Environmental Pollution 235 (2018) 74–84

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

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

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

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

ARTICLE INFO

Article history:
Received 21 March 2017
Received in revised form
2 August 2017
Accepted 8 December 2017

Keywords:
Landfill
Polyfluoroalkyl
Perfluoroalkyl
Leachate
Degradation

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,

☆ This paper has been recommended for acceptance by Dr. Chen Da.

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

https://doi.org/10.1016/j.envpol.2017.12.030
0269-7491/

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.