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Poly- and Perfluoroalkyl Substances (PFAS) in Landfills: Occurrence, Transformation and Treatment

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ABSTRACT

Landfills have served as the final repository for > 50 % municipal solid wastes in the United States. Because of their widespread uses and persistence in the environment, per- and polyfluoroalkyl substances (PFAS) (>4000 on the global market) are ubiquitously present in everyday consumer, commercial and industrial products, and have been widely detected in both closed (tens ng/L) and active (thousands to ten thousands ng/L) landfills due to disposal of PFAS-containing materials. Along with the decomposition of wastes in-place, PFAS can be transformed and released from the wastes into leachate and landfill gas. Consequently, it is critical to understand the occurrence and transformation of PFAS in landfills and the effectiveness of landfills, as a disposal alternative, for long-term containment of PFAS. This article presents a state-of-the-art review on the occurrence and transformation of PFAS in landfills, and possible effect of PFAS on the integrity of modern liner systems. Based on the data published from 10 countries (250 + landfills), C4-C7 perfluoroalkyl carboxylic acids were found predominant in the untreated landfill leachate and neutral PFAS, primarily fluorotelomer alcohols, in landfill air. The effectiveness and limitations of the conventional leachate treatment technologies and emerging technologies were also evaluated to address PFAS released into the leachate. Among conventional technologies, reverse osmosis (RO) may achieve a high removal efficiency of 90–100 % based on full-scale data, which, however, is vulnerable to the organic fouling and requires additional disposal of the concentrate. Implications of these knowledge on PFAS management at landfills are discussed and major knowledge gaps are identified.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of fluorinated aliphatic chemicals that contain at least one carbon atom on which all the hydrogen atoms are replaced by fluorine atoms (Buck et al., 2011). These synthetic chemicals have been commercially manufactured in 1950s (3M,2022). Due to their extraordinary water- and oil-resistance, chemical stability, and heat resistance (Darlington et al., 2018), PFAS have been widely used in consumer, commercial and industrial products or processes, within>200 categories and subcategories, including fire-fighting foam, textile and apparel, electroplating, ammunition, and others (Glüge et al., 2020).

Due to the unique molecular structure and strong C–F bond energy (536 kJ mol^{-1}), PFAS are very persistent to environmental degradation. Conventional biological and abiotic water/wastewater treatment processes have been reported unable to effectively degrade PFAS. As such, PFAS have been widely detected in various environmental media including water (Crone et al., 2019), sediment (White et al., 2015), soil (Brusseau et al., 2020; Schroeder et al., 2021), air (Barber et al., 2007; Morales-McDevitt et al., 2021) and wildlife even in the remote artic areas (Kelly et al., 2009). A widespread presence of PFAS in human blood has also been documented in the United States (Olsen et al., 2017) and Europe (Göckener et al., 2020).

Increasing toxicological data have associated PFAS exposure with various adverse health effects such as damage to the liver and immune system (Ojo et al., 2020), birth defects (Fenton et al., 2021) and delayed development (Gaballah et al., 2020). While most studies have been focused on understanding health risks associated with the legacy long-chain PFAS such as perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) (Fenton et al., 2021), recent works also reported that some emerging short-chain substitutes may pose comparable

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Nomenclature

AC	Activated carbon
BDD	Boron-doped diamond
C&D	Construction and demolition
Cl-PFAES	Chlorinated polyfluorinated ether sulfonic acid
COD	Chemical oxygen demand
DiPAP	Di-substituted polyfluorinated phosphate ester
DiSAmPAP	Bis(*N*-ethyl perfluoroalkylsulfonamidoethane) phosphate
DOM	Dissolved organic matter
FASA	Perfluoroalkane sulfonamide
FASSA	Perfluoroalkane sulfonamido acetic acid
FASE	Perfluoroalkyl sulfonamidoethanol
Fe/TNTs@AC	Fe-doped carbon-supported titanate nanotubes
FOSA	Perfluorooctane sulfonamides
FOSE	Perfluorooctane sulfonamidoethanols
FTCA	Fluorotelomer carboxylic acid
FTMAP	Fluorotelomer mercaptoalkyl phosphate ester
FTOH	Fluorotelomer alcohol
FTSA	Fluorotelomer sulfonate
FTUCA	Fluorotelomer unsaturated carboxylic acid
GCL	Geosynthetic clay liner
HDPE	High density polyethylene
IX	Ion exchange
LLDPE	Linear low-density polyethylene
MSW	Municipal solid waste
PFAA	Perfluorinated alkyl acids
PFAS	Per- and polyfluoroalkyl substances
PFCA	Perfluoroalkyl carboxylate
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPA	Perfluoroalkyl phosphonic acid
PFPiA	Perfluoroalkyl phosphinic acid
PFSA	Perfluoroalkyl sulfonate
PFSiA	Perfluoroalkyl sulfinate
RO	Reverse osmosis
TOC	Total organic carbon
UV	Ultraviolet

toxic effects (Nian et al., 2020; Rericha et al., 2022).

Landfills have been serving as the major final repository for long-term disposal of solid waste from residential, commercial and industrial sources for many decades. Not surprisingly, PFAS have been widely detected in both historic and active landfills (Propp et al., 2021). PFOA and PFOS have been reported at concentrations of up to ppb or higher levels in the leachate (Solo-Gabriele et al., 2020) and at up to ng/m^3 in the air (Wang et al., 2020) at municipal solid waste (MSW) landfills. To ensure the long-term effectiveness and protectiveness of landfill facilities for PFAS containment, it is very critical to understand the occurrence and transformation of PFAS at landfills and the possible effect of PFAS on the integrity of the liner system. Cost-effective technologies to remove/treat PFAS in leachate are also urgently needed to prevent potential release of PFAS into the environment.

Several reviews have been published about PFAS in landfills. Hamid et al. (2018) discussed the occurrence and trends of PFAS based on studies published prior to 2018. Gates et al. (Gates et al., 2020) discussed the potential interactions between PFAS and landfill liners based on limited studies on the actual performance of liners in the presence of PFAS. Wei et al. (2019) and Travar et al. (2020) reviewed technologies for treating PFAS in landfill leachate focusing on the treatment chemistry. However, studies and publications on treatment of PFAS, including leachate treatment, have been rapidly growing in recent years, and there is a need to critically review and update the latest knowledge on treating PFAS in landfill leachate. To fill out this critical knowledge gap, this review article aims to provide a comprehensive, state-of-the-art review on the occurrence and transformation of PFAS in various landfill media from solid waste to leachate and landfill air, and treatment of PFAS at landfills, with a special focus on real-world leachate conditions, liner integrity, and technology implementability. The specific objectives were to: 1) overview the occurrence and transformation of PFAS in landfills; 2) identify factors that could impact the PFAS release from waste; 3) examine potential effect of PFAS on the integrity of the modern liner system based on the current state of knowledge; 4) discuss the state of treatment technologies for PFAS in landfill leachate and their compatibility with landfill operations, and 5) identify key knowledge gaps and propose future actions for improved management of PFAS in landfills.

2. Occurrence and fate of PFAS in landfills

2.1. PFAS in solid waste

Due to its widespread use, PFAS have been detected in different types of solid waste. Venkatesan and Halden (2013) investigated the U.S. sewage sludge for 13 PFAS and found the mean total concentration of PFAS was 539 ± 224 ng/g-dry-weight (dw). PFOS was the most abundant PFAS tested with an average concentration of 403 ± 127 ng/g-dry-weight, followed by PFOA (34 ± 22 ng/g dw) and PFDA (26 ± 20 ng/g dw). Choi et al. (2019) analyzed MSW composts collected from five U.S. states for 17 PFAAs and found that short-chain perfluoroalkyl acids (PFAAs) and perfluoroalkyl carboxylates (PFCAs) were predominated in MSW composts. Specifically, PFAAs ranged from 28.7 to 75.9 ng/g dw for composts that included food packaging waste and 2.38 to 7.6 ng/g for those without the food packaging waste. The remarkable contribution of food packaging waste to the elevated PFAS content is not surprising because PFAS have been used as grease-proofing agents in many compostable food packaging products (FDA, 2020).

Munoz et al. (2022) characterized PFAS in 47 organic waste products in France and reported the following sequence of PFAS concentrations in different wastes: municipal sewage sludge and related compost ≈ compost of residual domestic wastes ≫ compost of municipal biowaste > digestate of urban waste ≫ agriculture wastes such as pig slurry, poultry manure, or dairy cattle manure. Their reported values are higher than those reported by Venkatesan and Halden (2013) and Choi et al. (2019) likely because this French study included 160 PFAS of 42 classes for analysis. The authors indicated the sum of some commonly targeted PFAAs in their tested urban wastes only represented on average 27 % of the total PFAS, suggesting that limiting PFAS analysis to a narrowly defined group could lead to dramatic underestimation of the total PFAS.

The composition of PFAS in solid waste have also changed remarkably over the last decade or so likely due to the shift of PFAS production and usage from long-chain to shorter chain alternatives. Munoz et al. (2022) found historical municipal biosolids and composts (1976–1998) from metropolitan France and Réunion Island were dominated by PFOS, *N*-ethyl perfluorooctanesulfonamido acetic acid, and cationic and zwitterionic electrochemical fluorination precursors to PFOS, while recent urban organic waste products (2009–2017) were dominated by zwitterionic fluorotelomers with fluorotelomer sulfonamidopropyl betaines as the prevalent class.

It is evident that data in terms of PFAS content in different types of solid waste is still limited. In addition, the majority of current studies have targeted only a small fraction of PFAS (<200 out of > 4000 on the global market), which can lead to serious underestimation. In terms of the compositional profile, there appears to be a trend to shift from long-chain to shorter-chain PFAS in solid wastes, which is in line with, but may not be fully accounted for by the recent substitution of some legacy PFAS (PFOS and PFOA) with shorter chain PFAS (e.g., GenX and PFBA).

2.2. PFAS in landfill leachate

PFAS in landfill leachate have been widely studied in recent years. Table 1 presents a summary of the type and levels of the most targeted PFAS in untreated landfill leachate from > 200 landfills (mostly for MSW) across 10 major countries. It should be noted that the number and type of PFAS tested varied widely from site to site, from the legacy PFAS (PFOA and PFOS) to 70 PFAS covering 13 classes of PFAS. Perfluoroalkyl carboxylates (PFCAs), perfluoroalkyl sulfonates (PFSAs), perfluoroalkyl sulfonamides and fluorotelomer sulfonates have been the most often targeted PFAS categories in these studies.

In general, C4-C7 PFCAs have been found most abundant in landfill leachate at United States (Huset et al., 2011), Europe (Fuertes et al., 2017) and Asia (Kameoka et al., 2022), while longer chain PFAS such as PFOS and PFOS-precursors were found dominant in sediment (Knutsen et al., 2019). The dominance of shorter-chain PFCAs in untreated leachate has been attributed to: 1) higher aqueous solubility and lower organic carbon–water partition coefficients than their longer-chain and sulfonate analogs (Higgins and Luthy, 2006); 2) a shift of industrial production from long-chain to short-chain PFAS in recent decades globally (Gallen et al., 2016; Yan et al., 2015); and 3) potential transformation to shorter-chain PFAAs under landfill conditions (Hamid et al., 2020a). Liu et al. (2021a) reported that the majority of PFAS present in the fresh liquid from waste collection vehicles were dominated by PFAA-precursors such as 6:2 fluorotelomer sulfonate (FTS) and 6:2 diPAP; however, the leachate from the landfills where the waste was deposited was dominated by PFAAs, suggesting the transformation of precursors to PFAAs occurred in the landfill.

Besides shorter-chain PFCAs, Lang et al. (2017) reported the abundance of 5:3 fluorotelomer carboxylic acid (FTCA) in leachate from 18 U.S. MSW landfills. The concentrations of 5:3 FTCA was at least three times higher than PFOA and PFOS combined (2.6 to 41,309 ng/L). Abundant 5:3 FTCA was also reported for leachate collected from five landfills in Florida, United States (Solo-Gabriele et al., 2020). Not only in the field studies, a laboratory study with model anaerobic landfill reactors filled with MSW also reported 5:3 FTCA being the most concentrated PFAS after the onset of methanogenesis, suggesting the occurrence of significant biotransformation of the C6 telomer precursors in the MSW (Allred et al., 2015). 5:3 FTCA is a known degradation product of fluorotelomer compounds (Abada et al., 2018). It appeared stable under methanogenic conditions (Allred et al., 2015), but it can be degraded under aerobic conditions (Wang et al., 2012). Thus its concentration can be significantly reduced in leachate after on-site treatment with aeration (Solo-Gabriele et al., 2020; Yin et al., 2017). Busch et al. (2010) analyzed three other FTCAs, namely, 2-perfluorohexylethanoic acid, 2-perfluorooctylethanoic acid, and 2-perfluorodecylethanoic acid in leachate from seven German landfills and found their contribution to the total PFAS was insignificant.

Although the production of PFOA and PFOS was voluntarily phased out by some major manufacturers in Europe and north America since early 2000s, they were still commonly detected in landfill leachate oftentimes among the most abundant PFAS in Asia (Yan et al., 2015), Australia (Gallen et al., 2017), Europe (Knutsen et al., 2019). The phase-out or restricted uses of PFOS and PFOA didn’t immediately eliminate them from leachate. This can be attributed to: 1) the inherent persistency of PFAS, 2) the prolonged leaching process from waste in landfills, 3) the wide variation in service lives of existing PFOS- and PFOA-containing products, which extends their disposal time at landfills, 4) continued use of existing stocks of PFOA/PFOS in the market, and 5) transformation of PFAA-precursors under landfill conditions(Hamid et al., 2020b; Zhang et al., 2013).

2.3. Key factors affecting PFAS level in landfill leachate

A number of studies have been made to identify potential relationships between PFAS concentration in landfills and the waste characteristics, leachate quality and environmental factors.

2.3.1. Waste type

Researchers have attempted to examine potential correlations between the PFAS concentration in leachate and the type of waste deposited in landfills. However, no clear correlation has been identified. Gallen et al. (2017) investigated 27 landfills in Australia and found landfills accepting primarily construction and demolition (C&D) waste (>50 % C&D) gave higher concentrations of PFAS than those accepting primarily MSW (>50 % MSW); however, they found the differences in mean concentrations were statistically significant due to high variability in both the PFAS concentration and waste profiles. In another study conducted in Victoria, Australia, Simmons (2019) reported no significant variation in PFAS concentrations by waste type at 19 landfills. In a recent study conducted in the U.S., Solo-Gabriele et al. (2020) also reported similar levels of total PFAS in untreated C&D and MSW leachate. However, the researchers in the U.S. noticed that the C&D leachate contained higher levels of PFHxS, likely due to its uses in sealants and waste repellants in building materials.

These findings are somewhat surprising given the variation of PFAS content in solid waste (**Section 2.1**); however, it may reflect the fact that PFAS has been widely applied in a variety of materials and consumer products. It should also be cautioned that these studies targeted only the dominant wastes and a small group of most commonly used PFAS, which were lumped as total PFAS for comparison. Potential transformation and “mixing” of PFAS from different types of waste materials need to be taken into account in the future studies.

2.3.2. Landfill age

PFAS concentrations in closed landfills/landfill cells were generally reported lower compared to active landfills. For example, in a landfill leachate study conducted at Germany (Busch et al., 2010), a total PFAS concentration of 31 ng/L was reported for leachate from a landfill closed in 1979, while the total PFAS concentrations in active landfills ranged from 1,889 ng/L and 12,819 ng/L. Similar results were reported in another two studies in Australia (Gallen et al., 2017; Gallen et al., 2016); however, Gallen et al. (2017) found the differences in mean concentrations were not statistically significant when the tested landfills were closed more recently (between 1993 and 2012).

In addition, concentrations of selected PFAS in leachate have been reported decreasing with the landfill age. For instance, lower concentrations of PFNA, PFBS and four PFAAs precursors (8:2 FTCA, 5:3 FTCA, MeFBSAA, MeFOSAA) were observed in leachate from older landfills among 18 MSW landfills tested in the United States (Lang et al., 2017). It was attributed to lowered mass of PFAS in more aged landfills and/or the recent change of PFAS profile due to the replacement of PFBS and PFNA for PFOS and PFOA (Lang et al., 2017). Likewise, a significant correlation between the concentration of PFBS and PFNA and landfill age was reported for 40 MSW landfills in Ireland (Harrad et al., 2019). Gallen et al. (2017) also reported a statistically significant negative correlation between PFHxA, PFHpA, PFOA, PFHxS and PFOS concentrations and landfill age in 27 landfills/landfill cells (including closed landfills) in Australia. However, Simmons (2019) and Solo-Gabriele et al. (2020) reported no significant correlation between PFAS concentrations and landfill age in their studies at Australia and United States. It may be due to the mixing of PFAS from different types of solid waste (including MSW, industrial waste, C&D, MSW ash) deposited in the studied landfills.

Table 1
Occurrence of PFAS in Untreated Landfill Leachate.

Country	Number of Landfills	Primary Waste Type	Years of Waste Disposal	Number of PFAS analyzed	Group of PFAS Analyzed	PFAS Analyzed	Dominating PFAS Chemicals	PFBS µg/L	PFHxA µg/L	PFHxS, µg/L	PFPeA µg/L	PFHpA µg/L	PFOA µg/L	PFOSµg/L	References
Australia	19	Mixed putrescible waste, solid inert waste and prescribed industrial waste (hazardous)	1964 - current	15	PFCAs, PFSAs, FASSAs, FTSAs	PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, MeFOSAA, EtFOSAA, 6:2FTS, 8:2 FTS	PFBS, PFHxA, PFOA, PFHxS, PFPeA, PFHpA, PFOS	0.04 – 23	0.11 – 7.74	0.03 – 2.41	ND – 2.07	0.04 – 1.7	0.09 – 3.12	0.02 – 1.2	Simmons (2019)
Australia	27	MSW, C&I, C&D	1969 - current	9	PFCAs, PFSAs	PFOA, PFOS, PFHxA, PFHpA, PFNA, PFDA, PFUnDA, PFDoDA, PFHxS	PFHxA, PFHxS, PFOA, PFOS, PFHpA	–	0.073–25	0.056–16	–	0.018–4.4	0.017–7.5	0.013–2.7	Gallen et al. (2017)
Australia	13	MSW, C&I, C&D, green waste	Closed; 1995 - Current	14	PFCAs, PFSAs	PFOA, PFOS, PFBA, PFHxA, PFHpA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, PFDS	PFHxA, PFHpA, PFOA	ND – 0.84	0.011–5.7	ND – 1.9	–	0.0022–3.5	0.019–2.1	0.037–1.1	Gallen et al. (2016)
Canada	28	–	–	13	PFCAs, PFSAs, FASAs	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFBS, PFHxS, PFOS, and PFOSA	PFHxA, PFBA, PFPeA, PFHpA, PFOA	–	0.695 (mean)	–	–	–	0.439 (mean)	0.279 (mean)	Li et al. (2012)
Canada	10	MSW, construction waste, industrial waste, and sewage sludge	–	2	PFCAs, PFSAs	PFOA, PFOS	–	–	–	–	–	–	0.0503–1.59	<0.0095–0.744	Gewurtz et al. (2013)
China	5	MSW	–	14	PFCAs, PFSAs	PFPrA, PFBA, PFPeA,PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA,PFTA, PFBS, PFHxS and PFOS	PFOA, PFBS, PFPrA	1.6–41.6	0.146–4.43	ND – 0.479	0.609–6.53	0.0754–5.83	0.281–214	1.15–6.02	Yan et al. (2015)
Finland	1	–	–	4	PFCAs, PFSAs	PFOA, PFOS, PFHxA and PFDA	PFOA, PFHxA, PFOS	–	0.049–0.2	–	–	–	0.076–0.27	0.087–0.14	PerKola and Sainio (2013)

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Table 1 (*continued*)

Country	Number of Landfills	Primary Waste Type	Years of Waste Disposal	Number of PFAS analyzed	Group of PFAS Analyzed	PFAS Analyzed	Dominating PFAS Chemicals	PFBS µg/L	PFHxA µg/L	PFHxS, µg/L	PFPeA µg/L	PFHpA µg/L	PFOA µg/L	PFOSµg/L	References
Country	**Number of Landfills**	**Primary Waste Type**	**Years of Waste Disposal**	**Number of PFAS analyzed**	**Group of PFAS Analyzed**	**PFAS Analyzed**	**Dominating PFAS Chemicals**	**PFBS µg/L**	**PFHxA µg/L**	**PFHxS, µg/L**	**PFPeA µg/L**	**PFHpA µg/L**	**PFOA µg/L**	**PFOSµg/L**	**References**
Germany	7	Classes I, II, partial unknown waste, one Class II only with filter cake from a WWTP	–	43	PFCAs, PFSAs, PFSiAs, PFPAs, FASAs, FASEs, FTCAs, FTUCAs, FTSAs	PFBA, PFPS, PFHxS, PFHpS, PFOS, PFNS, PFDS, 6:2 FTS, PFHxSi, PFOSi, PFDSi, PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFPeDA, PFHxDA, PFHpDA, PFOcDA, 3,7m2-PFOA, PFOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE, MeFBSA, MeFBSE, FHEA, FOEA, FDEA, FHUEA, FOUEA, FDUEA, PFHpPA, PFOPA, PFDPA	PFBA, PFBS, PFHxA, PFOA	–	–	–	–	–	–	–	Busch et al. (2010)
Ireland	40	MSW	–	10	PFCAs, PFSAs, FASAs, FASEs	PFOS, PFBS, PFHxS,PFOA, PFNA,PFOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE	PFBS, PFOA, PFOS, PFHxS, PFNA	<0.0001–17	–	<0.0001–2.6	–	–	0.009–11	<0.0001–7.4	Harrad et al. (2019)
Japan	2	MSW	1974 - Present	17	PFCAs, PFSAs	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTrDA, PFTeDA, PFHxD, PFODA, PFBS, PFHxS, PFOS, PFDS	PFBA, PFPeA, PFHxA, PFOA, PFODA	0.013–0.13	0.048–0.15	0.0078–0.0087	0.03–0.27	0.037–0.038	0.012–0.28	0.0055–0.1	Kameoka et al. (2022)
	3	Industrial	1994 - Present				PFBS, PFBA, PFPeA, PFHxA, PFOA	0.21–27	0.08–23	0.022–0.17	0.014–12	0.0055–3.2	3–24	0.099–0.38	

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Norway	2	MSW	1972–2006	16	PFCAs, PFSAs, FASAs, FTS, FTUCA	PFBS, PFHxS, PFOS, PFDcS, PFBA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFOSA, *N*-EtFOSA, 6:2 FTS, 6:2 FTUCA	PFOS, PFHxA, PFOA, PFNA, PFHpA, PFHxS	<0.005	0.59–0.757	0.089–0.281	–	0.215–0.277	0.532–0.767	0.455 – 2.92	Eggen et al. (2010)
Country	**Number of Landfills**	**Primary Waste Type**	**Years of Waste Disposal**	**Number of PFAS analyzed**	**Group of PFAS Analyzed**	**PFAS Analyzed**	**Dominating PFAS Chemicals**	**PFBS µg/L**	**PFHxA µg/L**	**PFHxS, µg/L**	**PFPeA µg/L**	**PFHpA µg/L**	**PFOA µg/L**	**PFOSµg/L**	**References**
Norway	10	Primarily MSW, in some cases industrial Waste, and contaminated soil and sewage sludge,	1960 - Present	28	PFCAs, PFSAs, FTSAs, FTOHs, FASAs, FASEs	PFBA, PFPeA, PFHxA, PFBS, PFHpA, HPFHpA, PFOA, PFNA, PFDA, PF-3,7-DMOA, PFUnDA, PFDoA, PFTrA, PFTA, PFHxDA, PFHxS, PFHpS, PFOS, PFDS, 4: 2 FTSA, 6:2 FTSA, 8: 2 FTSA, 8: 2 FTOH, FOSA, EtFOSA, MeFOSA, EtFOSE and MeFOSE	PFBA, PFPeA, PFHxA, PFBS	0.0071–4.2	0.05–1.4	0.011–0.15	0.029–0.58	0.019–0.77	0.066–1.8	0.015–0.16	Knutsen et al. (2019)
Spain	4	MSW	–	16	PFCAs, PFSAs	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, PFHpS, PFOS, PFDS	PFOA, PFHxA, PFPeA, PFBA	ND – 0.529	0.102–0.692	ND	ND - 0.267	ND - 0.078	0.387–0.584	ND - 0.043	Fuertes et al. (2017)
United States	5	MSW	–	2	PFCAs, PFSAs	PFOA, PFOS	–	–	–	–	–	–	0.177–2.5	0.026–0.092	Clarke et al. (2015)
United States	18	MSW	1966 - Present	70	PFCAs, PFSAs, FTSAs, n:2 FTCAs, n:3 FTCAs, FTUCAs,		5:3 FTCA	0.0034–3.4	0.0413–8.858	ND – 1.328	0.0234–3.194	0.032–3.133	0.0301–4.989	ND – 0.801	Lang et al. (2017)

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Country	Number of Landfills	Primary Waste Type	Years of Waste Disposal	Number of PFAS analyzed	Group of PFAS Analyzed	PFAS Analyzed	Dominating PFAS Chemicals	PFBS µg/L	PFHxA µg/L	PFHxS, µg/L	PFPeA µg/L	PFHpA µg/L	PFOA µg/L	PFOSµg/L	References
					FASAAs, MeFASAA, EtFASAA, DiPAP, DiSAmPAP, PFPIA, FTMAP										
United States	4	MSW with some non-municipal solid waste (e.g., industrial, construction, and demolition)	1982 - Present	24	PFCAs, PFSAs, FASAs, FTSA, FASAAs	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTDA, FOUEA, PFBS, PFHxS, PFOS, PFDS, 6:2 FTS, 8:2 FTS, Me-FBSA, Me-FBSAA, FOSA, FOSAA, Me-FOSAA, Et-FOSAA.	PFBA, PFPeA, PFHxA, PFHpA, PFBS	0.28–0.89	0.27–0.79	0.16–0.7	0.12–1.5	0.1–0.34	0.38–1	0.056–0.16	Huset et al. (2011)
Country	**Number of Landfills**	**Primary Waste Type**	**Years of Waste Disposal**	**Number of PFAS analyzed**	**Group of PFAS Analyzed**	**PFAS Analyzed**	**Dominating PFAS Chemicals**	**PFBS µg/L**	**PFHxA µg/L**	**PFHxS, µg/L**	**PFPeA µg/L**	**PFHpA µg/L**	**PFOA µg/L**	**PFOSµg/L**	**References**
United States	5	MSW, C&D, MSW ash (MSWA), MSWA/MSW-gas condensate (GC)	1981 - Present	11	PFCAs, PFSAs, FTCAs	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, 5:3 FTCA	PFHxA, PFHxS, PFBS	ND − 5.51	0.589–4.27	0.176–4.63	ND − 1.79	0.255–1.32	0.259–2.86	0.12–1	Solo-Gabriele et al. (2020)

Table 2
Occurrence of PFAS in Air at Landfills.

Country	Number of Landfills	Primary Waste Type	Years of Waste Disposal	Number of PFAS analyzed	Group of PFAS Analyzed	PFAS Analyzed	Dominating PFAS Chemicals	8:2 FTOH, pg/m3	6:2 FTOH, pg/m3	10:2 FTOH, pg/m3	PFBA, pg/m3	PFOA, pg/m3	References
Canada	2	MSW	–	22	PFSAs, PFCAs, FTOHs, FOSAs, FOSEs	PFBS, PFHxS, PFOS, PFDS, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA, PFOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH	8:2 FTOH, 6:2 FTOH, 10:2 FTOH, PFBA	1,290–17,381	987–6,462	310–2,151	101–102	2.98–46.2	Ahrens et al. (2011)
China	3	MSW	–	29	FTOHs, FOSAs, FOSEs, PFCAs, PFSAs, FTUCAs, FTSAs, Cl-PFAESs, diPAPs	6:2 FTOH, 8:2 FTOH, 10:2 FTOH, *N*-MeFOSA,*N*-EtFOSA, *N*-MeFOSE, *N*-EtFOSE, TFA, PFPrA, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoDA, PFBS, PFHxS, PFOS, 6:2 FTUCA, 8:2 FTUCA, 6:2 FTSA, 8:2 FTSA, 6:2Cl- PFAES, 8:2Cl- PFAES, 6:2 diPAP, 8:2 diPAP.	8:2 FTOH, 6:2 FTOH, 10:2 FTOH, PFBA	164–11,723	88–7,704	8.6–1,150	N.D. – 717	129–8,781	Wang et al. (2020)
China	2	MSW	–	23	FTOHs, FOSAs, FOSEs, PFCAs, PFSAs, diPAPs	6:2 FTOH, 8:2 FTOH, 10:2 FTOH, *N*-MeFOSA,*N*-EtFOSA, *N*-MeFOSE, *N*-EtFOSE, TFA, PFPrA, PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoDA, PFBS, PFHxS, PFOS, 6:2 diPAP, 8:2 diPAP.	8:2 FTOH, 10:2 FTOH, PFBA, PFOA	42–1,600	ND – 110	13–540	180–590	22–100	Tian et al. (2018)
Germany	2	Sanitary, compost, plastic, electronic devices	–	30	FTOHs, FTAs, FASAs, FASEs, PFSAs, PFCAs	4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, 12:2 FTOH, 6:2 FTA, 8:2 FTA, 10:2 FTA, MeFOSA, EtFOSA, MeFBSA, MeFOSA, PFOSA, MeFOSE, MeFBSE, EtFOSE, PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFBS, PFHxS, PFHpS, PFOS	FTOHs, PFBA	41–433.6	8.2–102.8	8.8–92.7	ND – 9.5	ND – 1.2	Weinberg et al. (2011)
Ireland	10	MSW	–	8	PFSAs, PFCAs, FOSAs, FOSEs	PFOS, PFBS, PFNA, FOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE.	PFOA	–	–	–	–	4.4–97	Harrad et al. (2019)

Based on these findings, though limited, PFAS leaching from landfills may decay over time, and it may take over decades to reach the lower-ppt levels (e.g., <30 ng/L). The causes for the decay in the PFAS concentrations may be due to: 1) depletion of PFAS in the waste via desorption and physical leaching over decades, 2) limited desorption and physical leaching after installation of final cover over the waste; 3) biotic and/or abiotic decomposition and transformation inside and outside the landfills, and 4) alteration of the source PFAS. However, such aging effects can vary depending on the type of waste as well as specific landfill settings and operating conditions.

2.3.3. *Operation mode*

Conventional landfills are normally operated like a “dry tomb”, which is designed to limit the liquid entering the disposed waste and thereby minimize the waste leaching. But it slows down the degradation of organic wastes and may pose long-term risks as the residual organics can continuously produce landfill gas and leachate and lead to settlement in the waste cells even after closure.

The concept of bioreactor landfill started in the early 1990s (Reinhart et al., 2002). Different from conventional landfilling, liquids (such as leachate, stormwater, wastewater, and/or sludge) are added in a controlled manner into the landfills to increase the water content and accelerate the biological processes for waste degradation and stabilization.

Laboratory-scale studies have shown that biological process could facilitate the release of certain PFAS into the leachate. Allred et al. (2015) studied the release of PFAS from MSW in laboratory-scale anaerobic bioreactors under the methanogenic conditions and observed higher concentrations of PFOS, PFOA, C9 and longer PFCAs, 6:2 FTCA and 5:3 FTCA in the bio-active reactors with/after the onset of methanogenesis, indicating biological processes were primarily involved in leaching of these PFAS. In a subsequent laboratory study, Lang et al. (2016) found the total PFAS concentrations in the bio-active reactor filled with carpets were one order of magnitude higher than in the abiotic reactors, confirming that biological processes were responsible for PFAS release from carpets; however, PFAS release from clothing was found more dependent on sample heterogeneity instead of biological activity.

The addition of liquids into wastes could accelerate waste decomposition. On the other hand, it may also lower PFAS concentrations in the leachate via dilution. In a study at two Canadian landfills, PFAS concentrations in leachate from a landfill with leachate recirculation were found generally lower than that from another landfill without leachate recirculation (Benskin et al., 2012). However, this study was based on one leachate sample from the landfill with leachate recirculation and its representativeness over the long term needs to be further verified.

The effect of bioreactor landfill operations on the PFAS concentrations in leachate has not been studied in the field scale yet. Laboratory studies have elucidated the role of biological waste decomposition on the temporal trends of PFAS in leachate; however, to enhance the rate of waste decomposition, some experimental conditions employed in the laboratory studies are not typical for field operations, such as a closed-loop leachate recirculation system and partially submerged waste. In this way, field data collected from bioreactor landfills becomes very critical to gain a better understanding of the effect of bioreactor operations on PFAS release from landfills.

2.3.4. *Leachate quality*

In general, wastes in the landfills undergo four (4) phases of decomposition/stabilization: 1) an initial aerobic phase with oxygen trapped in the waste, (2) an anaerobic acid phase after oxygen depletion, (3) an initial methanogenic phase with a reduction of CO_2 produced, and (4) a stable methanogenic phase (ATSDR, 2008). Along with waste decomposition, the composition of landfill leachate could change over time.

Significant positive correlations between PFAS concentrations and pH in leachate have been reported at landfills in Australia (Gallen et al., 2017), Ireland (Harrad et al., 2019), and the United States (Solo-Gabriele et al., 2020). The pH in MSW landfill leachate normally varies from 4.5 to 9 (Kjeldsen et al., 2002). It decreases in the anaerobic acid phase and becomes neutralized when the waste moves into the initial methanogenic phase (Kjeldsen et al., 2002). The change in pH not only affects the speciation of PFAS in leachate but also the surface charge of the waste materials (Higgins and Luthy, 2006), thereby affecting the PFAS-solids interactions and the adsorption/desorption rates. A higher pH could lower the adsorption of anionic PFAS by increasing repulsive electrostatic forces between the anionic functional groups of PFAS and negative surface potential of the solids (Wu et al., 2020).

Landfill leachate normally contains high concentrations of dissolved organic matter (DOM), ranging from 30 to 29,000 mg/L as total organic carbon (TOC) (Kjeldsen et al., 2002). Gallen et al. (2017) reported a significant positive correlation between the concentrations of nine PFAS (i.e., PFOA, PFOS, PFHxA, PFHpA, PFNA, PFHxS, PFDA, PFUnDA and PFDoDA) with TOC in landfills and attributed it to the hydrophobic interactions/partitioning of PFAS with/to the DOM in leachate.

Unlike DOM, a weak or statistically insignificant correlation has been reported between PFAS concentration and chemical oxygen demand (COD) (Solo-Gabriele et al., 2020; Yin et al., 2017). Although organic matters in leachate contribute to a large fraction of chemical oxygen demand (COD), both organic and inorganic components can contribute to the overall COD. Up to 1/3 of COD in leachate can be due to the inorganic substances such as ammonia and sulfides (Kylefors et al., 2003), which could complicate the correlation analysis with PFAS concentration.

Negative correlations have been reported between electrical conductivity and PFAS concentration in leachate (Yan et al., 2015; Yin et al., 2017). The elevated electrical conductivity normally reflects a higher content of electrolytes or a higher ionic strength. According to the classical double-layer theory, the higher ionic strength can compress the double layer and suppress the negative surface potential on the waste surface and subsequently enhance adsorption of anionic PFAS by reducing the electrostatic repulsion (Higgins and Luthy, 2006). Meanwhile, elevated electrical conductivity is usually associated with an increase in multivalent cations, which could act as bridges between negatively charged PFAS and negatively charged solids to facilitate adsorption. Moreover, high ionic strengths could pose a stronger “salting out effect” on hydrophobic chemicals, resulting in a decreased solubility and lower activity coefficient of PFAS in leachate (Kim et al., 2015). Nevertheless, Benskin et al. (2012) reported a positive correlation between electrical conductivity and the concentrations of 7 anionic PFAAs in leachate from a conventional MSW landfill. It was attributed to the weakened electrostatic attractions between PFAS anions and positively charged materials in the waste (such as boehmite and alumina) under high ionic strengths.

In short, the release of PFAS from waste materials could be impacted by the changes of pH, DOM and electrical conductivity in leachate. The adsorption/desorption of PFAS can be primarily explained by electrostatic interactions and hydrophobic interactions with waste materials, while other mechanisms such as ligand and ion exchange should also be considered. Leachate composition generally varies considerably between landfills or even waste cells depending on the waste composition, waste age, and landfilling technology (Kjeldsen et al., 2002), and could be one of reasons that lead to the wide variation of PFAS concentrations in leachate among landfills.

2.4. *PFAS in landfill air*

The release of PFAS from landfills into the atmosphere has been reported in North America (Ahrens et al., 2011), Europe (Harrad et al., 2020; Weinberg et al., 2011) and Asia (Tian et al., 2018; Wang et al., 2020). Table 2 presents a summary of studies on the occurrence of PFAS in the air at the MSW landfills. In these studies, neutral PFAS (fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamidoethanols (FOSEs) and perfluorooctane sulfonamides (FOSAs)) and anionic PFAS (PFCAs and PFSAs) have been most often tested.

Studies have demonstrated that neutral PFAS, primarily FTOHs and, to a lesser extent, FOSEs and FOSAs, were predominant in the air at landfill sites (Ahrens et al., 2011; Weinberg et al., 2011). Typically, 8:2 FTOH was found the most abundant, followed by 6:2 FTOH and 10:2 FTOH (Wang et al., 2020; Ahrens et al. 2011). 8:2 FTOH and 6:2 FTOH have been reported to be the most abundant PFAS in consume products (Vestergren et al., 2015), which are normally disposed of at MSW landfills. These chemicals have a stronger tendency to release into air due to their relatively lower solubilities ($1.32x10^{-11} \sim 4.8x10^{-5}$ mol/L) and higher vapor pressures ($0.2 \sim 876$ Pa at 25 °C) (ITRC, 2020; Lei et al., 2004).

Anionic PFAS have also been detected in the landfill air, with PFBA being the most abundant and PFHxA/PFOA to a lesser extent (Ahrens et al., 2011; Tian et al., 2018). The presence of PFBA has been often associated with the shift of PFAS manufacturing since the early 2000s (Wang et al., 2020; Weinberg et al., 2011). However, it is also noteworthy that the shorter-chain PFAS, especially C4 and C6 PFAS, can often result from decomposition of their longer-chain analogues, and they can be released into the air along with other typical gases (methane) as well as water vapor. Among PFCAs, Ahrens et al. (2011) found PFCAs of even-numbered carbons were more abundant than the odd-carbon-chain PFCAs in landfill air, and they attributed it to the presence of telomer-derived products that contain only even-carbon-chain PFCAs. Although PFOS was commonly detected in landfill leachate, its air emission was reported very limited at multiple sites (Ahrens et al., 2011; Tian et al., 2018; Weinberg et al., 2011), which can be due to its stronger adsorption to the solids.

In general, total PFAS concentrations were higher in the air at the central/downgradient of landfills compared to the upwind/reference sites (Ahrens et al., 2011; Glüge et al., 2020; Tian et al., 2018) and the concentrations at active landfills were higher than those at inactive landfills (Weinberg et al., 2011). However, in a study of 10 landfills in Ireland, Harrad et al. (2020) reported no significant difference between PFAS concentrations at downwind and upwind locations. It should be noted that FTOHs, which were the potentially predominant PFAS in the landfill air based on other studies, were not included in this Ireland study. In addition, the local meteorological and geographical conditions may also play a role in the atmospheric distribution and transport of the air-phase PFAS.

3. PFAS with modern liner systems

Modern landfills are normally constructed with a composite liner system and a leachate collection/removal system at the bottom to impede downward migration of leachate into underlying soil and aquifers. Despite the different design requirements for non-hazardous and hazardous wastes, materials used for the bottom liner system typically include synthetic flexible membranes (i.e., geomembranes), compacted clay liners, and/or geosynthetic clay liners (GCLs).

Geomembranes are widely used as hydraulic barriers due to their non-porous structure and ease of installation (USEPA, 2002). They are typically made of high density poly ethylene (HDPE), linear low-density polyethylene (LLDPE), low density linear polyethylene and flexible polypropylene (USEPA, 2002). Although geomembranes are known of their low permeability, studies have indicated some substances in leachate, such as volatile organic compounds and phenolic compounds (Touze et al., 2012), are able to diffuse through geomembranes at a faster rate compared with other common contaminants. Battista et al (2020) investigated the diffusion of PFOA and PFOS through a commercial LLDPE geomembrane. Adsorption tests suggested the partitioning coefficients of PFOA and PFOS onto LLDPE were in the range of 0.9–1.4 and 2.8–5.3, respectively. These values are at least two orders of magnitude lower than that reported for other common hydrophobic organic compounds detected in leachate (e.g., benzene and polychlorinated biphenyls). Despite the use of a relatively thin LLDPE geomembrane (0.1 mm) and elevated concentrations of PFOA and PFOS (1.1 mg/L; typical concentrations in landfill leachate are in μg/L or lower), the concentrations of these two legacy PFAS remained below the detection limits (0.1 μg/L) in the receptor cells after 202 days of testing at 23, 35 and 50 °C. Although there is no published data for HDPE yet, diffusion through HDPE was expected even lower given the greater tortuosity of the diffusive path through the amorphous zone and around the polyethylene crystals (Di Battista et al., 2020).

Compacted clay liners and/or GCLs are normally constructed/installed underneath geomembrane to further minimize the potential migration of hazardous constituents. Adsorption of PFAS onto natural soils and clay have been reported in recent years; however, the extent of adsorption was found to vary substantially depending on the soil/clay properties (e.g., organic content, anion exchange capacity, iron oxides, etc.) (Li et al., 2019), characteristics of PFAS (e.g., functional group, chain length, etc.) and water chemistry conditions (e.g., pH, salinity, DOM, etc.) (Mukhopadhyay et al., 2021). Adsorption or retention of PFAS to the clay liner needs to be evaluated on a site-specific basis.

Sometimes, due to lack of naturally occurring clay soil, on-site natural soil may be mixed with processed clay minerals such as bentonite to reach the desired hydraulic conductivity. Li et al. (2015) investigated the PFAS adsorption capacity of sodium bentonite and their potential impact on the swelling and hydraulic conductivity of bentonite under simulated leachate conditions. It was found that bentonite showed limited adsorption for 10 PFCAs and 3 PFSAs due to electrostatic repulsion between anionic PFAS and the negatively charged bentonite surface. In addition, the researchers observed that the presence of PFAS (at up to 10 ppb) showed no conspicuous effect on the swelling of bentonite and the hydraulic conductivity of sand-bentonite mixture, suggesting that the presence of PFAS would not impede the effectiveness of bentonite to restrict permeation of other contaminants in leachate.

The interactions and transport of PFAS through the landfill liner system is still a subject of active research. There is very limited data published to date. Rowe and Barakat (2021) modelled the transport of PFOS in a hypothetical single lined landfill and suggested that an intact HDPE geomembrane without any holes is likely to serve an effective diffusion barrier to PFOS. However, when holed wrinkles are present in the geomembrane, the underlying groundwater would likely be impacted by PFOS-containing leachate. The extent of contamination will depend on the level of leachate head, the location, shape, and length of holed wrinkles, the geomembrane-GCL interface transmissivity, the GCL hydraulic conductivity, and the PFAS concentration in leachate (Rowe and Barakat, 2021). As such, the quality of the installed geomembrane is extremely critical in minimizing leakage, regardless of PFAS concentrations.

4. Treatment for PFAS-containing leachate

4.1. Existing leachate treatment systems

Conventional wastewater treatment systems such as activated carbon (AC) adsorption, flocculation, biological treatment, and membrane separation have been employed at landfills to treat leachate before discharging it to the municipal wastewater treatment plants or natural waterbodies. Although they may not have been designed for PFAS treatment, some existing leachate treatment facilities may be able to reduce the PFAS concentrations in leachate to various degrees.

4.1.1. Activated carbon adsorption

AC adsorption of PFAS in landfill leachate has been evaluated in both laboratory (Johnson, 2019) and full (Busch et al., 2010; Kameoka et al., 2022) scales. In general, AC is more effective in removing long-chain PFAS than short-chain analogues and PFSAs are more prone to AC adsorption than PFCAs (Johnson, 2019; Kameoka et al., 2022). This observation is consistent with findings from other studies on PFAS removal in groundwater (Liu et al., 2019; Rodowa et al., 2020). Adsorption of PFAS on AC is primarily due to hydrophobic interaction (Park et al., 2020; Zhang et al., 2016), though electrostatic attraction may also play a role when positively charged adsorption sites are present (Cantoni et al., 2021). As such, the adsorption preference is often correlated with the hydrophobicity of PFAS or the carbon chain length for PFAS of the same homologs. Because the hydrophobicity of PFAS can be expressed by their octanol–water partition coefficients (K_{ow}) (Pauletto and Bandosz, 2022), their adsorption behavior may also be predicted based on the K_{ow} values.

With respect to the acidic functional groups, sulfonate PFAS usually have a greater K_{ow} than carboxylate PFAS with the same number of fluorinated carbons (Pauletto and Bandosz, 2022). As a result, PFSAs are often more preferentially adsorbed by AC than PFCAs.

The fundamental properties of the adsorbents, such as surface charge/functionalities, pore size/volume, and specific surface area, have been found to play important roles in PFAS removal in drinking water (Cantoni et al., 2021) and are expected to affect the PFAS removal in leachate as well. However, it should be noted that due to the stronger matrix effect in landfill leachate (i.e., the presence of high concentrations of DOM and co-contaminants) than in drinking water, the PFAS adsorption rate and capacity by AC may be inhibited due to stronger competition of co-solutes in leachate and blockage or fouling of the micropores/mesopores in AC (Yu et al., 2012).

Because AC adsorption only transfers from leachate to the adsorbent phase, namely it does not degrade PFAS. Moreover, regeneration of PFAS-laden spent AC has been a challenge in the field because it often requires costly and toxic solvent (e.g., methanol) to desorb PFAS, and the spent regenerant needs additional disposal and handling. As such, incineration of spent AC has been often practiced, which, however, is rather costly due to the high temperature (>900 °C) required to destroy PFAS (Sonmez Baghirzade et al., 2021).

4.1.2. Ion exchange

Ion exchange (IX) is another separation technology commonly used to remove cations or anions in water because of its simplicity, effectiveness, selectivity and ability to recover (Liu et al., 2021b). Various commercial anion exchange resins have been shown effective to remove anionic PFAS from drinking water and groundwater and have been successfully applied in both laboratory and full scales (Woodard et al., 2017; Zaggia et al., 2016).

In general, PFAS anions are removed by IX resins via: 1) electronic interactions between the positively charged functional groups on the resins and the negatively charged head group of PFAS, 2) hydrophobic effects that drive PFAS towards hydrophobic matrix (typically polystyrene) of the resins, 3) van der Waals interactions, and 4) hydrogen bonding (Boyer et al., 2021). Its performance could vary significantly based on the properties of the adsorbent (e.g., hydrophobicity, pore structure, and functional groups), properties of PFAS (e.g., functional heads, chain length, and molecular structure), and water chemistry (e.g., pH, DOM, and co-contaminants). Compared to AC, IX is much more effective for treating short-chain PFAS (Dixit et al., 2021). However, IX process is generally a stoichiometric process and the removal of short-chain PFAS may be inhibited if high concentrations of long-chain PFAS are co-present (Liang et al., 2022; Zeng et al., 2020). Moreover, IX resins are generally more expensive than AC and more vulnerable to organic fouling.

Compared to AC, IX is less commonly used at landfills. According to the USEPA (2000), <1 % of indirect non-hazardous landfills employed IX processes as part of their wastewater treatment systems due to its vulnerability to fouling by organics and solids. A pretreatment system with a cartridge filtration unit is normally needed to remove suspended solids in leachate (USEPA, 2000), or IX can be used as a polishing step prior to the final discharge of treated leachate.

Like AC, IX does not degrade PFAS and its regeneration is rather difficult and costly. Some resins such as Purolite® A520E (Zaggia et al., 2016) and ECT® Sorbix LC1 IX (Woodard and Sinnett, 2019) are considered non-regenerable and need to be shipped offsite for incineration after single use, while those considered regenerable require a combination of salt, base and organic solvents for regeneration (Boyer et al., 2021), and the treatment and disposal of the spent regenerant can be quite costly (Liu et al., 2021b).

4.1.3. Biological treatment

Biological treatment is one of the most used technologies for wastewater treatment at landfills (USEPA, 2000). Yet, the extremely strong C-F bond makes PFAS inherently persistent to biological treatment (Zhang et al., 2022). In fact, a few studies (Fuertes et al., 2017; Gewurtz et al., 2013; Solo-Gabriele et al., 2020) have shown that aerobic biological treatment processes resulted in elevated total PFAS concentrations in leachate after aeration. It indicates limited effectiveness of biological degradation of PFAS and possible biotransformation of precursor compounds (such as FTOHs, FTSs, or PAPs) in leachate to PFAAs (Hamid et al., 2020c; Liu et al., 2021a). Yan et al. (2015) reported a decrease in the concentrations of PFPrA, PFHxA, PFHpA, PFBS and PFOS after MBR treatment. It was ascribed to the loss of PFAS due to water-to-air transfer by bursting bubbles in the aeration tanks and the sorption of PFAS on the activated sludge as the concentrations of these PFAS in the sludge phase increased (Yan et al., 2015).

4.1.4. Membrane separation

Ultrafiltration and reverse osmosis (RO) have been used to treat landfill leachate for years (USEPA, 2000). Ultrafiltration is normally used to intercept emulsified oils and suspended solids (USEPA, 2000); however, it was ineffective to remove dissolved substances like PFAS (Crone et al., 2019).

RO has been demonstrated effective in PFAS removal. Busch et al. (2010) reviewed the concentration/composition of PFAS in untreated and treated leachate from 22 landfills in Germany and found the effluent samples from RO showed the lowest total PFAS concentrations (15.2–129 ng/L), followed by AC adsorption (9.26 – 4079 ng/L) and nanofiltration (621 – 1257 ng/L). A removal rate of approximately 90 % – 100 % for PFAAs was reported in the RO systems at five MSW landfills in China (Yan et al., 2015) and a leachate treatment facility in Japan (Kameoka et al., 2022).

Despite its high effectiveness on PFAS removal, RO is a physical separation process that does not destroy PFAS. PFAS are concentrated in the retentate stream (a.k.a., concentrate), which is typically 20 – 25 % by volume of the feed leachate (Peters, 1998). The RO concentrate usually contains higher concentrations of inorganic salts and organic compounds in addition to PFAS and needs to be properly disposed of or further treated.

Recirculation of RO concentrate back to the waste area has been considered as an economical option for landfill owners and operators; however, the consequence of this practice on leachate quality and gas production should be carefully evaluated. In a study at a MSW landfill in Poland, Talalaj and Biedka (2015) found that recirculation of RO concentrate to the waste cells could accelerate waste decomposition by increasing the waste moisture and decreasing pH to 7.2, which was conducive for methanogenesis; however, it also led to an increase of TSS in leachate, which could adversely impact the RO operation due to intensification of the membrane fouling. In addition, elevated ammonia and chloride concentrations was reported in leachate after recirculation of RO concentrate in both laboratory (Morello et al., 2016) and field (Calabrò et al., 2018) studies, coupled with doubled free ammonia gas emission (Morello et al., 2016). High concentrations of ammonium could inhibit methanogenesis and reduce the production of methane gas. An inhibition of biological waste decomposition due to high levels of ammonium (up to 15,000 mg NH_4-N/L) after recirculation of RO concentrate and leachate was reported at a MSW landfill in Australia

(Chamem et al., 2020). Recirculation of RO may also lead to an increase of the leachate generation rate as evidenced by 10-year data collected from an Italian landfill (Calabrò et al., 2018), whereas another laboratory study indicated leachate production was not influenced by RO concentrate injection (Morello et al., 2016).

Other alternatives for RO concentrate disposal/treatment include deep well injection, evaporation ponds (Kanchanapiya and Tantisattayakul, 2022), solidification (Hunce et al., 2012) and advanced oxidation processes (Zhang et al., 2019). They are either costly or remain at the laboratory scale.

In summary, RO is one of the most promising technologies that have been applied for landfill leachate treatment in full scale. Its effectiveness on PFAS removal has been demonstrated in both laboratory and field scale. But it is inherently energy-intensive (Lin and Elimelech, 2015). Moreover, the presence of elevated concentrations of suspended solids, salts and metals in leachate could adversely affect the RO operation and cause more frequent and severe membrane fouling/scaling (Tałałaj et al., 2019). The treatment/disposal of RO concentrate enriched with PFAS and other co-contaminants in leachate needs to be carefully addressed.

4.2. Emerging treatment technologies for PFAS in leachate

In recent years, a number of non-destructive and destructive emerging treatment technologies have been developed and applied for treating PFAS in water. However, given the unique challenges associated with landfill leachate (i.e., complex matrix components, highly variable strengths, and lack of solid regulatory drivers), only few of them have been applied for treating PFAS in leachate.

4.2.1. Foam fractionation

Foam fractionation is one of the emerging technologies that have been tested for leachate treatment. It introduces gas bubbles into the liquid to concentrate surface-active PFAS molecules at gas–liquid interface of the bubbles. The resulting PFAS-concentrated foam at the air–liquid interface is then separated for further treatment or disposal.

Robey et al. (2020) investigated the effectiveness to concentrate PFAS in foam produced via the bubble aeration of landfill leachate. A mean removal rate of 69 % was reported for 21 mass-labeled and 37 native PFAS compounds with sequestration peaked at PFHpA and PFOS. PFAS removal efficiency was reported declined for all longer-chain compounds and the shortest-chain species including PFBA, PFPrS, PFPeA and PFBS. McCleaf et al. (2021) investigated the performance of foam fractionation for PFAS removal from leachate in both batch and continuous modes. >90 % of PFOA, PFOS, PFHxS, and PFHpA were removed for both modes, while a lower removal of 0–50 % was observed for PFBA, PFPeA, PFDA, FOSA, EtFOSAA and MeFOSAA. The PFAS removal efficiencies were reported to be correlated with the magnitude of the original PFAS concentrations, instead of the chain length or air–water partitioning coefficient, although it appeared not consistent with the study by Robey et al. (2020). Similarly, a low removal rate of 10 % was reported for short-chain PFAS by Krogerstrom (2021) in another laboratory study with landfill leachate, where an average removal of 67 % was reported for long-chain PFAS.

In field scale, OPEC Systems conducted a full-scale field trial at Telge Landfill in Sweden and reported their treatment system removed PFOA, PFOS, PFHxS by>98 %, while the removal efficiency for short-chain PFAS such as PFBA, PFBS, and PFPeA varied between 4.4 % and 26.7 % (OPEC-Systems, 2021).

While the current foam fractionation practices mainly use air bubbles, ozonated air has been tested to improve the PFAS removal efficiency in water. Dai et al. (2019) reported that the total PFAS removal increased from 81 % to 95 % by switching from air to ozonated air in a laboratory foam fractionation study. The improved removal rate was attributed to the formation of hydroxyl radicals with ozone, which could serve as a strong electron binder around the interface of the gas bubbles to increase the partition of PFAS to the gas bubbles (Dai et al., 2019).

Like RO, foam fractionation is a physical separation process. It will generate a PFAS-concentrated foam at a volume of approximately 20–25 % of the feed leachate (McCleaf et al., 2021; Robey et al., 2020) that needs to be further treated or disposed of. Strategies for managing these PFAS-concentrated foam need to be further explored.

4.2.2. Electrochemical oxidation

Electrochemical oxidation has been successfully tested to reduce DOM and ammonia nitrogen in landfill leachate before (Cabeza et al., 2007). Recent studies (Radjenovic et al., 2020; Sharma et al., 2022) have shown that electrochemical oxidation could combine oxidation by hydroxyl radicals generated via anodic oxidation of water and direct electron transfer onto the anode to mineralize PFOA, PFOS and other PFAS in water.

Pierpaoli et al. (2021) investigated the effectiveness of electrochemical oxidation to degrade PFOA and PFOS in a MSW landfill leachate using a bench-scale reactor equipped with boron-doped diamond (BDD) electrodes. An average removal efficiency of 80 % and 78 % were reported for PFOA and PFOS at a current density of 75 mA/cm^2 after eight hours. A higher removal rate of 100 % and > 97 % were reported for PFOS and PFOA in another study with landfill leachate after 2 h of recirculation at a current density of 150 mA/cm^2 (Maldonado et al., 2021). Clearly, increasing the current density could enhance the degradation of PFAS. However, degradation of short-chain PFAAs, particularly PFBA, PFBS, and PFPeA, were found more challenging. An increase of PFBA, PFBS, and PFPeA concentrations were observed due to the degradation of longer-chain analogues, preferential conversion of PFSAs to PFCAs, and possible transformation of precursors present in leachate (such as 6:2 FTSA) (Maldonado et al., 2021).

Formation of toxic halogenated byproducts, such as perchlorate, have been reported in the electrochemical oxidation treatment with landfill leachate. Due to the associated health effects, these byproducts are subject to stringent regulations. For example, perchlorate is regulated in multiple states in the United States (USEPA, 2014). In a study by Maldonado et al. (2021), up to 12 mM of perchlorate was observed due to the oxidation of high concentrations of chloride in leachate. Although low current density and/or shorter treatment time could reduce the generation of perchlorate, it could also compromise the PFAS removal. Additional treatment measures may need to be evaluated if removal of these toxic halogenated byproducts is warranted.

The current electrochemical oxidation studies for leachate treatment are largely limited to laboratory bench-scale systems. In addition to the formation of halogenated byproducts, high energy consumption is another major obstacle for full-scale implementation of electrochemical oxidation (Mandal et al., 2017). The BDD electrodes are rather expensive to upscale and the high cost of the substrate material (Niobium, Tungsten, Tantalum) for BDD film deposition further limits the large-scale manufacturing of BDD electrodes (Radjenovic et al., 2020). Although not reported in the current studies, potential electrode fouling by adsorption of DOM and precipitation of inorganics have been documented in the studies with real waste streams (Radjenovic et al., 2020) and must be considered in field operation.

4.2.3. Photocatalytic oxidation

Heterogeneous photocatalytic oxidation with semiconductor-based catalysts has shown effective on degradation of PFAS in water under ultraviolet (UV) irradiation (Li et al., 2020a; Xu et al., 2017). Although commercial titanium dioxide photocatalyst has been found ineffective for PFAS degradation (Chowdhury et al., 2021), new classes of photocatalysts have been explored in recent years to enhance PFAS degradation by introducing different base catalysts, imparting a metal dopant, manipulating PFAS adsorption modes and molecular orientation, increasing light absorbability, and inhibiting electron/hole recombination (Leonello et al., 2021; Li et al., 2020b).

The application of photocatalytic oxidation to remove and destruct

PFAS in landfill leachate has been very limited. Tian et al. (2021) tested a two-step 'Concentrate-&-Destroy' technology (Fig. 1) for treating 18 PFAS from a MSW landfill leachate through bench- and pilot-scale experiments. The technology employed an innovative adsorptive photocatalyst (Fe-doped carbon-supported titanate nanotubes, Fe/TNTs@AC). To mitigate the leachate matrix effect on the photocatalytic process, PFAS are first adsorbed on Fe/TNTs@AC that is designed to bind the anionic PFAS through both hydrophobic interactions with the tails and electrostatic interactions/surface complexation with the head groups (Li et al., 2020b). Subsequently, the PFAS-loaded solids are separated from the leachate and then subjected to UV irradiation for PFAS destruction, which will also regenerate the material for repeated uses. Despite the strong matrix of the leachate (e.g., TOC = 418.6 ~ 440.1 mg/L), Fe/TNTs@AC was able to selectively adsorb most of the PFAS from the leachate, with a binary PFOA-TOC separation factor of 110 for PFOA, which effectively concentrated PFAS on the photoactive sites. At a dosage of 10 g/L, fresh Fe/TNTs@ AC removed > 95 % of 13 PFAS from the leachate within 2 h, 86 % after first regeneration, and 74 % when reused three times. In a follow-up pilot study, Fe/TNTs@AC removed > 92 % of 18 PFAS in 8 h under the field conditions, and when the PFAS-laden solids were subjected to the UV-H_2O_2 system, ~84 % of 16 PFAS in the solid phase were degraded. However, the authors noticed Fe/TNTs@AC was less effective for PFBA and PFPeA likely due to transformation of precursors and/or longer-chain homologues into these short-chain PFAS and competition of adsorption sites by the longer-chain PFAS (Tian et al., 2021).

Most studies on photocatalytic oxidation of PFAS have been conducted in laboratory under a well-mixed condition and primarily focused on material development and treatment mechanisms. Only a limited number of bench-scale studies have been conducted to develop innovative treatment system/reactor designs (Thomson et al., 2020). The reactor system design is critical for industrial applications particularly for heterogeneous photocatalysts because its treatment efficiency could be limited by obscured catalyst surface, reduced illuminated catalyst surface area, and higher rates of photon scattering (Loeb et al., 2019). To ensure the long-term viability of photocatalytic oxidation for full-scale applications, the material stability and reusability, ease of operation and cost effectiveness should also be considered.

4.2.4. Plasma-based treatment

Plasma-based treatment is another advanced oxidation method that has been demonstrated effective for PFAS destruction in water (Lewis et al., 2020). It relies on a combination of highly reactive oxidative (e.g., •OH, O, HO_2•, O_2•–, O_3, H_2O_2) and reductive species (e.g., free and aqueous electrons e_{aq-}, hydrogen radicals, etc.), which are generated during the formation of an electrical discharge between a high voltage and a grounded electrode in the vicinity of water (Singh et al., 2019).

Singh et al. (2021) tested a bench-scale gas-discharge reactor to degrade PFAS in leachates collected from three MSW landfills. PFAS in the leachate was concentrated by argon bubbling to the gas–liquid interface and then degraded by reactive species generated by plasma in the argon air (Singh et al., 2021). 90 % of PFOA and PFOS, >99.9 % of long-chain PFAAs, and 10–99.9 % of short-chain PFAAs were removed after 10 min of treatment. The PFAS degradability was reported decreasing with decreased PFAS chain length due to 1) the lower surface activity of shorter-chain PFAS, which controlled PFAS concentrations at the gas–liquid interface, and 2) formation of shorter-chain PFAS due to stepwise breakdown of longer-chain analogues and precursors (Singh et al., 2021). PFSAs were reported being degraded more rapidly than PFCAs of the same carbon number due to higher surface activity with an extra –CF_2 group in a PFSA molecule (Singh et al., 2021).

High concentrations of organic matter in leachate was reported not to compromise the efficacy of plasma treatment due to the pre-concentration of PFAS at the plasma (gas)-liquid interface; however, high concentrations of ionic species could make the leachate highly electrically conductive and thereby decrease the PFAS removal rate through decreasing the liquid–gas contact area (Singh et al., 2021). Generation of toxic halogenated byproducts like chlorite and chlorate due to oxidation of high concentrations of chloride in leachate was also reported (Singh et al., 2021).

The current leachate treatment studies with plasma-based technologies remain in the laboratory-scale configurations. Scaling up the plasma reactors is still a challenge. For example, one major issue to scale up the atmospheric pressure plasma technology is that the discharges tend to be filamentary in form and very localized, which makes it difficult to work with large flow rate (Foster et al., 2018). Moreover, the costs, long-term system reliability and operational safety need to be evaluated.

5. Conclusions and perspectives

With more PFAS-containing wastes being landfilled and the fast-evolving regulations, there is an urgent need to understand the long-term effectiveness/protectiveness of landfills for PFAS disposal. This article provides a state-of-the-science review on the occurrence and transformation of PFAS at landfills in both leachate and air, the potential

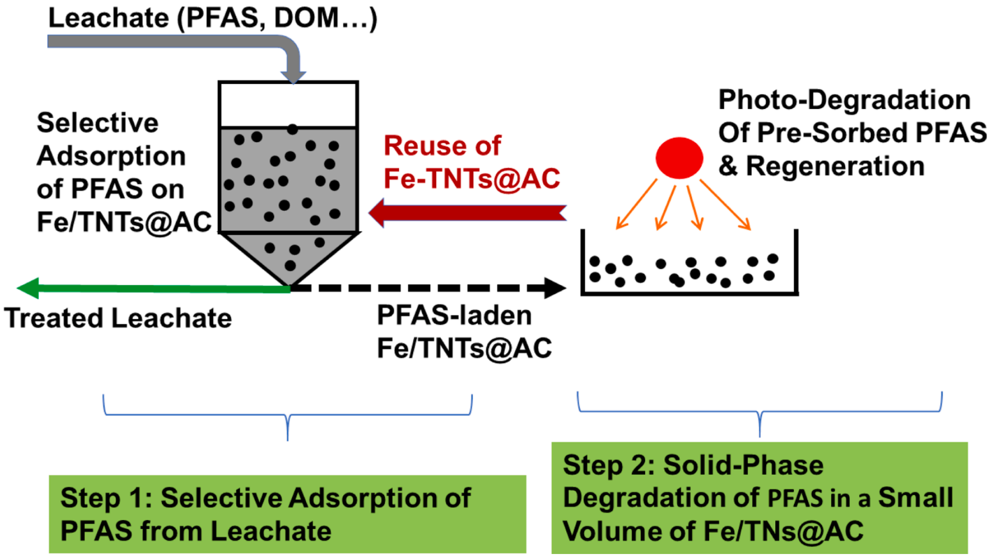

Fig. 1. A two-step concentrate-&-destroy process for selective adsorption and solid-phase degradation of PFAS from landfill leachate.

interactions of PFAS with landfill liners and the efficacy of wastewater treatment technologies for proper management of PFAS-containing leachate from landfills.

The dominance of C4-C7 PFCAs in landfill leachate have been documented in recent studies. The abundance of 5:3 FTCA has also been reported in untreated landfill leachate in a few studies at United States. However, 5:3 FTCA is conducive to aerobic degradation, so its concentration could decrease significantly after aeration treatment. There is no significant correlation reported between PFAS concentrations in leachate and the type of waste in the landfills yet. It is likely because of the widespread presence of PFAS across various products deposited at landfills and only small fractions of PFAS (out of $>$ 4000 used globally) being targeted for analysis.

Elevated concentrations of PFAS have been reported in air at landfills compared to upwind direction. The predominant PFAS in landfill air are different from that reported for leachate. Neutral PFAS, primarily FOTHs, have been reported dominating likely due to their lower solubilities and higher vapor pressure. Anionic PFAS has also been detected in the air with PFBA being dominant.

There have been very limited data available on the interactions between PFAS and the landfill liner systems. The presence of PFAS in leachate is not anticipated to significantly alter the hydraulic conductivity of GCL or compacted soil liner. Existing data suggested undamaged and properly installed geomembrane is expected to serve an effective barrier for PFOS; however, data on the diffusivity of other PFAS than PFOS across different types of geomembrane materials and their impact on the service life of geomembranes are still lacking.

To manage leachate discharge from landfills, the most common approach is off-site treatment at publicly owned treatment works. However, most of the existing wastewater treatment systems are not designed to target PFAS. Among conventional treatment technologies, RO has been proven highly effective to remove most PFAS from leachate; however, aside from the strong matrix effect on the membrane fouling, the treatment/disposal of PFAS-concentrated RO concentrate needs to be carefully evaluated. Recirculation of the RO concentrate back to landfills may change the leachate quality/quantity and affect the waste decomposition. AC adsorption is more effective to remove long-chain PFAS and PFSAs compared to PFCAs. Ion exchange can be employed as a polishing step in the treatment train for removal of short-chain PFAS; however, their effectiveness and reactive lifetime need to be verified under the leachate conditions. Emerging treatment technologies such as the 'concentrate-and-destroy' technology using adsorptive photocatalysts, the plasma-based treatment, foam fractionation, and other advanced oxidation processes have been explored for PFAS removal and destruction. Among those emerging treatment technologies, foam fractionation is expected to be less energy-intensive and requires low capital investment. The operation cost to remove PFAS from a Swedish landfill was reported to be \$0.12 USD/m^3 leachate treated (OPEC-Systems, 2021). However, foam fractionation could result in a PFAS-concentrated foam at a volume of approximately 20–25 % of the feed leachate that needs further treatment/proper disposal. The handling and treatment/disposal cost for the PFAS-concentrated foam should be considered in the cost analysis. Except for foam fractionation, most of the other emerging technologies are still limited in the laboratory or pilot scale for PFAS removal in leachate. Very limited data is available in terms of their potential capital and operation costs to treat PFAS in landfill leachate. Singh et al. (2021) reported that the electrical energy per order of an enhanced contact plasma reactor to treat PFOA and PFOS in three MSW landfill leachate samples ranged from 20 to 36 kwh/m^3, approximately equal to \$1.4 – 2.6 USD/m^3 based on an averaged industrial consumer electricity cost of \$0.0726 USD/kwh in United States in 2021. It should be noted that the mean concentrations of PFOA and PFOS in Singh's leachate samples were 2400 ± 300 and 270 ± 90 ng/L, respectively. However, the reported average $\sum$PFAS was 65,000 ± 33,000 ng/L and reportedly dominated by short-chain PFAS. Thus, the actual cost could be much higher, and moreover, the capital cost and other operation and maintenance costs remain unknown.

The rising concern about the health effects of PFAS has triggered growing attention among the scientific and regulatory communities. In June 2022, the U.S. Environmental Protection Agency (U.S. EPA) further lowered their interim drinking water health advisory values of PFOA and PFOS to 0.004 and 0.02 ng/L, even below the PFAS's minimum reporting level of 4 ng/L. According to the 2021 PFAS Strategic Roadmap, USEPA is now collecting data from landfills to inform decision making about a future rulemaking to restrict PFAS discharges from landfills.

To better understand the various sources of PFAS at landfills, data related to the content and speciation of PFAS in different kinds of solid wastes are lacking. As such, there is a need to develop rapid and cost-effective analytical approaches to identify and determine various classes of PFAS in landfills. The advancement of analytical methods and development of standard solutions for more PFAS (liner and branch) and their precursors will help gain a better understanding of the long-term fate and transport of PFAS at landfills.

Although studies have investigated PFAS in the air at landfills, data is still lacking to characterize PFAS in landfill gas, the gaseous byproduct generated along with waste decomposition. Data regarding the efficacy of the current landfill gas collection and control systems on the emission of gaseous PFAS, and PFAS behaviors through the flare/internal combustion engines will be critical to ensure proper management of potential PFAS gaseous releases from landfills.

Among these emerging technologies for leachate treatment, foam fractionation is the only one that has been deployed in full scale for leachate treatment. The 'concentrate-and-destroy' concept appears most promising because pre-concentrating PFAS on either the photoactive materials or to the gas–water interface could avoid the strong matrix effect on the subsequent degradation (photo or plasma) processes. To this end, more PFAS-selective and more reactive adsorptive photocatalysts or air-floating approaches need to be further explored or improved.

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

The authors do not have permission to share data.

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