milk at higher concentrations than PFBS and PFOA (Kowalczyk et al., 2012, 2013).

6.6. Muscle/meat

Measured concentrations of PFAS in muscle/meat are generally lower than concentrations in blood and offal. Kowalczyk et al. (2013) showed that in dairy cattle, PFOA concentrations in muscle were one hundredth of the concentrations in the liver, and PFOS concentrations in muscle were one tenth of the concentrations in the liver. In pigs, PFOS concentrations were an order of magnitude lower in pork muscle compared to liver, but PFOA concentrations were similar between these two tissues (**Table S3**) (Chen et al., 2018). In one study conducted in Australia, PFOS + PFHxS concentrations in duck breast muscle samples were approximately nine times lower than the corresponding liver concentrations (Senversa, 2018).

7. Conclusion

This review has compiled known information about PFAS in livestock and game species, as a source of dietary exposure in humans, and briefly summarised adverse health effects in experimental animal studies. Broader collection and publication of baseline data in livestock and game species is required to enable development of predictive models on the uptake and elimination of PFAS, and transfer to edible tissues. This would enable more accurate risk-based management of PFAS exposure in humans via livestock and game animals. One key gap in the literature has been that very few studies assessed if any detrimental effects on welfare or production occurred due to PFAS exposure. Thus it is suggested that future research measure both PFAS dynamics and potential detrimental health effects in livestock and game animals.

While the only conclusive evidence of health effects of PFAS in animals are from high dose studies and exposure scenarios, there remains concern globally about potential human and animal health effects of these widely distributed, persistent chemicals. Multiple studies have asserted that PFAS consumption by humans in food and water is the most significant route of exposure, and this is likely the same in livestock and game species. Applying the precautionary principle, food regulators wish to understand the potential risks to the human population from consumption of PFAS in livestock and game products.

As there are only two to five studies per livestock species published in the international scientific literature, with low sample sizes and dosing regimes that do not necessarily reflect environmental relevance, there is uncertainty with respect to the tissue distribution and clearance information. It is therefore difficult to assess human health risks associated with the consumption of livestock products with a high degree of certainty. More research into the concentrations of PFAS in livestock and game populations is required globally, to better understand the potential transfer of these chemicals into the human food chain. There are many PFAS, and PFAA precursors, which are known to be widely distributed in the environment that still require investigation in livestock and game species, and the use of total fluorine measurements could be explored. Further detailed studies into the pharmacokinetics of PFAS in livestock, and more comprehensive approaches to health assessment of exposed stock, are also necessary. In addition, assessment of new (or replacement) PFAS should continue, preferably prior to their large-scale use to ensure new PFAS chemicals are less harmful than the original longer perfluorocarbon chain compounds (e.g. PFOS and PFOA) (Briels et al., 2018).

Additional clearance and tissue distribution data in livestock are required to enable confident assessment of potential tissue concentrations via non-invasive serum sampling. Determination of serum/tissue partition coefficients enables prediction of body burdens of various PFAS from blood serum samples in various classes of stock. Such data enable appropriate management of PFAS exposed livestock. Generally, livestock will eliminate PFAS over time if the contaminated source (e.g. stock water) is removed (EPA Vic, 2020). It is important to understand how long it takes different PFAS in different species to reduce to a level that minimises the exposure to humans consuming the animal products (EPA Vic, 2020).

Understanding concentrations of PFAS in the edible tissues of game species is also important for public health, and data currently being collected at contaminated locations will enable informed management of hunting. From an animal health perspective, further work will be needed to demonstrate whether PFAS cause toxic effects on free-living vertebrates (Costantini et al., 2019). Collection of additional data in livestock and game species (and their relevant exposure media) could also enable development of pharmacokinetic/pharmacodynamic models on the oral uptake and elimination of PFAS in different tissues, which could then be used to potentially estimate uptake and concentrations in edible tissues (van Asselt et al., 2011).

Once the transfer of PFAS through the food chain can be quantified, it will enable the effect of future pollution events on the consumer to be characterised using parameterised food chain models, which would allow the responsible authorities to take appropriate measures to ensure consumer confidence and health.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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