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

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ABSTRACT

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

1. Introduction

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

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

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Abbreviations

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

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

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

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

2. Methodology

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

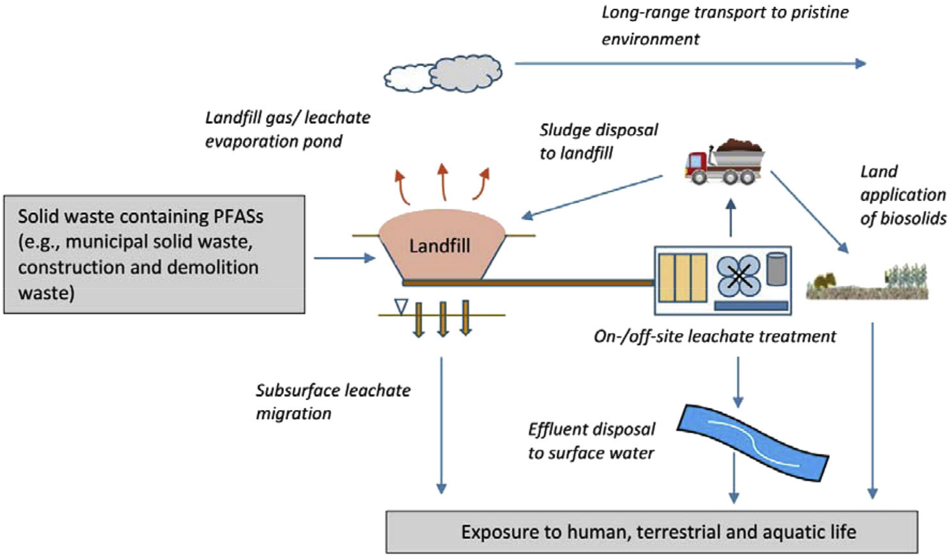

Fig. 1. Environmental pathways of per- and polyfluoroalkyl substances (PFASs) originating from solid wastes.

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

3.1. Occurrence and sources of PFASs in leachate

3.1.1. Perfluoroalkyl acids (PFAAs)

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

3.1.2. Fluorotelomer-based substances

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

3.1.3. Perfluoroalkyl sulfonamide derivatives

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

3.1.4. Polyfluoroalkyl phosphate esters (PAPs)

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

3.2. General observations and geographical trends of PFASs in leachate

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

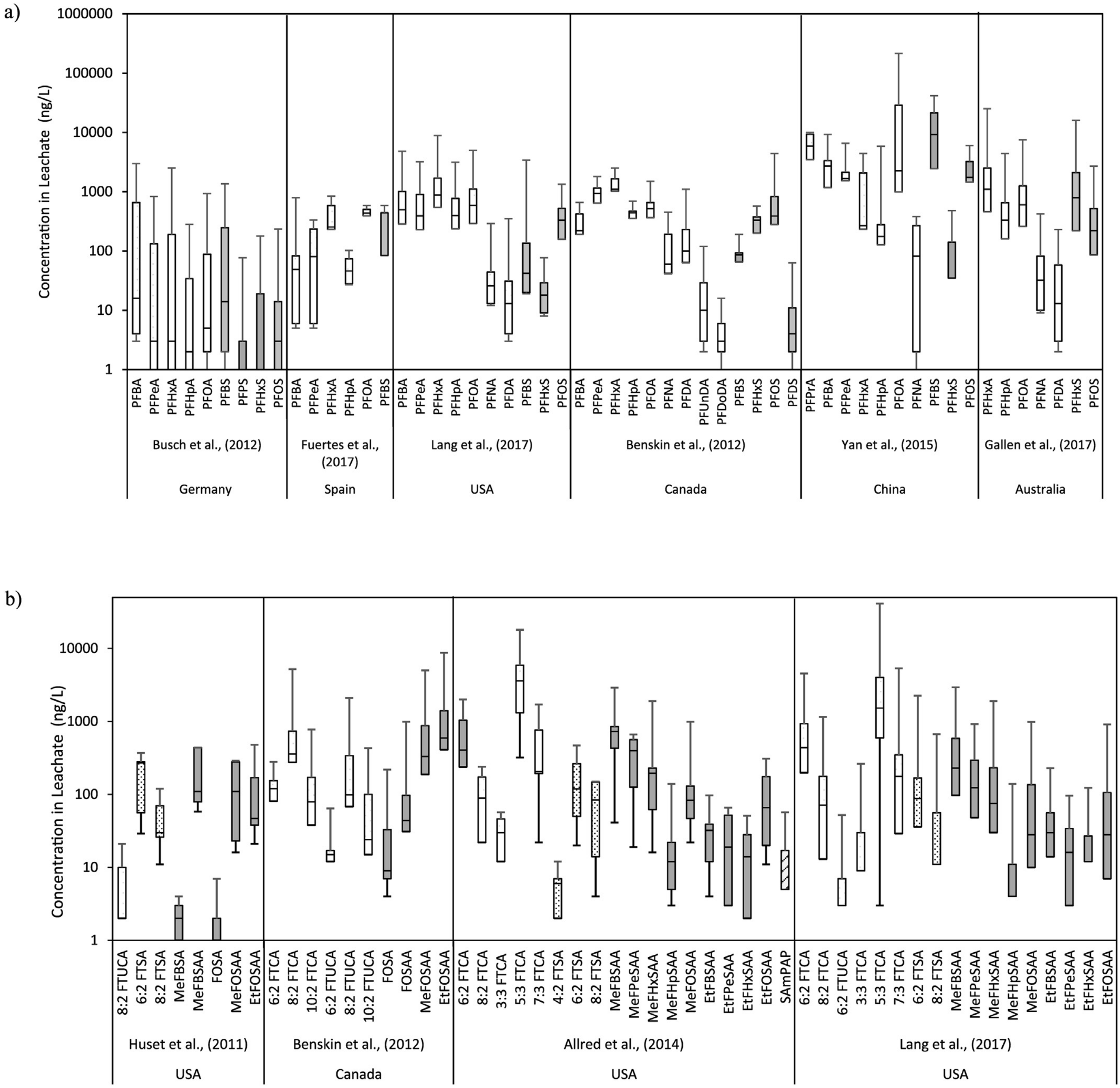

Fig. 2. Concentration ranges of (a) perfluoroalkyl carboxylic acids (PFCAs; plotted with white box) and perfluoroalkyl sulfonic acids (PFSAs; plotted with grey box); For countries with multiple studies, the study with highest number of landfill leachate samples are plotted in 2(a). Concentration ranges of (b) fluorotelomer carboxylic acids (plotted with white box), fluorotelomer sulfonic acids (black dotted box) and perfluoroalkane sulfonamide derivative substances (plotted with grey box) and polyfluorinated phosphate ester (plotted with slanted line) in landfill leachate in different countries. Analytes with ≥50% detection frequency has been plotted in 2(b). The box plots represent the 1st quartile, median and 3rd quartile values. The negative and positive error bars represent the difference between 1st quartile and minimum value, and maximum and 3rd quartile value, respectively. Note the logarithmic scale of Y-axis.

(Acronyms for 2(b): FTCA: fluorotelomer carboxylic acid, FTUCA: fluorotelomer unsaturated carboxylic acid, FTSA: fluorotelomer sulfonates, MeFASAA: methyl-perfluoroalkane sulfonamide acetic acid (C4-C8), EtFASAA: Ethyl-perfluoroalkane sulfonamide acetic acid (C4, C5, C6, C8), SAmPAP: 2 (*N*-ethylperfluorooctane-sulfonamido) ethyl phosphate).

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

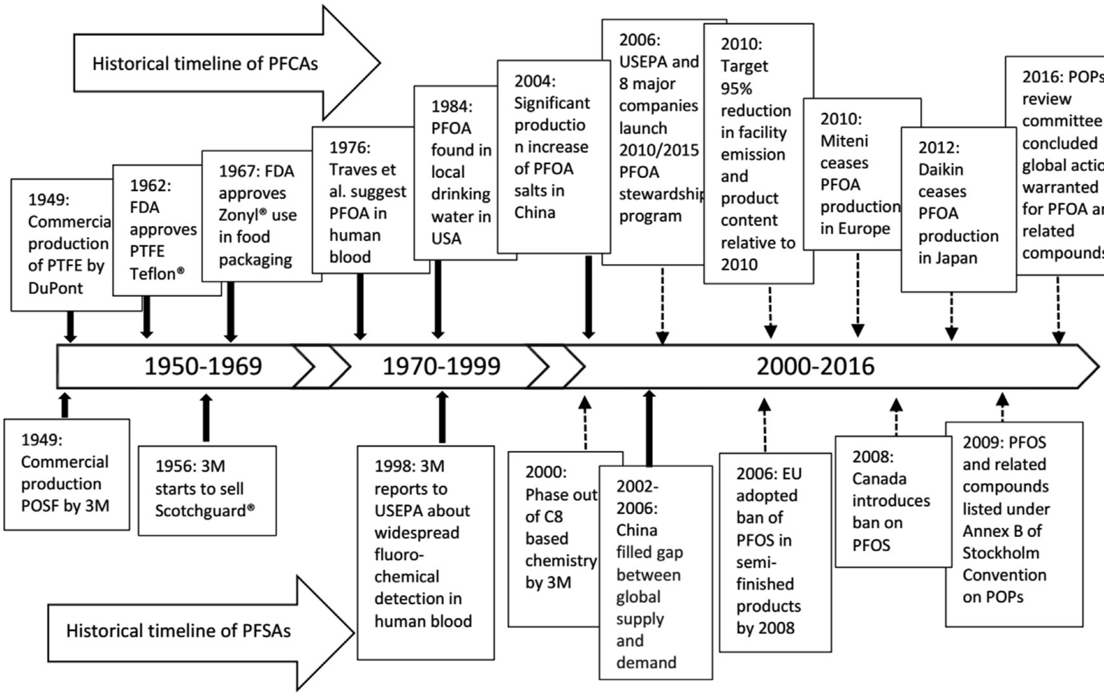

Fig. 3. Timeline of the production, commercialization and legislation of perfluoroalkyl carboxylic acids (PFCAs; at the top) and perfluoroalkyl sulfonic acids (PFSAs; at the bottom). Events and actions that may have resulted in increased concentrations in the environment and important findings are indicated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

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

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

3.2.1. Concentration and trends in the USA and Canada

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

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

3.2.2. Concentration and trends in European Countries

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

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

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

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

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

3.2.3. Concentration and trends in Australia

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

3.2.4. Concentrations and trends in China

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

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

3.3. Estimating PFASs mass discharged with landfill leachate

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

3.4. Occurrence of PFASs in landfill ambient air

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

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

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

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

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

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

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

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

3.5.1. *Effect of leachate physiochemical properties*

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

3.5.2. *Biological processes inside landfills*

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

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

3.5.3. *Effect of climate*

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

3.5.4. *Effect of waste age and waste composition*

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

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

4. Fate of PFASs in leachate treatment systems

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

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

5. Future research directions

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

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

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

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

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

6. Conclusions

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

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

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2017.12.030.

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