Exhibit D



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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

GRAPHICAL ABSTRACT

- 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.



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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

Corresponding author.

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

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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; H₂S, 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.

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 riskbased 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 PFAScontaining 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 s	elect PFAS in	ı landfill le	eachate and	US EPA	risk-based	thresholds
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0	ξŲ, β												
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
	Tapwater RSL $(HQ = 1.0)$	60		40		59		6000		390		n/a	n/a
EPA limit (ng L^{-1})	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 PFAScontaining 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 μ g 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 $380 \ \mu g \ kg^{-1}$ 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} 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 μ g 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 $\mu g kg^{-1}$ 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 μ g kg⁻¹ (median: 0.4 μ g kg⁻¹) in electronic and electrical equipment (EEE) and as high as 2.2 μ g kg⁻¹ (median: 1.4 μ g 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 $\mu g \; kg^{-1}$ in fly ash and from 3.1 to 77 μ g 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 μ g 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 μ g 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 μ g 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 μ g kg⁻¹, respectively, with concentrations as high as 50,000 μ g kg⁻¹ for PFOA and 460,000 μ g 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 biosolids composting, especially short-chain aerobic (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 physicalchemical 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 \sum 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

Table 2

Number of PFAS measured and \sum PFAS among published landfill leachate studies.

average is notably similar to the estimated average \sum PFAS concentration reported by Lang et al. (2017) using Monte Carlo simulation. Often, the \sum 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 \sum 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).

Matrix	Number of samples	Number of PFAS detected (in Method)	Average \sum PFAS (ng L ⁻¹)	\sum PFAS range (ng L ⁻¹)	Country	Reference
	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)
MSW LL	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 \sum 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 STerminal and SPrecursor 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 Σ 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 $>2000 \text{ ng L}^{-1}$ 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 $\sum_{24} \text{PFAS}$ 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 (CH₄), and 50% carbon dioxide (CO₂), 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 H₂S_(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
	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)
Separation technologies	Ion exchange resins	High maturity levelCan 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 Uses additional chemicals for		
	Chemical Oxidation	•Controllable by varying pH and temperatures	 bot identifiant elements 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)
Destruction technologies	Electrochemical	 98–99.7% effectiveness demonstrated Operates at ambient temperatures No chemicals required Lower energy consumption compared to incineration 	Expensive electrode materialsPerchlorates 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 commercializationLow 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 Unlikely to be effective 		Berhanu et al. (2023); Huang and Jaffé (2019)
	Constructed wetlands	•No demonstrated benefits for PFAS treatment	 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 RCRAcompliant 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 μ g 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 ground-water 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 m³ 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 m³ 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_0) of



Fig. 3. Flowchart depiction of annual \sum 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 m³ 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 PFAScontaining 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

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