

From: [Bruce Cowger](#)
To: [Coffin Butte Landfill Appeals](#)
Cc: [Bruce Cowger](#)
Subject: Testimony regarding LU-24-027: PFA forever chemicals and CBL
Date: Sunday, October 5, 2025 8:16:25 AM
Attachments: [A critical review of perfluoroalkyl and polyfluoroalkyl substances \(PFAS\) landfill disposal in the United States - full article.pdf](#)
[Per- and polyfluoroalkyl substances \(PFAS\) distribution in landfill gas collection systems - leachate and gas condensate partitioning - ScienceDirect.pdf](#)
[Some landfill 'burps' contain airborne PFAS, study finds - American Chemical Society.pdf](#)
[Volatile and semi-volatile organic compounds in landfill gas - Composition characteristics and health risks - ScienceDirect.pdf](#)
[What happens in a landfill, doesn't stay in a landfill - Washington State Department of Ecology.pdf](#)

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To: Benton County Board of Commissioners

From: Bruce Cowger 37194 Helm Drive, Corvallis, OR 97330

Subject: Testimony regarding LU-24-027, the application to expand the Coffin Butte landfill

I strongly urge Benton County's Commissioners to reject Republics' bid to expand the Coffin Butte Landfill. The siting of a solid waste landfill in a wet climate was a mistake, one that should be rectified as soon as possible so as to reduce the emission of hazardous pollutants.

I have a Physics degree (1978, Willamette University) and a Mechanical Engineering Degree (1978, Stanford University).

Three of the attached studies cite the magnitude and variety of polyfluoroalkyl acids (PFAs) being released from landfills. These are being inhaled and absorbed by residents around the Coffin Butte Landfill (CBL) and bring significant health risks as detailed in the attached study "Volatile and semi-volatile organic compounds in landfill gas" by Pan and Liu.

Some of these forever chemicals escape into the air by fugitive emissions which Republic admits are occurring and some from collected landfill gas (LFG) passing through flares. LFG flares are no hotter than 850 °C which is insufficient to mineralize PFAs as documented in "A critical review of perfluoroalkyl and polyfluoroalkyl substances (PFAS) landfill disposal in the United States" by Tolaymat, et al. The full 18 page Tolaymat study is attached and describes why the only control Benton County has over PFA and other pollutants is to switch its municipal solid waste (MSW) disposal to a site with less rainfall, such as the Columbia Ridge Landfill near Arlington, Oregon.

Let me also call your attention to p11 of the Tolaymat study which describes how FTOH (fluorotelomer alcohols) are the majority of non-methane organics in LFG. Unfortunately for the health of Benton County residents these alcohols photooxidize into shorter chain PFCAs (Perfluoroalkyl carboxylic acids) which are very mobile and readily bioaccumulate in animals and humans. Typical human health impacts are high cholesterol, decreased antibody response, kidney cancer and reduced fetal growth (see the YouTube video "How One Company Secretly Poisoned The Planet" at <https://www.youtube.com/watch?v=SC2eSujzrUY>).

These health risks are completely contrary to Benton County's Comprehensive Plan which includes the goal of "maintaining and improving the quality of the air, water and land resources of the state." Similarly, PFAs and other emissions from CBL undercut Benton County's stated Core Value of "health considerations in all actions."

The US average blood concentration for all PFAs is just under 6 ppb (parts per billion), much higher than the 2 ppb generally regarded as safe for human health. Please deny Republic's request to continue operating the point source for PFA air and water pollution that is Coffin Butte Landfill.



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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

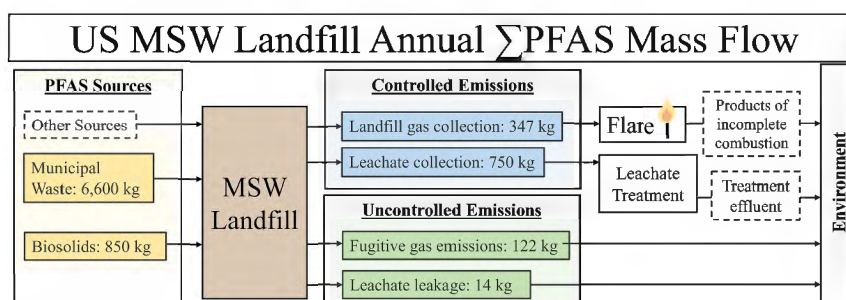
^d The Center for Environmental Measurements and Modeling, Office of Research and Development, U.S. Environmental Protection Agency, Research Triangle Park, NC, USA

^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



ARTICLE INFO

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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

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 Contaminant 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)	DF	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 Σ PFAS concentration in MSW of $50 \mu\text{g kg}^{-1}$ 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

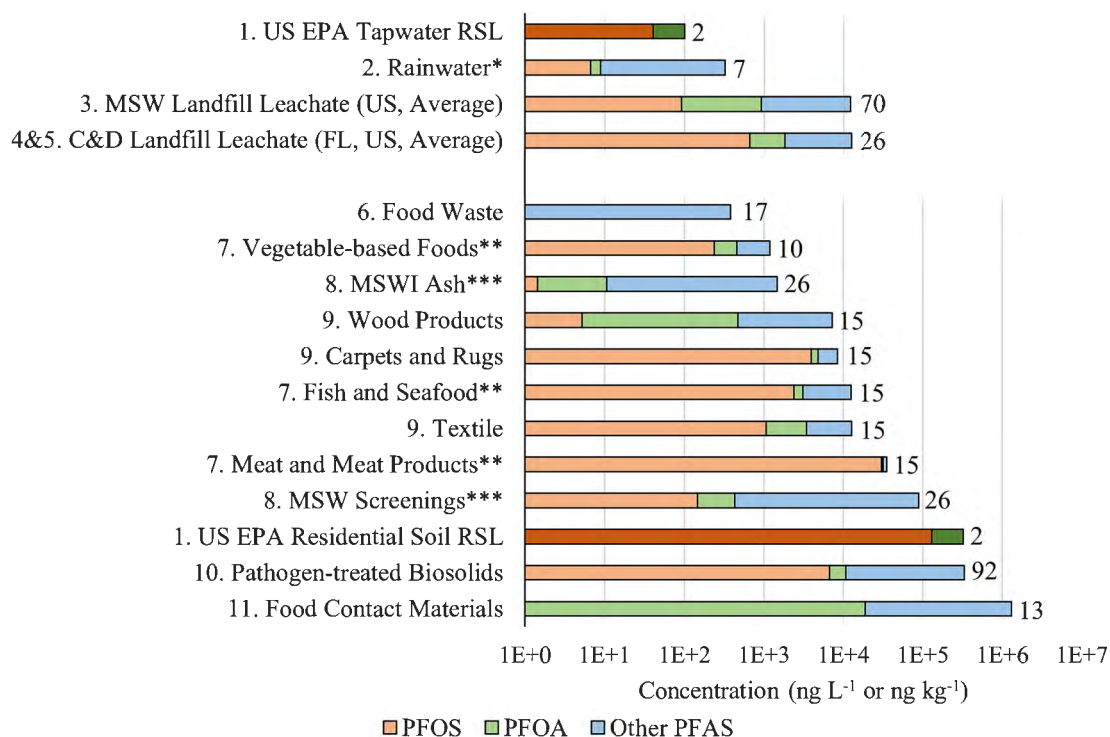


Fig. 1. PFAS concentrations and compositions measured in various products, wastes, and the environment compared to MSW landfill leachate.

* 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 Σ_{13} PFAS in food packaging as high as $8500 \mu\text{g kg}^{-1}$; 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 Σ_8 PFAS was $28 \mu\text{g kg}^{-1}$ before oxidation and $380 \mu\text{g kg}^{-1}$ 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 Σ 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 Σ_{17} PFAS = $0.38 \mu\text{g kg}^{-1}$); 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} PFAS of 5 and $18 \mu\text{g kg}^{-1}$, 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} PFAS in various household and consumer products found textiles, floor covering, and car interior materials represented the three highest maximum concentrations (78 , 38 , and $36 \mu\text{g kg}^{-1}$, respectively); the highest non-biodegradable median PFAS concentration was from insulation ($3.6 \mu\text{g kg}^{-1}$) (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 – $0.43 \mu\text{g kg}^{-1}$ PFOS among all electronic products is provided by Garg et al. (2020). Σ_{15} PFAS reported by Bečanová et al. (2016) ranged as high as $11.7 \mu\text{g kg}^{-1}$ (median: $0.4 \mu\text{g kg}^{-1}$) in electronic and electrical equipment (EEE) and as high as $2.2 \mu\text{g kg}^{-1}$ (median: $1.4 \mu\text{g kg}^{-1}$) 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 Σ_9 PFAS of $45 \mu\text{g kg}^{-1}$ of biosolids; Thompson et al. (2023a, 2023b) reported mean Σ_{92} PFAS of $500 \mu\text{g kg}^{-1}$ (dry) in untreated biosludge and $330 \mu\text{g kg}^{-1}$ (dry) 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 Σ_{21} PFAS in fly and bottom ash from three facilities in China, with concentrations ranging from 1.5 to $88 \mu\text{g kg}^{-1}$ in fly ash and from 3.1 to $77 \mu\text{g kg}^{-1}$ in bottom ash. Based on the concentrations of PFAS in a laboratory leaching study, the average minimum Σ_{26} PFAS in MSWI ash from a US facility was $1.5 \mu\text{g kg}^{-1}$ (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 Σ_{11} PFAS (Kim et al., 2016); sludge effluent collected from an electronics industry location in South Korea contained 91 $\mu\text{g kg}^{-1}$ of Σ_{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 $\mu\text{g kg}^{-1}$ (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 $\mu\text{g kg}^{-1}$, respectively, with concentrations as high as 50,000 $\mu\text{g kg}^{-1}$ for PFOA and 460,000 $\mu\text{g kg}^{-1}$ 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 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 digester 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 $\sum$ 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

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).

Table 2

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

Matrix	Number of samples	Number of PFAS detected (in Method)	Average $\sum$ PFAS (ng L ⁻¹)	$\sum$ 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 $\sum$ 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 $\sum$ 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 (Beeson 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

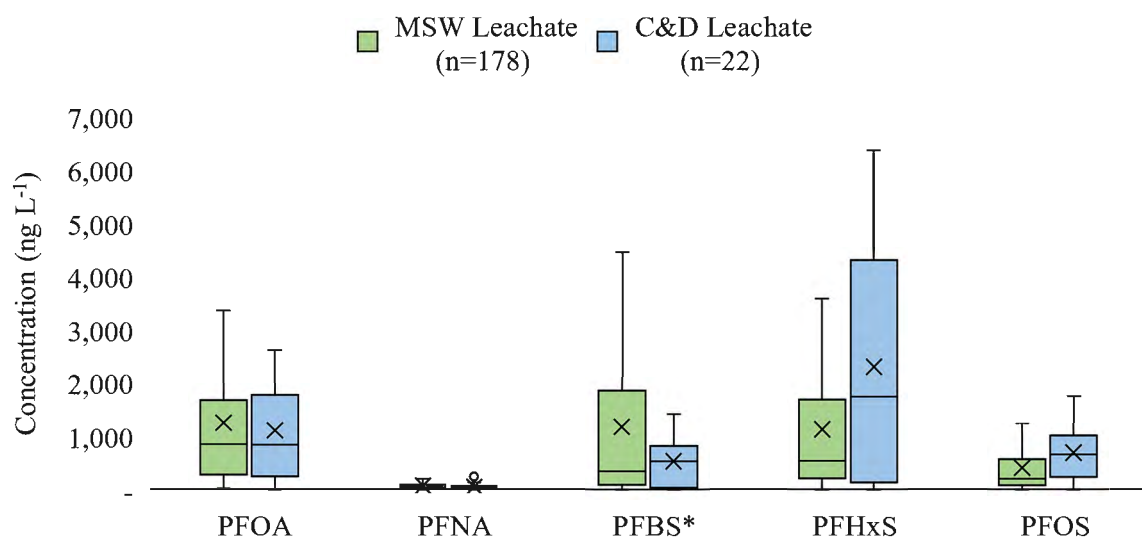


Fig. 2. Average concentrations of five PFAS with US EPA tapwater RSLs. Data from Gallen et al. (2017), Solo-Gabriele et al. (2020), and Chen et al. (2023).

* 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; $\sum_{17}\text{PFAS}$ concentrations averaged $45,000 \text{ ng L}^{-1}$.

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, $\sum_{24}\text{PFAS}$ and $\sum_{28}\text{PFAS}$ concentration was as high as $377,000 \text{ ng L}^{-1}$ (average $68,000 \text{ ng L}^{-1}$), 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 $\sum_{24}\text{PFAS}$ 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 $\sum_{51}\text{PFAS}$ 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 $\sum_{92}\text{PFAS}$, 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 $\sum_{19}\text{PFAS}$ 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 $\text{H}_2\text{S}_{(\text{g})}$ 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 FTA), 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 • Highly effective method • Can be used for regeneration of spent materials	• Pretreatment of leachate might be required • Secondary treatment of concentrated PFAS required • As a standalone method, not practical for large volumes of leachate • Uses additional chemicals for treatment	GW, AFFF, LL	Smith et al. (2022); Robey et al. (2020); Malovanyy et al. (2023)
	Incineration				
	Chemical Oxidation	• Controllable by varying pH and temperatures	• 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 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 • Can be used to regenerate GACs • Catalytic microwave treatment could result in ~65–67% effectiveness	• High capital costs		Moriwaki et al. (2005); Vecitis et al. (2008); Babu et al. (2016)
	Microwaves		• 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 • Full fluorine balance needed		Hori et al. (2006)
	Supercritical water oxidation	• High maturity and close to commercialization • Low residence times required	• High-pressure and temperature processes can be energy intensive	GW, AFFF, 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 • Laboratory demonstrations only and thus low technology readiness level	LL	Travar et al. (2020)
	Biological processes	• Limited aerobic and anaerobic degradation of PFOS by bacteria and fungi reported	• Slow kinetics • Longer-chain PFAS converted to shorter-chain; no mineralization • Unlikely to be effective • Does not result in a concentrated PFAS stream that can be adequately managed • Environmental release of PFAS		Berhanu et al. (2023); Huang and Jaffé (2019)
	Constructed wetlands	• No demonstrated benefits for PFAS treatment		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 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 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 (L₀) of

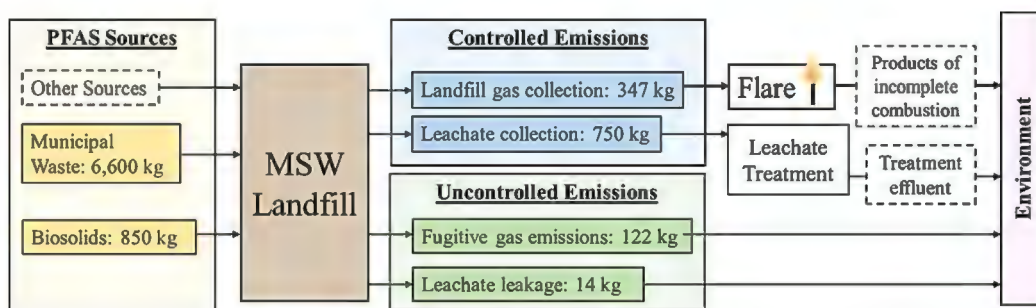


Fig. 3. Flowchart depiction of annual Σ PFAS loading and release at MSW and C&D debris landfills based on current understanding in the literature. Dashed lines represent PFAS streams which have not been quantified to any extent in the literature.

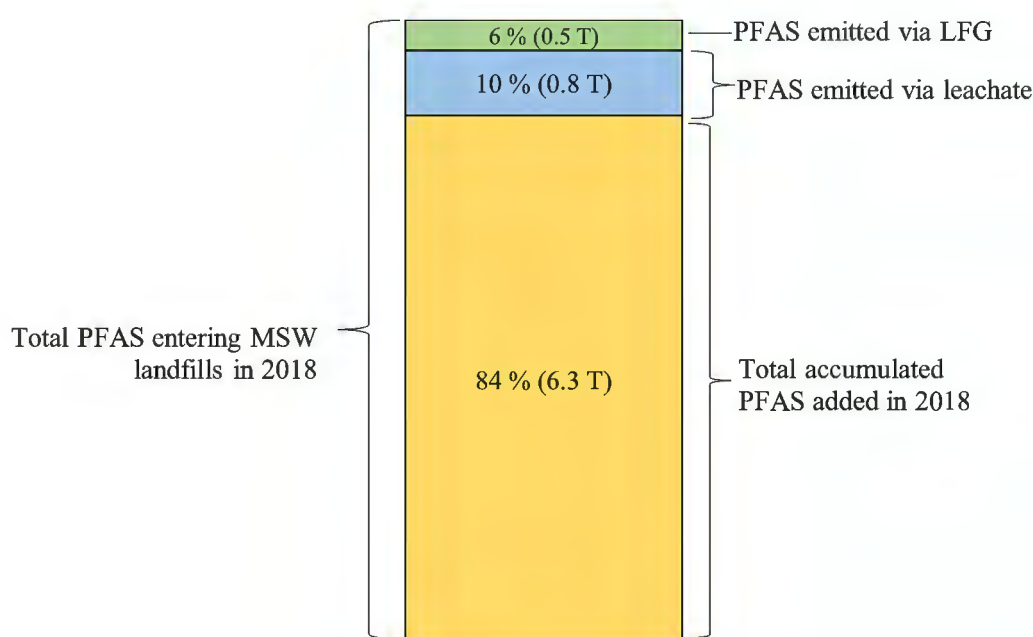


Fig. 4. Estimated PFAS mass balance for US MSW landfills.

MSW. Jain et al. (2021) estimated MSW methane emissions of 68 m³ of methane per metric ton (Mg) of waste, or approximately 136 m³ 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 Σ 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 Σ 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-reviewed 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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Per- and polyfluoroalkyl substances (PFAS) distribution in landfill gas collection systems: leachate and gas condensate partitioning

Thomas J. Smallwood ^a, Nicole M. Robey ^a, Yalan Liu ^a, John A. Bowden ^{a b}, Thabet M. Tolaymat ^c, Helena M. Solo-Gabriele ^d, Timothy G. Townsend ^a  

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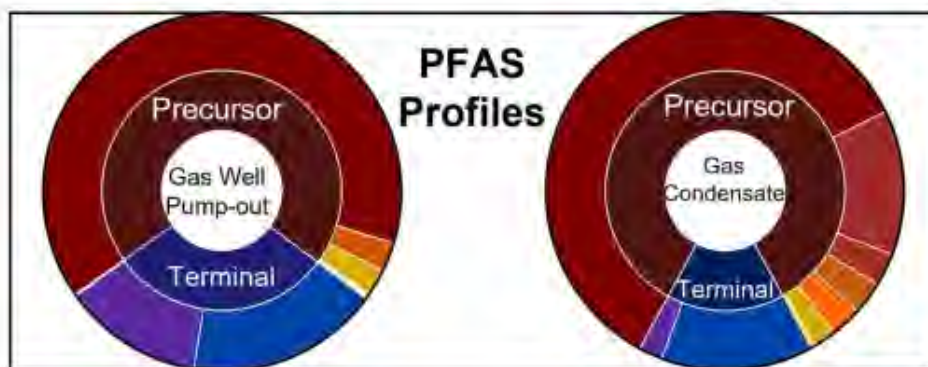
Highlights

- Two leachates were collected from landfill gas systems: condensate and well pumpout.
- Gas condensate is not colored, contains ammonia, alkalinity, and some metalloids.
- Gas well pump-out is dark in color, resembles leachate, high chloride concentrations.
- Condensate has lower total PFAS abundance, but greater diversity of species.
- PFAS which are more abundant in condensate include semi-volatile FTOHs and FASEs.

Abstract

While per- and polyfluoroalkyl substances (PFAS) have been reported extensively in municipal solid waste (MSW) landfill leachate, they have rarely been quantified in landfill gas or in discrete landfill liquids such as landfill gas condensate (LGC), and the potential for PFAS to partition to the condensate has not been reported. LGC and leachate collected from within gas wells known as gas well pump-out (GWP) from three MSW landfills underwent physical-chemical characterization and PFAS analysis to improve understanding of the conditions under which these liquids form and to illuminate PFAS behavior within landfills. LGC was observed to be clear liquid containing ammonia and alkalinity while GWP strongly resembled leachate – dark in color, high in chloride and ammonia. Notably, arsenic and antimony were found in concentrations exceeding regulatory thresholds by over two orders of magnitude in many LGC samples. LGC contained a lower average concentration of Σ PFAS (19,000ngL) compared to GWP (56,000ngL); however, LGC contained more diversity of PFAS, with 53 quantified compared to 44 in GWP. LGC contained proportionally more precursor PFAS than GWP, including more semi-volatile PFAS which are rarely measured in water matrices, such as fluorotelomer alcohols and perfluoroalkane sulfonamido ethanols. This study provides the first detailed comparison of these matrices to inform timely leachate management decisions.

Graphical Abstract



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Introduction

Waste decomposition within a landfill produces gas (landfill gas; LFG) consisting primarily of methane and carbon dioxide, which are often collected and either flared to oxidize methane or processed into a renewable fuel source. During the vacuum collection of LFG, gas from the warm, moist interior of the landfill comes into contact with ambient air temperatures, and that water vapor condenses to form a liquid which can accumulate in the landfill gas collection and control

system (GCCS). GCCS liquids are often broadly described as gas condensate [6], but true leachate, derived from within the waste mass itself, which collects in the gas wells (referred to here as gas well pump-out, or GWP), constitutes another major source of liquids in the GCCS. Liquids from a landfill GCCS are normally mixed with leachate intercepted at the bottom of the lined landfill but could be isolated if desired. Commonly referred to as landfill gas condensate (LGC), GCCS liquids are removed from pipes and wells installed and operated to extract and convey the gases produced as a result of waste decomposition. A diagram of a GCCS is included in Fig. 1 to illustrate the discrete sources of these liquids. LGC typically represents a small fraction of a landfill's total leachate generation and is commonly mixed with other sources of landfill leachate for disposal. The US Environmental Protection Agency (US EPA) estimated landfill LGC generation for an individual landfill site in the range of 2700 to 7500L per day [6]. For comparison, the landfills used in this study generate approximately 100,000L of leachate per day. An example calculation to estimate LGC generation based on LFG production is included in Section S1 of the supplemental information (SI).

Per- and polyfluoroalkyl substances (PFAS) are an emerging constituent of concern which present an operational challenge with landfill leachate due to their toxicity, persistence, and lack of viable treatment technologies [14], [16], [20], [28], [31], [39], [5], [56], [59], [60]. Studies have reported diverse and significant concentrations of PFAS in landfill leachate worldwide [28], [5], [8]. The most well-studied PFAS, perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), have been detected in leachate at concentrations ranging from ng L^{-1} to $\mu\text{g L}^{-1}$ [16]. In the US, PFOA and PFOS have been measured at concentrations as high as $50 \mu\text{g L}^{-1}$ [47] in landfill leachate, and PFAS regulations are being promulgated across the US and globally [19], [42], [49], [54], [53]. Although landfill leachates are not usually subject to regulation, the recent proposal to designate PFOA and PFOS as CERCLA Hazardous Substances [55] may determine how leachate is managed in the future, as any wastewater treatment effluent requirements will have upstream impacts on leachate management decisions.

While leachate has been described extensively in scientific literature, recent studies have placed focus on studying the behavior of PFAS (e.g., partitioning, transformation, and potential volatilization) within municipal solid waste (MSW) landfills [29]. Gas and air measurement of PFAS is comparatively limited in the literature; however, PFAS have been measured in ambient air above landfills and wastewater treatment plants [22], suggesting that they are either volatilizing or aerosolizing and entering the air [23]. To this point, PFAS have been shown to travel great distances through the air and have been measured in soil, water, and biota in remote locations such as the arctic [62]. The term “semi-volatile” is defined in different ways across the literature in reference to PFAS as well as other chemicals and might refer to compounds which possess a specified range of vapor pressures, or by their observed partitioning behaviors. Several PFAS, including fluorotelomer alcohols, perfluoroalkane sulfonamides and sulfonamido substances, and terminal perfluoroalkyl acids, have been quantified using both gas and liquid chromatography, indicating their presence in both phases [36], [37]. Because of their different origins, LGC and GWP are expected to differ significantly from each other in chemical composition. Differences between LGC and GWP are

anecdotally recognized within the solid waste industry, although some misconceptions persist. To explore these differences, with a focus on PFAS, LGC and GWP were collected from three MSW landfills in central Florida and underwent PFAS analysis and other characterizations. This is the first study of its kind to evaluate these landfill liquids, and the results of this investigation provide critical insight for the management of discrete sources of leachate within the landfill, and also shed light on the partitioning and transformation behavior of PFAS within MSW landfills.

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Section snippets

Sample collection

Twelve LGC and five GWP samples were collected from three active MSW landfills (referred to as Landfill A, B, and C) in Florida between July and October 2021. Table S1 provides detailed information regarding each site. GWP samples were collected using in-place submerged pumps already installed in the gas wells. The flexible hose connecting these pumps to the leachate collection system was removed from the pipes and used for collection. LGC samples were collected at the condensate knockouts at ...

Physical-chemical parameters

Although little has been published comparing LGC and GWP matrices, an understanding of the sources of each effluent type informs the expectation of significant differences between the two. For example, GWP, like landfill leachate, is expected to be dark in color and higher in organic matter and water-soluble constituents, such as chloride. Ammonia, on the other hand, might be present under certain conditions as either a gas or water-soluble ion and is likely to be found in both sample types. ...

Conclusion

The results of this study shed light on the conditions under which gas condensate and leachate are produced, and these findings provide valuable information about the fate and behavior of PFAS within MSW landfills. As PFAS regulations are promulgated at the state and federal level, as well as internationally, the results of this study contribute to a better understanding of how PFAS in waste contribute to PFAS in landfill effluent, which includes leachate and LFG. While the two leachate ...

CRediT authorship contribution statement

Thomas J. Smallwood: Conceptualization, Methodology, Validation, Data curation, Formal analysis, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Nicole M. Robey:** Conceptualization, Methodology, Validation, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Yalan Liu:** Conceptualization, Methodology, Validation, Data curation, Formal analysis, Investigation, Writing - original draft, Writing - review & editing. **John A. Bowden:** ...

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Timothy Townsend reports financial support was provided by United States Environmental Protection Agency. ...

Acknowledgments

The researchers have received support from the New River Solid Waste Association and the U.S. Environmental Protection Agency under the Science To Achieve Results (STAR) grant program (EPA-G2018-STAR-B1; Grant#: 83962001-0) to investigate the occurrence, source, and fate of PFAS in landfills. Data presented herein did not undergo a formal quality assurance review as outlined in "U.S. EPA Office of Research and Development's Quality Management Plan for Scientific Research." This manuscript was ...

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H. Hamid *et al.*

[Review of the fate and transformation of per- and polyfluoroalkyl substances \(PFASs\) in landfills](#)

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Occurrence, behaviors, and fate of per- and polyfluoroalkyl substances (PFASs) in typical municipal solid waste disposal sites

2024, Water Research

Citation Excerpt :

...These volatile PFASs tend to appear in the gas phase, which is discussed in detail in Section 2.2. Highly concentrated PFASs exist in landfill gas (Smallwood et al., 2023; Titaley et al., 2023). If the landfill gas is not efficiently collected, these PFASs may enter the ambient air via the discharge of landfill gas....

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

2023, Science of the Total Environment

Citation Excerpt :

...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. 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....

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Some landfill 'burps' contain airborne PFAS, study finds

PressPacs (/pressroom/presspacs.html) June 26, 2024

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"Landfill Gas: A Major Pathway for Neutral Per- and Polyfluoroalkyl Substance (PFAS) Release" (<https://pubs.acs.org/doi/10.1021/acs.estlett.4c00364>)
Environmental Science & Technology Letters

Many municipal landfills "burp" gas from decomposing organic matter rather than letting it build up. And burps from buried waste containing per- and polyfluoroalkyl substances (PFAS) can release these "forever chemicals" into the air, say researchers in ACS' *Environmental Science & Technology Letters*. Their study reports unexpectedly high levels of airborne PFAS at three landfills and demonstrates that vented gases and liquid by-products called leachates could transport similar amounts of these contaminants to the environment.

Many municipal landfills "burp" gas from decomposing organic matter rather than letting it build up. And burps from buried waste containing per- and polyfluoroalkyl substances (PFAS) can release these "forever chemicals" into the air, say researchers in ACS' *Environmental Science & Technology Letters*. Their study reports unexpectedly high levels of airborne PFAS at three landfills and demonstrates that vented gases and liquid by-products called leachates could transport similar amounts of these contaminants to the environment.

Some consumer products and commercial waste, such as children's clothing (</pressroom/newsreleases/2022/may/childrens-products-labeled-water-or-stain-resistant-may-contain-pfas.html>), cosmetics (</pressroom/newsreleases/2022/october/beauty-products-with-fluorinated-ingredients-may-also-contain-pfas.html>) and wastewater treatment sludge solids (</pressroom/presspacs/2023/march/toilet-paper-is-an-unexpected-source-of-pfas-in-wastewater.html>), contain PFAS — and they ultimately end up in landfills. Timothy Townsend and colleagues previously established that PFAS-containing waste can contaminate the water that seeps through landfills. This leachate is usually captured and treated before entering the environment. Landfills also produce gas that can be captured and controlled, but unlike leachate, it's often released untreated. The burped gas is mostly made up of methane and carbon dioxide; however, two recent studies also discovered a subset of airborne PFAS called fluorotelomer alcohols, which have the potential to be toxic when inhaled and can be transported long distances. Since the prevalence of PFAS-contaminated landfill vapors isn't yet widely known, Townsend, Ashley Lin and their team wanted to identify and measure them in vented gas at three sites in Florida.

The researchers pumped landfill gas from pipes through cartridges filled with resin that captured the airborne PFAS. They freed the compounds from the cartridges with organic solvents and analyzed the extracts for 27 neutrally charged PFAS, including fluorotelomer alcohols. Surprisingly, some of the fluorotelomer alcohol levels were up to two orders of magnitude higher than previous studies at other landfills. Three of these alcohols (abbreviated 6:2, 8:2 and 10:2) comprised most of the vaporized contaminants measured at each site. The researchers also collected leachate samples at the Florida sites and analyzed them for ionic PFAS commonly found in water samples. From this data, they estimated that the annual amount of fluorine (as a proxy for PFAS content) leaving the landfills through gas emissions could be similar to, or even greater than, the amount leaving through leachates.



Landfill burps from three sites in Florida, such as this one, contain high levels of airborne PFAS, including fluorotelomer alcohols.
Ashley Lin

Because landfills are repositories for PFAS, this work indicates that vented gas from these sites should be considered in future mitigation and management strategies to reduce potential inhalation exposure and release to the environment. Some landfills burn the vapors or trap them for energy production, and the team suggests that further research is needed to determine the degree of removal these treatments provide for airborne contaminants.

The authors acknowledge funding from the Florida Department of Environmental Protection and from the U.S. Environmental Protection Agency under the Science to Achieve Results grant program.

###

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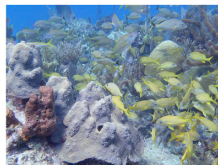


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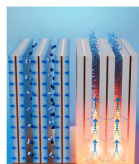
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Review article

Volatile and semi-volatile organic compounds in landfill gas: Composition characteristics and health risks

Qi Pan ^{a b c}, Qing-Yu Liu ^{a b}, Jing Zheng ^{a b c}, Yan-Hong Li ^c, Song Xiang ^{a b}, Xiao-Jie Sun ^c, Xiao-Song He ^{a b}[Show more](#) ▼[Outline](#) | [Share](#) [Cite](#)<https://doi.org/10.1016/j.envint.2023.107886> ↗[Get rights and content](#) ↗Under a Creative Commons [license](#) ↗

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Highlights

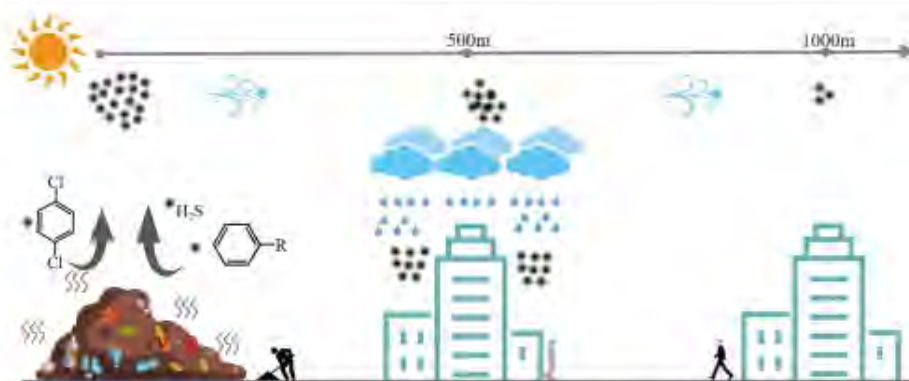
- VOC and SVOC data were collected from 72 landfills in 20 countries.
- Aromatic and halogenated compounds are the major sources of carcinogenic risks.
- The risks posed by ethyl acetate in landfill gas also deserve attention.
- Landfill gas health risk was influenced by meteorological conditions.

Abstract

Gas emitted from landfills contains a large quantity of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs), some of which are carcinogenic, teratogenic, and

mutagenic, thereby posing a serious threat to the health of landfill workers and nearby residents. However, the global hazards of VOCs and SVOCs in landfill gas to human health remain unclear. To quantify the global risk distributions of these pollutants, we collected the composition and concentration data of VOCs and SVOCs from 72 landfills in 20 countries from the core database of Web of Science and assessed their human health risks as well as analyzed their influencing factors. Organic compounds in landfill gas were found to primarily result from the biodegradation of natural organic waste or the emissions and volatilization of chemical products, with the concentration range of $1 \times 10^{-1} - 1 \times 10^6 \mu\text{g}/\text{m}^3$. The respiratory system, in particular, lung was the major target organ of VOCs and SVOCs, with additional adverse health impacts ranging from headache and allergies to lung cancer. Aromatic and halogenated compounds were the primary sources of health risk, while ethyl acetate and acetone from the biodegradation of natural organic waste also exceeded the acceptable levels for human health. Overall, VOCs and SVOCs affected residents within 1,000m of landfills. Air temperature, relative humidity, air pressure, wind direction, and wind speed were the major factors that influenced the health risks of VOCs and SVOCs. Currently, landfill risk assessments of VOCs and SVOCs are primarily based on respiratory inhalation, with health risks due to other exposure routes remaining poorly elucidated. In addition, potential health risks due to the transport and transformation of landfill gas emitted into the atmosphere should be further studied.

Graphical abstract



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Keywords

Waste; Landfill gas; Volatile organic compounds; Semi-volatile organic compounds; Risk assessment

1. Introduction

The generation of municipal solid waste (MSW) has drastically increased with the rapid growth rates of economy and population. As in other countries, there was a significant increase in the production of China's MSW with 235 million tons in 2020, a 90.4% increase compared to 2010 ([Ministry of housing., 2021](#)). Globally, annual generation of MSW is expected to reach 3.4 billion tons by 2050 ([Haza et al., 2018](#)).

Landfill is one of the most important means of MSW disposal worldwide. Large amounts of gases are produced in landfills through complex physical, chemical, and biochemical processes and being emitted into the atmosphere through cover soils or landfill wells, thereby threatening the safety and health of landfill workers and nearby residents. Waste in landfills includes natural organic materials, such as vegetables, meat, and trees, and synthetic substances, such as chlorinated disinfectants and petrochemicals, which releases a large variety and quantity of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) ([Slack et al., 2005](#)).

Previous studies on landfills have focused on the following three aspects: (1) landfill gas, in particular, methane and greenhouse gas emission ([Lucernoni et al., 2017](#)); (2) landfill leachate composition, degradation and treatment characteristics, and groundwater pollution characteristics ([Abiriga et al., 2020](#), [Mehmet et al., 2020](#), [Oturán et al., 2015](#), [Yang et al., 2021](#)); and (3) degradation of organics and humus formation ([Liu et al., 2021](#), [Yang and Antonietti, 2020](#)). Previous studies on landfill gas have primarily focused on methane, and the human health risks and transformation characteristics of VOCs and SVOCs remaining overlooked. Many VOCs and SVOCs in landfill gas are mutagenic, teratogenic, and carcinogenic and can cause human health risks, such as irritating respiratory tract, damaged central nervous system, and even cancer ([Allen et al., 1997](#), [Butt et al., 2016](#)). Human health risks posed by VOCs and SVOCs in landfill gas have been lately receiving attention but have remained spatially limited to individual landfills ([Liu et al., 2016a](#), [Piccardo et al., 2022](#), [Wang et al., 2021](#)). Only few studies have been conducted about the compositions and concentrations of VOCs and SVOCs ([Wu et al., 2018](#); [Yao et al., 2019](#)). Therefore, this study aimed to (1) globally characterize the sources, compositions, and concentrations of VOCs and SVOCs produced by landfills; (2) assess their potential human health risks to workers and nearby residents and address the biological mechanisms underlying those risks; and (3) identify their primary drivers. ([Yang and Antonietti, 2020](#))

2. Materials and methods

2.1. Data collection

Scientific studies published during 2002–2022 were searched with the subject words of “VOC, SVOC, VOCs, SVOCs, volatile organic compound, semi-volatile organic compound, volatile organic compounds, or semi-volatile organic compounds” and “landfill gas” in the core database of Web of Science. As shown in [Table S1](#), 53 publications, including 72 landfills in 20 countries, were obtained on the VOC and SVOC data worldwide. For the lowest concentrations, the terms used in the original studies included “ND” (not detected) and “<LOQ” (below limit of quantification), and n.a. (not available) was used to indicate studies reporting only maximum concentrations.

2.2. Sampling and measurement techniques

The sampling techniques applied in 53 studies included sampling bags, vacuum canisters, glass-bombs, sorbent tubes, and flux boxes, with the sampling bags being the most common one. The measurement methods applied in 53 studies included gas-vent sampling, velocity measurement, surface flux chambers, production model of landfill gas, concentration ratios estimated relative to methane, and inverse dispersion modeling, with the surface flux chambers being the most common one. Most of these studies in our data chose helium as gas matrix during the measure process ([Chiriac et al., 2007](#), [Ding et al., 2012](#), [Fang et al., 2012](#), [He et al., 2015](#), [Kim et al., 2012](#), [Lim et al., 2018](#)), and the rest used nitrogen as gas matrix ([Duan et al., 2020](#), [Liu and Zheng, 2020](#)).

2.3. Risk assessment

Based on carcinogenicity, the International Agency for Research on Cancer (IARC) classifies VOCs and SVOCs into various groups, including Group 1 (carcinogenic to humans), Group 2A (probably carcinogenic to humans), and Group 2B (possibly carcinogenic to humans) ([Yao et al., 2019](#)). Based on the concentration data of VOCs and SVOCs extracted from 53 studies ([Table S1](#)), human health risk assessment was conducted to estimate their health risks to landfill workers and nearby residents. However, we excluded the VOC and SVOC data from organized emissions, such as soil pores and manifolds before burning in a flare. Exposure to pollutants in the atmosphere occurs via the following three pathways: dermal absorption, ingestion, and respiratory inhalation. As the exposed population at landfills are primarily workers and the pollutant carcinogenicity parameters are incomplete, human health risk assessment conducted in this study was limited to respiratory inhalation. Non-carcinogenic and carcinogenic effects of exposure to odorous volatile compounds by inhalation were evaluated via the methodology recommended by the United States Environmental Protection Agency ([Usepa, 2009](#)). The exposure concentration (EC, $\mu\text{g}\cdot\text{m}^{-3}$) can be estimated as follows:

$$EC = \frac{Ca \times ET \times EF \times ED}{AT \times 365 \times 24} \quad (1)$$

where Ca is airborne pollutant concentration ($\mu\text{g}\cdot\text{m}^{-3}$), ET is daily exposure time ($8\text{h}\cdot\text{d}^{-1}$), EF is exposure frequency ($260\text{d}\cdot\text{yr}^{-1}$), ED is exposure duration (in 25yr), and AT is the average time of effect expressed as 75yr for carcinogenic effect and 25yr for non-carcinogenic effect.

The carcinogenic risk (CR) of individual pollutants (ΣCR) can be estimated using Eq. (2)

$$CR = EC \times IUR \quad (2)$$

where IUR is inhalation unit risk ($\mu\text{g}\cdot\text{m}^{-3}$)⁻¹. When $IUR=2\times 10^{-6}$ ($\mu\text{g}\cdot\text{m}^{-3}$)⁻¹, two additional cancer cases (upper bound) per one million people are expected if exposed to 1 μg of chemical per m^3 of air every day during the average time of effect (Piccardo et al., 2022). When $CR>1$, carcinogenic risk is considered to be possible.

The non-carcinogenic risk of individual pollutants (HI) can be estimated using Eq. (3)

$$HI = \frac{EC}{RfC \times 1000 \mu\text{g}/\text{m}^3} \quad (3)$$

where RfC is the reference concentration ($\text{mg}\cdot\text{m}^{-3}$). When $HI>1$, non-carcinogenic risk is considered to be possible. The toxicological parameters of VOCs and SVOCs are shown in Table S5.

2.4. Statistical analysis

Statistical analysis was conducted using GraphPad Prism 8.0. Figures and maps were produced using OriginPro 2018 and ArcGIS 10.8, respectively.

3. Results and discussion

3.1. Sources, compositions, and concentrations of VOCs and SVOCs in landfill gas

3.1.1. Sources of organic pollutants in landfill gas

Organic compounds in landfill gas were found to primarily result from the biodegradation of natural organic waste or the emissions and volatilization of chemical products (Duan et al., 2021a, Duan et al., 2021b). As presented in Table 1, hydrocarbon, nitrogen, oxygen, and sulfur compounds and terpenes originate through the biodegradation of organic waste, such as kitchen waste, vegetables, and fruits, and their compositions and concentrations depend on the biodegradation stage of waste. The aromatic and halogenated compounds originate from the direct volatilization of chemical products or the biological degradation of natural organic wastes (Duan et al., 2014). The aromatic compounds primarily originate from petrochemicals, such as coal tar, asphalt, and crude oil, among which benzene was the most common, which is derived from chemical products, such as gasoline, paints, and varnishes (Table S2). The halogenated compounds primarily originated from chemical products, such as disinfectants, pesticides, and degreasers (Dincer et al., 2006), among which the most common was tetrachloroethylene, which is derived from dry cleaners, dye solvents, and detergents (Table S3).

Table 1. Sources of organic compounds in landfills.

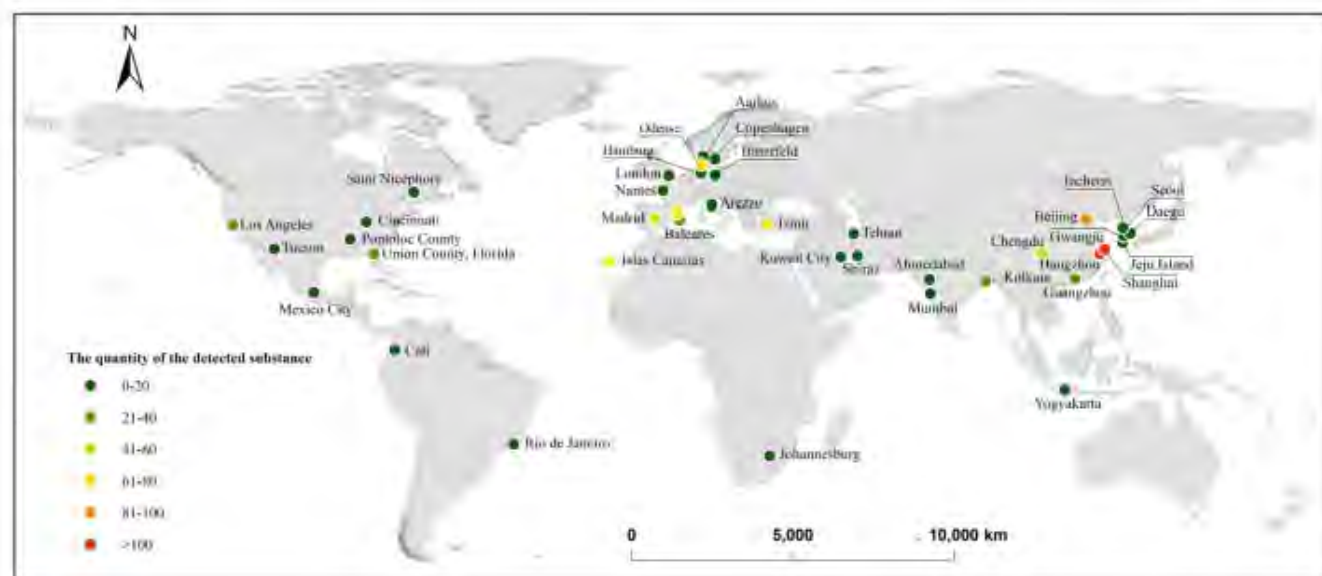
Compounds	Sources	References
Oxygenated compounds	Vegetables, fruits, kitchen waste, yard waste, medical waste, solvents, and plastic packaging	(Duan et al., 2021a, Zou et al., 2003, Butt et al., 2016)
Sulfocompounds	Kitchen waste, gypsum drywall, and wall board	(Duan et al., 2021b, James and Stack, 1997, Lucernoni et al., 2017, Yaghmaien et al., 2019, Butt et al., 2016)
Nitrogen compounds	Proteins, rubber, and kitchen waste	(Abiriga et al., 2020, Chiriac et al., 2007, James and Stack, 1997, Wu et al., 2017)
Hydrocarbons	Edible oils, paper, plastic packaging, fragrant detergents, solvents, refrigerants, waste cooking oil, and pesticides	(AlAhmad et al., 2012, Duan et al., 2021b, Lucernoni et al., 2017)
Terpenes	Yard waste, plant waste, household aromatic cleaners, air fresheners, and pharmaceuticals	(Duan et al., 2021b, James and Stack, 1997, Zou et al., 2003)
Aromatic compounds	Coal tar, asphalt, crude oil, plastic bags, paints, vehicle exhaust emissions, food containers, food with high fat content, paper, yard waste, solubilizers, and yard/green waste.	(Abiriga et al., 2020, AlAhmad et al., 2012, Baghanam et al., 2020, James and Stack, 1997, Liu et al., 2016b, Zou et al., 2003, Randazzo et al., 2022)
Halogenated compounds	Pesticides, insecticides, disinfectants, cleaners, plastics, dry cleaners, paint strippers, dye solvents, degreasers, soaps, paints, varnishes, and refrigerants	(AlAhmad et al., 2012, Dincer et al., 2006, Gonzalez et al., 2013, Piccardo et al., 2022, Butt et al., 2016)

3.1.2. Compositions of VOCs and SVOCs in different countries

VOCs and SVOCs in 72 landfills of 20 countries were reported. The top five reported countries were China, South Korea, the United Kingdom, Spain, and the United States (Figure S1), all belonging to the high-income class, except China that belongs to the mid-to-high income class. Briefly, the level of the national economic growth was positively correlated with the study of VOCs and SVOCs in landfills.

These 72 landfills analyzed globally were located in 40 cities. As shown in Fig. 1, the top three cities with the most detected pollutants were Shanghai ($n=132$), Hangzhou ($n=116$), and Beijing ($n=91$). However, no >40 pollutants were detected in landfill gas emissions from most of the remaining cities. Aromatic compounds were the most reported landfill gas in all countries (Figure S2). The aromatic compounds included toluene, ethylbenzene, and xylenes, as detected in landfills on several continents, including Europe, North America, and Asia. As the major components of benzene, toluene, ethylbenzene, and xylene (BTEX), these three compounds are widely present in

chemicals expended by humans in everyday life, such as gasoline, dyes, and paints. The halogenated compounds, in particular, tetrachloroethylene and trichloroethylene, were also commonly reported. Among the top 50 reported VOCs and SVOCs in landfill gas, a small number of sulfur compounds was present, with methyl mercaptan ($n=12$) and dimethyl sulfide ($n=12$) being detected frequently primarily through the degradation of sulfur-containing food waste, such as methionine.



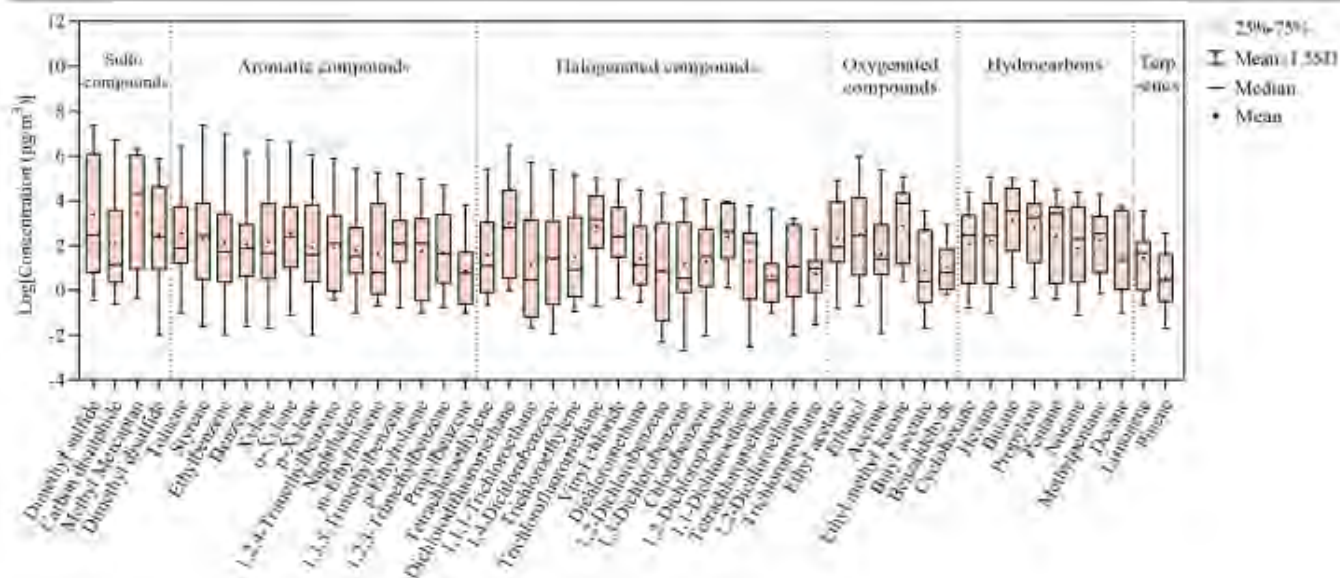
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Fig. 1. Quantity and spatial distribution of VOCs and SVOCs detected in landfill gas in cities.

3.1.3. Concentrations of VOCs and SVOCs

A significant spatial difference was observed in the concentrations of VOCs and SVOCs in landfill gas worldwide. The concentrations of the same pollutants in the different countries varied by 6–8 orders of magnitude over a range of $1 \times 10^{-1} - 1 \times 10^6 \mu\text{g}/\text{m}^3$. As shown in Fig. 2, the aromatic compounds were present at very high concentrations in narrow ranges in certain landfills. Compared with those of the aromatic compounds, the overall concentrations of the halogenated compounds were slightly low but differed widely between specific pollutants. Although a small number of sulfur compounds were reported in a few studies, their overall concentrations were high. For example, dimethyl sulfide, and methyl mercaptan generally occurred at higher concentrations than the other pollutants. In China, this was primarily because the high proportion of kitchen waste was characterized by high salt, oil, and water contents, which is more likely to produce high concentrations of sulfur and oxygen compounds (Liu et al., 2018). The overall concentrations of oxygen and hydrocarbon compounds were less variable than those of sulfur compounds and showed a decreasing trend with the increasing carbon atoms.



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Fig. 2. Concentrations of the top 50 VOCs and SVOCs detected in landfill gas in cities.

The concentrations of VOCs and SVOCs in landfill gas were affected by the sampling and measurement method. As show in [Table S4](#), the chemical composition of the sampling bag method may undergo a significant loss or change during storage, or vacuum canisters may lose analytes owing to wall effect, matrix effect, and condensation and VOCs or SVOCs loss in sorbent tubes due to reaction between sulphur compounds and carbon-based sorbent tubes. In addition, human activities will also affect the concentration of VOCs and SVOCs. The dumping and compacting work of waste in the landfill will promote the volatilization of VOCs and SVOCs in the waste, resulting in an increase in the concentration in the air ([Chiriac et al., 2007](#), [Duan et al., 2014](#)).

3.2. Potential human health risk assessment

Because only the respiratory inhalation is considered and the dermal absorption and ingestion is not considered, the results of human health risk assessment may be not completely accurate. As the concentration data of VOCs in the papers are not all sampled in the way of simulating human respiratory inhalation, the results of human health assessment in this paper can only be called potential human health risk assessment.

3.2.1. Impacts of VOCs and SVOCs on human health

(1) Cancer risks posed by the inhalation of VOCs and SVOCs

The inhalation of VOCs and SVOCs can cause cancer ([Liu et al., 2014](#), [Yang et al., 2017](#)). As presented in [Table 2](#), chloroform, tetrachloromethane, hexanal, and 1,4-dioxane can cause breast cancer. Benzene may cause leukemia, and 1,4-dichlorobenzene may cause head and neck cancer. The lung is the major target organ of polycyclic aromatic hydrocarbons (PAH) carcinogenicity, and the inhalation of

PAHs can cause lung cancers; PAHs can also cause skin, bladder, and urinary system cancers as a function of different routes, concentrations, and exposure environments. For example, the increased risk of skin cancer follows high dermal exposure to PAHs, whereas the increased risk of bladder and other organ cancers is primarily found in workers in industries with high exposure to PAHs (Boffetta et al., 1997).

Table 2. Carcinogenic health effects of VOCs and SVOCs.

Diseases	Compounds	References
Breast cancer	Chloroform, tetrachloromethane, hexanal, and 1,4-dioxane	(Calderon et al., 2022)
Leukemia	Benzene	(Zhou et al., 2015)
Head and neck cancer	1,4-dichlorobenzene	(Taware et al., 2018)
Lung cancer	PAHs	(Ben et al., 2004, Boffetta et al., 1997, Bosetti et al., 2005)
Skin cancer	PAHs	(Boffetta et al., 1997)
Bladder cancer	PAHs	(Boffetta et al., 1997, Bosetti et al., 2005)
Carcinoma of urinary system	PAHs	(Bosetti et al., 2005)

(2) Non-carcinogenic risks posed by the inhalation of VOCs and SVOCs

The impacts of VOCs and SVOCs on human health are primarily concentrated in the respiratory system as respiratory inhalation is the major route of human exposure to VOCs and SVOCs. As presented in Table 3, α -pinene, 2-methyl-1,2-butadiene, and aldehydes, such as hexanal and octanal, are associated with the deterioration of lung function (Cakmak et al., 2014), while chlorobenzene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, and 1,4-dichlorobenzene can cause asthma. Asthma can lead to irreversible emphysema and permanent lung damage. Long-term lung damage increases the risks of asthma attack and heart disease (Kim et al., 2013).

Table 3. Non-carcinogenic health effects of VOCs and SVOCs.

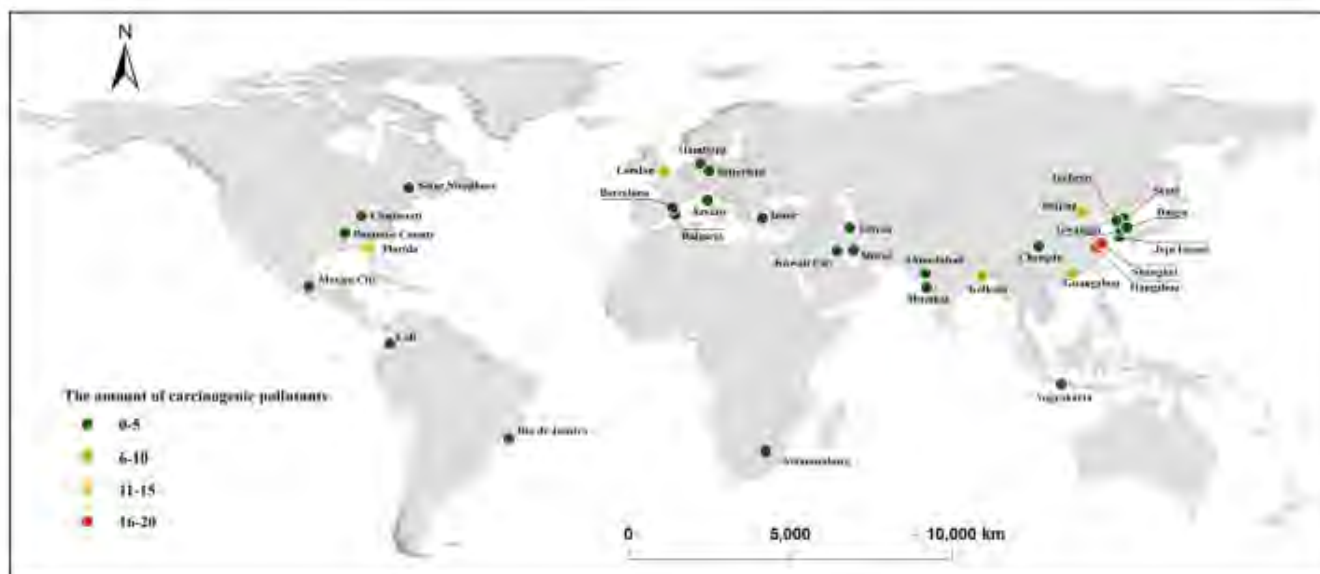
Diseases	Compounds	References
Lung function deterioration	Hexanal, octanal, nonanal, decanal, benzene, styrene, α -pinene, 2-methyl-1,2-butadiene, naphthalene, and 2-furancarboxaldehyde	(Cakmak et al., 2014, Martins et al., 2012)

Diseases	Compounds	References
Asthma	Benzene, toluene, ethylbenzene, p-xylene, m-xylene, styrene, chlorobenzene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, and 1,4-dichlorobenzene	(Martins et al., 2012, Rumchev et al., 2004)
Allergies	Formaldehyde, hexane, octane, nonane, decane, toluene, ethylbenzene, p±m-xylene, o-ethyltoluene, m-ethyltoluene, p-ethyltoluene, and chlorobenzene	(Kim et al., 2013, Lehmann et al., 2001, Martins et al., 2012)

In addition to adversely affecting the human respiratory system, VOCs and SVOCs can cause allergies, headache, eye irritation, nausea, and vomiting (Duan et al., 2014, Kim et al., 2013, Martins et al., 2012). As presented in Table 3, naphthalene and other aromatic compounds, such as benzene, toluene, and ethylbenzene, can cause allergic reactions in addition to asthma and lung function deterioration (Lehmann et al., 2001, Martins et al., 2012, Rumchev et al., 2004), while chlorobenzene and alkanes, such as hexane and octane, increase the likelihood of allergic reactions to milk and egg albumen (Lehmann et al., 2001). Atopic reactions increase the risk of allergies, which can cause shock and eventually death.

3.2.2. Global distribution of potential human health risks

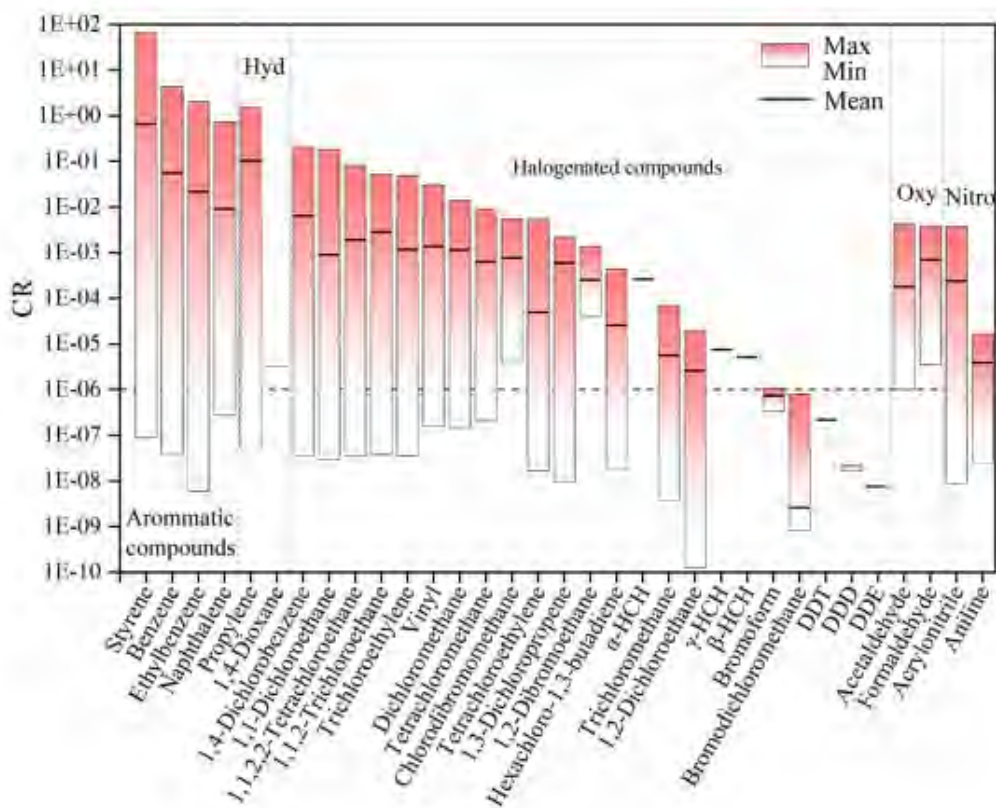
The carcinogenic risks of various landfill pollutants at the maximum (CR_{max}) and mean (CR_{mean}) concentrations were estimated for the 40 cities worldwide. Globally, 30 out of 34 pollutants may cause cancer in humans. As shown in Fig. 3, Hangzhou ($n=20$), Shanghai ($n=19$), and Union County of Florida ($n=12$) exhibited the largest number of carcinogenic VOCs and SVOCs, with the remaining cities exhibiting less than ten such compounds. The aromatic and halogenated compounds were the major sources of carcinogenic risks (Figs. 4 and S3). The aromatic compounds were commonly detected with high exceedance rates in landfills, presenting the highest carcinogenic risk. The halogenated compounds exceeded risk limits at a slightly lower exceedance rate than that of the aromatic compounds; however, their frequent detections suggested a high carcinogenic risk. Formaldehyde, acetaldehyde, acrylonitrile, and aniline were also identified as carcinogenic (Fig. 4). Propylene oxide was detected in only one landfill despite its high carcinogenic risk ($CR_{max}=1.5$). Therefore, more attention should be paid to the carcinogenic aromatic and halogenated compounds detected with high exceedance rates, such as ethylbenzene, benzene, carbon tetrachloride, and methylene chloride.



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Fig. 3. Number and spatial distribution of VOCs and SVOCs with carcinogenic risk in landfill gas in cities.



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Fig. 4. Carcinogenic risk assessment of VOCs and SVOCs detected in landfill gas in cities (Hyd: hydrocarbons; Oxy: oxygenated compounds; and Nitro: nitrogen compounds).

The non-carcinogenic risks of pollutants at the maximum ($HI_{\max}$) and mean (HI_{mean}) concentrations detected were also assessed for the 40 cities worldwide. As shown in Fig. 5, Shanghai (n=29), Hangzhou (n=27), and United County of Florida (n=20) released more non-carcinogenic pollutants than the other cities (n<20). Both $HI_{\max}$ and HI_{mean} were greater than unity for 38 out of 52 pollutants worldwide, indicating that most organic compounds in landfill gas caused non-carcinogenic risks (Fig. 6). Pollutants with excess non-carcinogenic risks primarily included aromatic, halogenated, and oxygen compounds. Being emitted from landfills, VOCs and SVOCs showed a lower exceedance rate for non-carcinogenic risks than for carcinogenic risks (Figure S4). The aromatic compounds, such as benzene, naphthalene, and toluene, and the halogenated compounds, such as trichloroethylene, tetrachloroethylene, and chlorobenzene, not only presented high human health risk but also were frequently reported in landfills, thereby emphasizing the urgent need to focus on them. Being a potential hazard to human health, ethyl acetate, typically reported in landfills, also deserves special attention in mitigating the health risks.



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Fig. 5. Number and spatial distribution of VOCs and SVOCs with non-carcinogenic risk in landfill gas in cities.

carcinogenic risk in North America and Europe but posed a human health risk in Asia. Organic matters such as aniline, acetone, methyl ethyl ketone, and methyl isobutyrate did not pose human health risks in North America but posed non-carcinogenic risks in Asia. Although oxygenated, sulfurous and nitrogen compounds were detected in landfills in Europe and North America but did not pose human health risks. However, most of these pollutants may pose human health risks in Asia owing to their high concentrations. The health risks from trichloroethylene were also lower in Europe than in Asia likely owing to the strict regulation of trichloroethylene issued by the European Chemicals Agency in 2010. Trichloroethylene has been banned for metal cleaners, which is a potential source of trichloroethylene detected at landfills ([Allen et al., 1997](#), [Chiriac et al., 2007](#)). Landfills in South America and Africa posed low human health risks, with only one or two hazardous pollutants in each country, primarily owing to the small number of local studies. No risk assessment was performed for Oceania and Antarctica owing to the lack of data.

3.2.4. Country-level potential human health risks

(1) North American countries

In North America, the United States was subjected to the highest health risks from landfill gas, followed by Canada and Mexico. The carcinogenic risks of landfill gas in the United States and Canada were primarily associated with the aromatic and halogenated compounds ([Figure S13](#)). In the United States, various landfill pollutants posed carcinogenic risks, with 1,4-dichlorobenzene, 1,1-dichloroethane, and styrene as the top three pollutants. However, ethylbenzene was the most hazardous landfill gas in Canada. According to the limited number of studies, no VOCs and SVOCs that caused human health risks were observed in Mexican landfills.

Similar to the results of the carcinogenic risk assessment, the United States exhibited more non-carcinogenic pollutants and higher non-carcinogenic risks at landfills than the other North American countries. The halogenated and aromatic compounds were the major sources of non-carcinogenic risk in the United States and Canada likely owing to the extensive use of such products ([Figure S14](#)). For example, 1,1,2-trichloroethane and benzene posed health risks in the United States. Such hazardous pollutants from landfills, including xylenes, ethylbenzene, toluene, naphthalene, trichloroethylene, and tetrachloroethylene, were fewer in Canada than in the United States. In Mexico, only toluene from landfills had potential health risks.

(2) European countries

The overall health risk of VOCs and SVOCs in landfill gas was lower in the European countries than in the United States, with slight differences between the countries ([Figure S15](#)). The European countries were in the following descending order of CR_{max} : the United Kingdom>Germany>Italy>Spain. The overall health risk was relatively high in the United Kingdom owing to the six halogenated compounds with carcinogenic toxicity: trichloroethylene, tetrachloroethane, vinyl chloride, 1,1,2,-trichloroethane, tetrachloroethylene, and benzene. Similar

to the United Kingdom, the halogenated compounds from landfills were the major source of carcinogenic risks in Germany. α -HCH and γ -HCH cause cancer risk but were rarely detected in landfill gas. Dichloromethane, vinyl chloride, trichloroethylene, and tetrachloroethylene required special attention as they not only have cancer risk but also have been frequently detected in landfill gas. In Italy and Spain, landfill gas exhibited low carcinogenic risk owing to the low CR_{max} value of pollutants and their small number detected.

The results of the non-carcinogenic and carcinogenic risk assessments were similar in the European countries. The UK and Germany were the two countries with the highest overall risk ([Figure S16](#)). The halogenated compounds, such as vinyl chloride, trichloroethylene, and 1,1,1-trichloroethane, were the major sources of non-carcinogenic risk. The non-carcinogenic risk of organic substances was lower in Italy than in the United Kingdom and Germany. No pollutants exhibited non-carcinogenic risk in Spain, as the pollutants (i.e., ethyl acetate, benzene, xylene, and vinyl chloride) detected in landfill gas remained at low concentrations. Overall, vinyl chloride and trichloroethylene showed both carcinogenic and non-carcinogenic risks that mostly exceeded the threshold in the European countries, which need to be reduced.

(3) Asian countries

The aromatic compounds from landfills were the major source of carcinogenic risks in Asia ([Figure S17](#)); however, the causes differed between the countries. As the oil-producing countries, the petrochemical industry is the mainstay of Kuwait and Iran, with a large quantity of the aromatic compounds being generated during production and processing. South Korea, China, India, and Turkey are the large consumers of petrochemicals, and the aromatic compounds primarily originate from the volatilization of used petrochemicals. The country ranking in terms of CR_{max} was of the following descending order: Kuwait>South Korea>China>Iran>India>Turkey. Kuwait, South Korea, and Iran were at a high risk from individual landfill pollutants but with fewer pollutants with carcinogenic risk. Overall, landfills in China were the ones that most threatened human health. A large variety of landfill pollutants in China caused carcinogenic risks, with predominantly halogenated compounds, such as trichloroethylene and methylene chloride, followed by aromatic compounds, such as styrene and benzene. Commonly used to make sanitary balls in China before 1993, naphthalene was also a potential source of carcinogenic risks. In addition, attention should be paid to formaldehyde, acetaldehyde, and acrylonitrile as these three pollutants not only pose high carcinogenic risks but also have been frequently detected in landfills. In India and Turkey, the carcinogenic risks of VOCs and SVOCs from landfills were rated low and largely associated with the oxygen, aromatic, and halogenated compounds. Most of the pollutants, such as benzene, styrene, and chloroform, showed the potential to cause carcinogenic risks in India but not in Turkey.

A large variety and quantity of VOCs and SVOCs with non-carcinogenic risks were found in Chinese landfills ([Figures S18 and S19](#)). The pollutants that exceeded the limits included the aromatic, halogenated, oxygen, and nitrogen compounds. In addition, carbon disulfide, frequently detected in Chinese landfills, exhibited non-carcinogenic risks (HI) well above the acceptable level. The

situations in South Korea and Kuwait were similar, as non-carcinogenic risks from landfills in both countries were caused by the aromatic compounds, predominantly benzene and xylene. A difference was that, in South Korea, carbon disulfide with high health risks were detected in multiple landfills. Landfills in Turkey, Iran, and India posed low health risks. There were 26 non-carcinogenic pollutants found in Turkish landfills, of which acetaldehyde, acrolein, 1,2-dichloropropane, 1,1-dichloroethylene, and trichloroethylene may pose non-carcinogenic health risks. In Iran, six non-carcinogenic pollutants were detected, among which benzene, xylene, and acetaldehyde showed the potential to cause non-carcinogenic health risks. Among the 18 non-carcinogenic pollutants detected in India, acrolein and benzene were the potential sources of human health risks.

(4) Other countries

Only few studies and data on VOCs and SVOCs existed from landfill in South American, Oceanic, and African countries, such as Brazil, Colombia, and South Africa. Considering the limited data, carcinogenic risks were based on the average concentrations of pollutants. Health risks primarily arose from formaldehyde and acetaldehyde in Brazil and from benzene in Colombia and South Africa ([Figure S11](#)). Acetaldehyde from landfills showed the potential to cause health risks in Brazil. Although detected, the aromatic compounds, such as acetone, toluene, and ethylbenzene, presented low concentrations and low risks. In Colombia and South Africa, no pollutants were detected that could harm human health ([Figure S12](#)).

Globally, the top two countries with comprehensive human health risks were China and the United States. In China, the landfill pollutants were characterized by high-risk values, rich diversity, and frequent detections, which are related to the rapid development of Chinese economy. Landfill in European countries generally posed a weak threat to workers and nearby residents.

3.3. Factors influencing the health risks of VOCs and SVOCs

After being produced from the biodegradation of waste or from their direct volatilization, VOCs and SVOCs enter the atmosphere through cover soils, thereby adversely affecting the life and health of residents around the landfill sites. Air temperature, relative humidity, air pressure, wind direction, and wind speed are the major factors that influence the health risks of VOCs and SVOCs. High temperatures aggravate the atmospheric pollution of VOCs and SVOCs, as they cause VOCs and SVOCs to diffuse fast in the air, thereby exerting a spatially wider negative impact on the environmental and public health. As water vapor in the air can adsorb VOCs and SVOCs, high relative humidity is not conducive to the diffusion of these compounds, thereby worsening the air pollution over landfills. In contrast, high air pressure decreases the atmospheric concentrations of VOCs and SVOCs, thereby decreasing both carcinogenic and non-carcinogenic risks. [Ding et al. \(2012\)](#) found that the air pollution and health risks of VOCs and SVOCs at a landfill site in Hangzhou severely grew under the high temperature, high relative humidity, and low air pressure. Sea breeze that increases air temperature, relative humidity, and air pressure could also exacerbate the

atmospheric pollution of VOCs and SVOCs over landfills in coastal areas. The wind direction and speed determine the diffusion direction and transport rate of VOCs and SVOCs, respectively. The faster the wind speed, the faster the migration rates of VOCs and SVOCs are and the stronger the dilution of the concentrations of VOCs and SVOCs, thereby decreasing health risks. VOCs and SVOCs not only transport in the atmosphere but also undergo chemical reactions under sunlight. [Duan et al. \(2014\)](#) detected more reduced sulfides in the air in winter owing to the decomposition and photochemical reactions of VOCs and SVOCs. [Allen et al. \(1997\)](#) suggested that the emissions of VOCs and SVOCs stimulate photochemical reactions in the air over landfills, thereby accumulating ozone, benzene, and vinyl chloride ([Butt et al., 2016](#)). Liu et al. (2016) revealed that BTEX can react with $-OH$ or NO_x to produce secondary organic aerosols, ozone, and other oxides, depending on the intensity of solar radiation, relative concentrations of aromatic compounds, and meteorological conditions. Atmospheric stability play an important role in the pollutant dispersion. [Tansel and Inanloo \(2019\)](#) estimated that their impact range expanded by 194–1164% in stable atmosphere conditions (early morning or night) compared to that in other conditions.

Several studies have been conducted on the transport distance of VOCs and SVOCs from landfills. As shown in [Figure S20](#), using the Gaussian model, [Liu et al. \(2018\)](#) quantified that reduced sulfide can travel as far as 1,000m. [Cai et al. \(2015\)](#) estimated from 1,955 landfills that VOCs and SVOCs transport in the range of 400–1,000m. [Lu et al. \(2013\)](#) determined that, among aromatic compounds released from a landfill site in Beijing, benzene can move approximately 1,500m under normal conditions. China's Pollution Control Standards for Municipal Solid Waste Landfills (Draft for Comments) - Preparation Notes (People's Republic of China, 2022) ([Pollution Control Standards., 2022](#)) required a buffer distance of 500–1,500m for the protection of residential areas from landfill gas. Currently, 5.57% of the landfills in China is located within 500m of residential areas, with 37.88% within 1,000m ([Table S6](#)), suggesting that landfills are already threatening the health of surrounding residents. The impact range of various VOCs and SVOCs emitted from landfills depends on multiple factors, such as landfill amount, waste composition, management level, geographical location, and meteorological conditions. In addition, complex mechanisms, such as superposition and synergy, play a significant role in the transport of VOCs and SVOCs; however, studies in this field of research are lacking.

4. Summary and outlook

Landfill gas is one of the major sources of VOCs and SVOCs in the environment. This study systematically addressed the composition characteristics, health risks, and influencing factors of VOCs and SVOCs and drawn the following main conclusions:

(1) Landfill gas is a non-negligible source of health risks for landfill workers and nearby residents. Landfill gas poses human health risks in almost all the countries considered, in particular, in China, followed by the developed European countries and the United States.

(2) The aromatic and halogenated compounds are the primary contributors to landfill gas hazards. As these substances primarily originate from the volatilization of chemical products, their hazards to human health can be reduced by improving the utilization rate of chemical products and reducing the total amount of these wastes entering landfills.

(3) The risks posed by compounds such as acetaldehyde, formaldehyde, and ethyl acetate also deserve special attention. Owing to their high concentrations and low thresholds, some VOCs and SVOCs caused human health risks beyond the acceptable levels. Typically, they were frequently detected with high exceedance rates.

(4) The health risks of VOCs and SVOCs from landfills are primarily influenced by air temperature, relative humidity, air pressure, wind direction, and wind speed. Owing to their increased transport distance, VOCs and SVOCs of different concentrations and hazards exerted a wider impact range and adversely affected residents within 1,000m of landfills.

This paper systematically summarized the composition, source and concentration of VOCs and SVOCs in landfill gas, and has a more complete and clear understanding of them. Potential human health risk assessment was conducted at the global, continental and national levels to fully understand the threat of VOCs and SVOCs to human health in different regions. However, due to the lack of toxicity parameters in other pathways, it is impossible to calculate the human health risk assessment of VOCs and SVOCs through other pathways. In addition, the propagation mechanism of VOCs and SVOCs diffusing from landfills to the air lacks further research. In the future, studies can be conducted to address the following aspects:

(1) Despite the non-negligible human health risks posed by VOCs and SVOCs, most of the previous studies only assessed their respiratory risks, with health risks from other pathways of exposure remaining unclear. Moreover, the lack of certain carcinogenicity parameters for VOCs and SVOCs renders it impossible to estimate their health risks.

(2) The reactions and transport pathways of VOCs and SVOCs in the atmosphere, as well as the mechanisms and roles of interactions between VOCs and SVOCs, remain poorly understood.

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.

Acknowledgments

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Appendix A. Supplementary material

The following are the Supplementary data to this article:

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Data availability

Data will be made available on request.

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...Exposure to toluene can cause a range of health effects, such as headaches, damage to the central nervous system [7], and cancer. Qi et al., [8] reported that aromatic compounds are the major sources of carcinogenic risks. Epidemiological studies proclaim that VOCs can cause various kinds of cancer [9]....

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What happens in a landfill, doesn't stay in a landfill

Reducing methane emissions from landfills is another powerful tool to combat climate change



Working face of an active landfill

When you toss your banana peel into the garbage, you're probably not thinking about how it will decompose in a landfill, emitting planet-warming carbon dioxide (CO₂) and methane gases into the atmosphere. But that's what happens.

In fact, decomposing organic matter in landfills (mostly food waste and yard debris) is a significant source of methane emissions in Washington, and municipal solid waste landfills are the third largest source of human-related methane emissions in the nation.

Methane gas, a super pollutant many times more damaging than CO₂ in the short term, is terrible for the climate.

That's why the Washington Legislature passed a new law in 2022 requiring owners and operators of municipal solid waste landfills, which are a certain category of landfills that may be either public or privately owned, to adopt stricter requirements for controlling methane.

We recently [proposed a new rule](#) to implement this law and put Washington at the forefront of the issue, along with California, Oregon and Maryland, which have passed similar rules.

What's the urgency?

To understand why it's important to quickly reduce methane emissions, consider these sobering facts:

Typically, 99% of landfill gas is CO₂ and methane in roughly equal proportions.

Methane is a potent greenhouse gas, just like CO₂, but it has approximately 83 times the global warming potential (or "GWP") of CO₂ during its first 20 years in the atmosphere.

Methane is responsible for more than 25% of the global warming experienced today.

Three methods of controlling methane gas

When adopted, the new rule will require landfill owners and operators to implement at least one of three methods to control methane emissions:

1. Combust methane with flares (open and enclosed)

The new rule requires landfills to eventually stop using open flares and to use enclosed flares that destroy 99% of methane gas. Most landfills have at least a flare, which burns methane gas, but open flares aren't as efficient as enclosed flares. Enclosed flares cover the flame in a cylindrical shroud, which helps distribute temperature evenly, destroying methane more efficiently.

2. Combust methane to create energy

Under the new rule, landfill owners and operators could use energy recovery devices – such as combustion engines or gas turbines – that destroy 97% of all methane gas.

Combusting or burning methane to generate energy requires separating methane gas from other landfill gas and then sending it to internal combustion engines or gas turbines, where it is combusted and turned into electricity.

3. Treat and process methane to sell

The new rule will require treatment and processing systems to maintain a methane leak rate of 3% or less. For methane to be sold and used, it must be separated from other components of landfill gas using treatment and processing technologies. When methane is pure enough, it's considered a renewable natural gas that can power transportation, cooking, home heating, electricity, and more.



An enclosed flare

"All three of these methods end with methane being combusted and turned into water and carbon dioxide," says Philip Gent, engineering and permitting section manager in the Air Quality Program. "As methane is more detrimental in the short term, the conversion of methane to carbon dioxide reduces the impact to the environment."

Most Washington landfills already have systems in place to control their methane gas emissions. However, some will need to install new equipment to meet the regulations. The State Legislature has appropriated \$15 million from Climate Commitment Act funds to help landfill owners obtain the technology required for compliance, and Ecology anticipates opening a new grant program in summer 2024 to distribute the money.



A compressor station, which is part of a landfill gas collection system

Protecting the environment and public health

Unfortunately, the combustion of landfill gas generates pollutants itself. However, a recent supplement to an Ecology [State Environmental Policy Act \(SEPA\) report](#) found that flares and other combustion devices are still the best way to reduce methane and other toxic air contaminants contained in landfill gas. Research shows that combusting landfill gas is better for public health and the environment than emitting methane to outdoor air without controls.

Perhaps the best way to prevent methane from escaping from landfills is to make sure the banana peel never makes it to the trash in the first place. “The reduction of organic waste in the landfill directly results in less methane produced in the landfill,” says Gent.

This is the goal of the [Organics Management Law](#) of 2022, which requires state and local governments, businesses, and other organizations to divert organic materials from municipal solid waste landfills toward beneficial uses, such as creating compost and crop nutrients, and getting food to people at risk of hunger.

Until all organic waste is diverted from landfills, this new rule will help mitigate harmful effects of methane emissions on the planet and people. It is another important way Washington is leading the effort to combat climate change and taking steps to reduce greenhouse gas emissions in the state by 95% below 1990 levels by 2050.

“Reducing methane emissions is one of the best strategies for mitigating the impacts of climate change in the near term,” says Bill Flagg, air quality planner and rulemaking lead.

While there are exceptions, this law will apply to all municipal landfills that received solid waste after Jan. 1, 1992.

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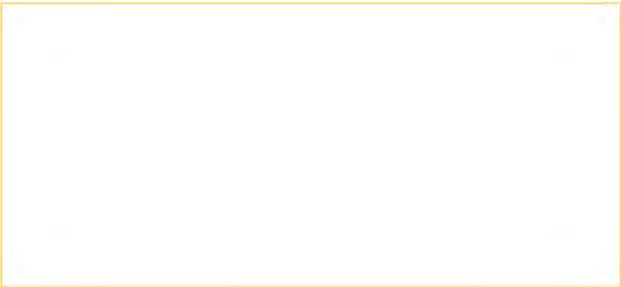


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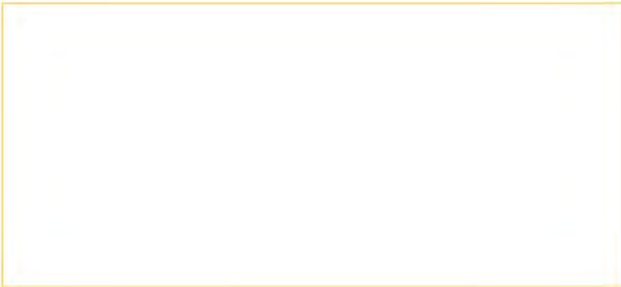
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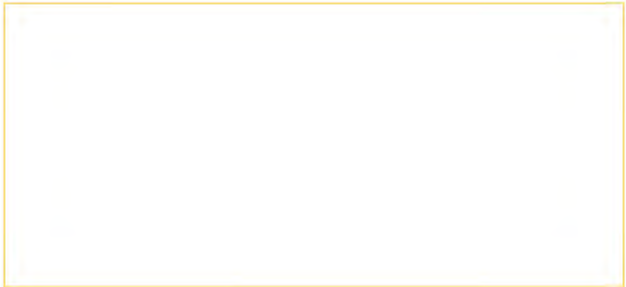
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