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Cite this: *Environ. Sci.: Processes Impacts*, 2019, **21**, 1970

Leachate emissions of short- and long-chain per- and polyfluoralkyl substances (PFASs) from various Norwegian landfills†

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Received 10th April 2019
Accepted 7th August 2019

DOI: 10.1039/c9em00170k

rsc.li/espi

Restrictions on the use of long-chain per- and polyfluoralkyl substances (PFASs) has led to substitutions with short-chain PFASs. This study investigated the presence of four short-chain PFASs and twenty-four long-chain PFASs in leachate and sediment from ten Norwegian landfills, including one site in Svalbard, to assess whether short-chain PFASs are more dominant in leachate. PFASs were detected in all sites. Short-chain PFASs were major contributors to the total PFAS leachate concentrations in six of ten landfills, though not in Svalbard. In sediment, long-chain PFASs such as perfluorooctanesulfonate (PFOS) and PFOS-precursors were dominant. Short-chain PFAS leachate concentrations ranged from 68 to 6800 ng L^{-1} (mean: 980 ± 1800; median: 360 ng L^{-1}), whereas long-chain concentrations ranged from 140 to 2900 ng L^{-1} (mean: 530 ± 730; median: 290 ng L^{-1}). Sediment concentrations, which contained mainly long-chain PFASs, ranged from 8.5 to 120 μg kg^{-1} (mean: 47 ± 36; median: 41 μg kg^{-1}). National $\sum_{28}$PFAS release from Norwegian landfills to the environment was estimated to be 17 ± 29 kg per year (median: 6.3 kg per year), which is in the same range as national emissions from the US, China and Germany after normalizing the data to a per capita emission factor (3.2 ± 5.5 mg per person per year). Results from this study are compared with previous and current studies in other countries, indicating a general trend that short-chain PFASs are dominating over long-chain PFASs in landfill leachate emissions.

Environmental significance

Compared to previous studies, where long-chain PFASs dominated leachate emissions, short-chain PFASs appear to now be more dominant in Norwegian landfills, reflecting the general market shift from long- to short-chain PFASs. National $\sum_{28}$PFASs release from Norwegian landfills was estimated to be 17 ± 29 kg per year, which is relatively lower than other countries such as China and Germany. However, the results in per capita emission factors (3.2 ± 5.5 mg $\sum_{28}$PFAS per person per year) were very similar of those derived in other regions. Landfill emissions in general appear to be a minor but not insignificant source of PFASs in the environment.

1. Introduction

Per- and polyfluoroalkyl substances (PFASs) in the environment are of concern because of their general persistence in combination with either potential aquatic mobility, long range transport, bioaccumulation, toxicity, or some combination thereof.[1] PFASs have been used in a variety of industrial processes and in commercial products over the past 60 years,[2,3] due to their unique chemical and physical properties, such as their thermal and chemical stability as well as their hydrophobic/lipophobic behavior.[2,4–6]

Regulations of PFAS in recent years[3,7,8] have led to restrictions of perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA) and other so-called "long-chain" PFASs.[9,10] This has resulted in these being replaced on the market with alternatives including so-called "short-chain" PFASs, such as perfluorobutanesulfonate (PFBS), which are considered to be less bioaccumulative.[9–11] Short-chain PFASs are defined by Buck *et al.* (2011)[10] and the Organisation for Economic Co-operation and Development (OECD) as perfluoroalkyl carboxylic acids (PFCAs) with a chain length of $<C_7$, and $<C_6$ for perfluoroalkane sulfonates (PFSAs).[9,10] Though short-chain PFASs are less bioaccumulative, they are of concern

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/c9em00170k

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because of their environmental persistence and aquatic mobility,[1] and because currently little is known about their (eco)toxicity.[12] As such, it is important to investigate to what extent these short-chain PFASs are being emitted to and occurring in the environment.

Landfill leachate from municipal solid waste are potential emission hotspots for PFASs,[13–16] though concentrations vary widely.[13–15,17,18] There are few available data on PFAS occurrence in Norwegian landfill leachates;[19,20] though the first available data, comprising data from ten landfills from 2003–2007 based on non-target screening, indicated short-chain PFASs were infrequently detected (or analysed for (ref. 21)), and long-chain PFASs clearly dominated.[20] Studies from North American landfills from 2006 to 2009 reported that specific short-chain PFASs were amongst the most dominant in landfills,[13,15] and this has also been reported in more recent studies from elsewhere such as China,[22] Germany,[17] Sweden,[23] and Spain.[24] It is therefore anticipated based on this shift of usage and data from other countries, that short-chain PFASs are emitted to a greater extent than long-chain PFASs from Norwegian landfill leachate.

The aims of this study are three-fold. The first is to characterize the aqueous (leachate) concentration and composition of PFASs being emitted from a diverse array of Norwegian landfills, alongside PFAS composition in leachate sediment. The second aim is, based on this data, to test the hypothesis that short-chain PFASs dominate landfill emissions over long-chain PFASs. The third aim is to compare these results with other regions, through the derived national per capita emission factors of short and long-chain PFASs. For this study, leachate and sediment was collected from ten diverse Norwegian landfills, including one in Svalbard, and analysed for PFASs. The collected data is further discussed in terms of the distribution of PFASs between collected sediment and water leachate, the influence of leachate characteristics, including pH, electrical conductivity (EC), leachate flow rates, dissolved organic carbon (DOC) and meteorological data (24 h and 2 week precipitation).

2. Methods

2.1. Site descriptions

Ten diverse Norwegian landfills, including one site in Svalbard (Table S1, (ESI†)), receiving primarily municipal solid waste (MSW) and in some cases industrial waste and contaminated soil and sewage sludge, were included. Yearly leachate volumes for each landfill are given in Table S2.† Due to confidentiality reasons, the identities of the landfills are anonymized. Some of the landfills were established in a period where there was few or no requirements for liners, leachate drainage nor control of the influence of storm water and groundwater dilution of the raw leachate. However, it is our opinion that the selected landfills represent typical Norwegian, and that unpolluted storm water and groundwater have relatively limited influence on the sampled raw leachate. A description of landfills, the sampling points, their hydrology and whether dilution by storm water/groundwater can be a consideration is presented in the ESI.†

2.2. Sampling

As an aim of this study was to characterize PFAS landfill emissions and sediment concentrations from all of Norway, available resources were used to sample from many locations rather than obtaining replicates or time series in individual locations. Sampling at each landfill was conducted mainly between April to June 2018 by landfill operators following sampling protocols and equipment provided by the authors. Sampling dates are given in Table S2.† Sampling of leachate (0.5 L, HDPE bottles, Eurofins Environment, Norway) was conducted as close as possible to where leachate leaves the landfill (either a retention pond, borehole, pumping station, stream, or underground culvert/leachate pipe accessed by a manhole; see Table S2, ESI†), in order to be representative of landfill emissions. Not all samples are considered raw leachate, but raw leachate diluted by storm water and groundwater (see the ESI†).

Sediment samples (500 g, Rilsan bags, Eurofins Environment, Norway) were collected from sedimentation ponds, if present, or by sandtraps in the underground culverts. The samples were placed in coolers with cooling elements and bubble wrap and shipped generally overnight and stored cold (*ca.* 4 °C) until analysis. To compliment this data, existing, recent PFAS data was also included when possible, provided by the site owners. These were obtained for four of the landfills from the same sampling points using similar protocols and analysis laboratories as in this study.

2.3. Analyses

The pH and EC were measured following method NS-EN ISO 10523:2012 and NS-ISO 7888:1985 = EN 27888:1993, respectively (Table S2†).

28 PFASs were quantified in leachate: short-chain PFCAs (PFBA, PFPeA and PFHxA), a short-chain PFSA (PFBS), long-chain PFCAs (PFHpA, HPFHpA, PFOA, PFNA, PFDA, PF-3,7-DMOA, PFUnDA, PFDoA, PFTrA, PFTA and PFHxDA), long-chain PFSAs (PFHxS, PFHpS, PFOS and PFDS), as well as fluorotelomer sulfonates (FTSAs: 4 : 2 FTSA, 6 : 2 FTSA and 8 : 2 FTSA), a fluorotelomer alcohol (FTOH: 8 : 2 FTOH) and PFOS-precursors (perfluorosulfonamides (FOSAs): FOSA, EtFOSA, MeFOSA and perfluoroalkylsulfonamide alcohols (FOSEs): EtFOSE and MeFOSE). In Table S3† the full names, PFAS class, and molecular formula for these are provided. In sediment, perfluorooctane sulfonamido acetic acids (FOSAAs) were also analysed (EtFOSAA, MeFOSAA and FOSAA). All sediment analyses and most leachate analyses (all except for leachate from landfill V and VIII), including the previous data provided by site owners, were carried out at the accredited laboratory Eurofins Environment Testing AS (sediment: centrifugation in methanol, followed ENVI-carb clean-up and UPLC/MS/MS analysis; leachate: analyzed by SPE-methanol elutriation followed by UPLC/MS/MS analysis, instrument: Agilent 6495 MS/MS. Column: Waters BEH 50 × 2.1 mm. Mobile phase: ammonium acetate (aq) and methanol). Due to logistical reasons, including contractual obligations, leachate from Landfill V and VIII were analyzed by the accredited ALS Laboratory Group Norway AS (method EPA537: SPE-methanol elutriation followed

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by LC/MS/MS analysis), which did not analyse PFBS, PFHpS, PFDS, PFBA, PFPeA, PFHpA, PFHxDA, 4 : 2 FTSA, HPFHpA and PF-3,7-DMOA.

2.4. Statistical analysis

Statistical analysis was performed with Statistica v. 13.1 (©1984–2016 by Statsoft, Tulsa, USA). All concentrations were Box–Cox transformed prior to statistical analysis. Physical–chemical properties (pH, DOC, EC and precipitation) were not transformed. Pearson product moment was used for testing for significant correlations. The significance level was set at $p = 0.05$. Principal component analysis was performed exploratively to check for correlations.

3. Results and discussion

3.1 Leachate concentrations

PFASs were detected in all landfill leachate samples. The total sum of 28 PFASs $\left(\sum_{28}\text{PFASs}\right)$ per landfill ranged from 320 to 11 000 ng L^{-1} (mean ± standard deviation: 1700 ± 2900; median: 630 ng L^{-1}), see Table 1 (data for individual PFAS is presented in Tables S4 and S5†). For the four short-chain PFASs these were from 68 to 6800 ng L^{-1} (mean: 980 ± 1800; median: 360 ng L^{-1}), whereas the 15 long-chain PFASs ranged from 140 to 2900 (mean: 530 ± 730; median: 290 ng L^{-1}). Hence, a substantial variation in leachate concentrations, more than two orders of magnitude, can be found at these diverse Norwegian landfills.

For four of the landfills (I, II, III and VI), there was data available from previous, recent sampling campaigns for comparison. The concentrations for landfills I, II and III were quite similar from the current and previous campaign (generally within a factor 1.5 of each other, or not statistically different), though landfill VI exhibited substantially higher concentrations in the current April 2018 data compared to the previous September 2017 data. The only landfill with a time series available was Landfill I, having 6 time points from 2017 to 2018. This time series found more variation in long-chain PFAS concentrations (relative standard deviation, rsd, of 81%) compared to short-chain PFASs (rsd of 14%). Time trends in PFAS leachate concentrations were studied for a Canadian landfill by Benskin *et al.*,[13] this study observed a peak in leachate emissions around mid-March to April for long-chain PFASs, but not for short-chain PFASs, which tended to be consistent throughout the year. Though the mechanisms of this are complex, and related to meteorological conditions (*e.g.* snow melt and precipitation), sorption and water properties (*e.g.* pH and ionic strength), less variability for short-chain PFASs than long-chain PFASs in landfill I is consistent with the Canadian landfill studied by Benskin *et al.*[13] However, what is inconsistent is the apparent difference in both short-chain and long-chain PFASs observed in landfill VI between September 2017 to April 2018, where the September 2017 concentrations seems more of a diluted version compared to the April 2018 sample; and may simply be due to a dilution event *e.g.* storm water. Later in this manuscript a correlation analysis is presented to other leachate parameters (pH, DOC, EC and precipitation).

Regarding the second-aim of this study to see if short-chain PFAS dominate in leachate emissions, in six out of ten landfills, there were higher concentrations of short-chain PFASs than long-chain PFASs, with short-chain to long-chain ratios ranging from 0.28 to 3.1 (Table 1). Overall, the short-chain PFBS contributed most to the $\sum_{28}$PFAS-concentration (30% based on mean concentrations). In leachate from Svalbard (landfill X), the major contributor was the long-chain PFOS (30%). At other landfills, PFOS contributed 1 to 20% of the $\sum_{28}$PFASs. Relatively lower overall PFOS-abundance and higher abundance of short-chain PFASs (especially PFBS) could indicate that short-chain PFASs are now the major contributors in landfill leachate, supporting the study's hypothesis. The role of time trends could also be considered here, as mentioned above, Benskin *et al.*[13] noticed a drop in long-chain PFASs after April, but not short-chain PFASs. Since sampling was mainly done in April, short-chain PFAS may have dominated further if sampling occurred later in the year. Sorption may also play a role. PFOS and other long-chain perfluoroalkyl acids sorb stronger to organic solids than some of their short-chain analogues;[25–27] for instance, average observed organic carbon partition coefficient, K_{oc} of PFOS and PFBS is 3.0 and 2.2, respectively.[28,29] Thus one would expect long-chain PFASs to leach slower than short-chain PFASs. The relatively high abundance of short chain PFAS, particularly PFBS, in leachate demonstrates that considerable amounts of PFBS (and PFBS precursor) containing waste have been deposed of in Norwegian landfills. The presence of PFOS in the leachates shows that the phase-out in new commercial products has not reduced their concentrations in leachate entirely; due to both the presence in waste being currently deposited at the landfills, and the lag-time of leaching from waste previously landfilled. There were significant correlations between $\sum_{28}$PFASs and several PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA), PFBS and 6 : 2 FTSA (Table S6†), indicating that these substances have correlating concentration and emission pathways across landfills.

In Table 2, leachate concentrations from the literature are compiled, in which it was possible for us to calculate the short-chain and long-chain concentrations and ratios in a manner similar to this study, because similar PFASs were analysed and the raw data was available. General caveats however with such comparisons are that the analysis methods can differ, PFASs analysed can differ, as can the type of leachate sampled (raw, diluted, treated), as indicated in Table 2, thus preventing exact comparisons, Eggen *et al.*[19] using non-target analysis (Table 2) reported relatively high $\sum_{16}$PFAS concentrations of 6123 ng L^{-1} for raw leachate in Norwegian landfills sampled in 2006 (which in comparison is higher than all landfills in this study except landfill VI), and a short- to long-chain ratio of 0.16; thus, indicating that short-chain PFASs have become more dominant in

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Table 1 PFAS concentrations (ng L^{-1}) in landfill leachates from landfill I-X (sampled in 2018). Values < LOQ are excluded from the calculations. PFDS, PF-3,7-DMOA, PFTrA, PFTA, PFHxDA, EtFOSA, MeFOSA, EtFOSE, MeFOSE, 4 : 2 FTSA and 8 : 2 FTOH were not detected > LOQ in any of the samples. n.a. = not analysed. I-S to III-S and VI-S = supplementary PFAS results obtained from the landfill operators (site-owner measurements) at the same respective sampling points as in this study

ID	Year, month (*n*)	∑Short-chain PFSAs[a] (<C6)	∑Long-chain PFSAs[b] (≥C6)	∑Short-chain PFCAs[c] (<C7)	∑Long-chain PFCAs[d] (≥C7)	∑Short-chain PFAS	∑Long-chain PFAS	Short : long-chain ratio	PFOS	PFOA	∑PFAS
I	2018, Jan. (1)	30	51	140	85	170	140	1.3	29	66	320
I-S	2017, Apr., June, Aug., Nov., 2018, May (5)	24 ± 10	150 ± 210	160 ± 24	110 ± 17	180 ± 26	260 ± 210	0.70 ± 0.40	100 ± 140	69 ± 10	470 ± 240
II	2018, April (1)	260	90	240	140	500	230	2.1	70	94	780
II-S	2016, April (1)	320	110	260	170	580	280	2.1	74	100	900
III	2018, April (1)	100	110	250	180	350	290	1.2	70	120	670
III-S	2018, April (1)	110	110	260	200	370	310	1.2	65	130	740
IV	2018, April (1)	47	26	380	110	430	140	3.1	15	72	590
V[e]	2018, May (1)	n.a.	87	68[e]	98	68[e]	190	0.37	51	98	432
VI	2018, April (1)	4200	200	2600	2700	6800	2900	2.3	120	1800	11 000
VI-S	2017, Sept. (1)	660	73	490	420	1200	490	2.3	36	270	2500
VII	2018, May (1)	850	220	1900	1000	2800	1200	2.3	65	660	4200
VIII[e]	2018, May (1)	n.a.	98	95[e]	240	95[e]	340	0.28	59	200	590
IX	2018, May (1)	7.3	71	130	210	130	280	0.47	36	170	420
X	2018, June (1)	7.2	200	130	200	130	400	0.33	160	120	540
Median	2016–2018 (18)	105	100	250	190	360	290	1.3	65	120	630
Mean ± SD		550 ± 1200	110 ± 58	510 ± 760	420 ± 690	980 ± 1800	530 ± 730	1.8 ± 0.94	68 ± 38	280 ± 460	1700 ± 2900
Min-max		7.1–4200	26–220	68–2600	85–2700	68–6800	140–2900	0.28–3.1	15–160	66–1800	320–11 000
Most abundant		(PFBS)	PFOS	PFHxA	PFOA	PFBS	PFOA	—	—	—	PFBS

[a] PFBS. [b] PFHxS, PFHpS and PFOS. [c] PFBA, PFPeA and PFHxA. [d] PFHpA, HPFHpA, PFOA, PFNA, PFDA, PFUnDA and PFDoA. [e] Sample analysis by ALS (different from others in this table); PFBS was not analysed in these samples, which could explain their relatively lower ∑short-chain concentrations.

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Table 2 Comparison of short- (PFBS, PFBA, PFPeA and PFHxA) and long-chain (PFHxS, PFHpS, PFOS, PFDS, PFHpA, HPFHpA, PFOA, PFNA, PFDA, PFUnDA, PF-3,7-DMOA, PFDoA, PFTrA, PFTA and PFHxDA) PFAS concentrations (ng L^{-1}) in landfill leachate from selected studies. Values < LOQ are excluded from the calculations

Area, year (number of landfills/raw, diluted, mixed)	∑Short-chain PFASs mean ± SD (min–max)	∑Long-chain PFASs mean ± SD (min–max)	Short : long-chain ratio	n PFASs	$\sum_n$PFASs	Reference
Norway, 2017–2018 (10, mixed)	980 ± 1800 (68–6800)	530 ± 730 (140–2900)	1.8 ± 0.94 (0.28–3.1)	28	1700 ± 2900 (320–11 000)	This study
Norway, 2006 (2, raw)	757	4784[a]	0.16	16	6123	Eggen *et al.*, 2010[19]
Canada, 2009 (1, raw)	2812 ± 1109 (1424–5150)	2719 ± 2160 (1021–7738)[b]	1.0	24	11 000 ± 10 000 (3800–3600)	Benskin *et al.*, 2012[13]
Sweden, 2015 (10, unknown)	171 ± 137 (<LOQ–508)	123 ± 78 (<LOQ–269)[c]	1.4	26	487 (0.30–1300)	Gobelius *et al.*, 2018[23]
Spain, 2015 (4, raw)	576 ± 317 (125–852)	506 ± 113 (413–663)[d]	1.1	16	1082 (639–1379)	Fuertes *et al.*, 2017[24]

[a] With the exclusion of PFHpS, HPFHpA, PF-3,7-DMOA, PFTA and PFHxDA; note this was measured using non-target analysis. [b] With the exclusion of PFHpS, HPFHpA, PFDA, PF-3,7-DMOA, PFTrA and PFHxDA. [c] With the exclusion of PFHpS, HPFHpA, PFOA, PFDA and PF-3,7-DMOA. [d] With the exclusion of HPFHpA, PF-3,7-DMOA and PFHxDA.

Norway recent years (though this data is influenced by a non-target analysis method being used).

Benskin *et al.*[13] reported higher $\sum_{24}$PFAS concentrations in flow-through (raw) leachate from a landfill in Canada (11 000 ± 10 000 ng L^{-1}) in 2010 than the Norwegian concentrations presented in this study. The mean short- to long-chain ratio was 1.0 in Benskin *et al.*,[13] indicating short-chain PFASs were not dominant as in six of the landfills in this study. It is noteworthy that the short-chain PFBS was found in lower concentrations in this 2010 Canadian landfill (mean: 94 ± 41 ng L^{-1}) than the mean of the 2018 Norwegian landfills (550 ± 1200 ng L^{-1}); potentially indicating increased landfilling of PFBS.

A study of Swedish landfills analysed in 2015 (ref. 23) reported $\sum_{26}$PFAS concentrations in the same range, although somewhat lower than this study (Table 2). The short- to long-chain ratio we calculate from their data was 1.4 (Table 2), which agrees with the general dominance of short-chain PFASs in landfills from this study. Four landfills from northern Spain were also sampled in 2015,[24] and the concentration of s in raw leachate was in the same range as $\sum_{28}$PFASs in this study (Table 2). The short- to long-chain ratio from the data in Fuertes *et al.*[24] was 1.1 (Table 2).

There are other leachate studies in the literature; however, it is not as clear to calculate the short-chain : long-chain PFAS ratio as done here. Thus, instead the sum PFAS concentrations and the presence of short-chain PFAS are discussed. According to Kallenborn *et al.*,[21] $\sum_{7}$PFASs in landfill leachate from five Norwegian landfills in 2004 ranged from 199 to 1538 ng L^{-1} (mean: 673 ± 552, median: 468), which is lower than this study, but also with fewer PFASs, with many short-chain PFCAs not analysed for or found in low levels. A 2010 study of 22 landfills in Germany by Busch *et al.*[17] reported $\sum_{43}$PFASs in untreated leachate from 31 to 12 922 ng L^{-1} (mean: 6086 ng L^{-1}), which is relatively higher than the $\sum_{28}$PFASs in this study, and perhaps influenced by including more congeners. This study reported short-chain PFASs were dominating, with the two most abundant congeners PFBA (mean contribution 27%) and PFBS (mean contribution 24%).[17] In 2012, Li *et al.* published $\sum_{13}$PFAS concentrations from 30 to 21 000 ng L^{-1} in leachate from 28 landfills and dumpsites in Canada,[18] these appeared dominated by long-chain PFCAs. In a study of four U.S. landfills sampled in 2006 by Huset *et al.*,[15] $\sum_{24}$PFASs ranged from 2688 to 7415 ng L^{-1} in raw leachates, though interestingly already with high concentration of PFBA (up to 1700 ± 63 ng L^{-1}) and PFBS (up to 890 ± 100 ng L^{-1}). In raw leachate from five municipal landfill sites in China, sampled in 2013, the $\sum_{14}$PFAA concentrations in 2015 ranged from 7280 to 292000 ng L^{-1} (mean: 82 100 ng L^{-1}),[22] which is much higher than $\sum_{28}$PFASs this study. PFOA and PFBS were the most abundant.[22] It appears from this review that short-chain PFBS and PFBA have been a dominant component of leachate in some areas since 2006, though in the more recent studies this seems to be more typically the case, as in Norway.

3.2 Emissions

The yearly amount of leachate generated at each landfill were provided by the landfill operators. This was used for estimation of the yearly PFAS release from each site, assuming the concentrations in Table 1 were consistent all year round (Table S7,† illustrated in Fig. 1); though as indicated above, sampling was done at time when concentrations of long-chain PFAS, in particular, are highest. The annual $\sum_{28}$PFAS release at each landfill was estimated to range from 9.2 to 510 g per year (mean: 160 ± 160 g per year; median: 100 g per year). For short-chain PFASs these were from 2.2 to 310 g per year (mean: 87 ± 98;

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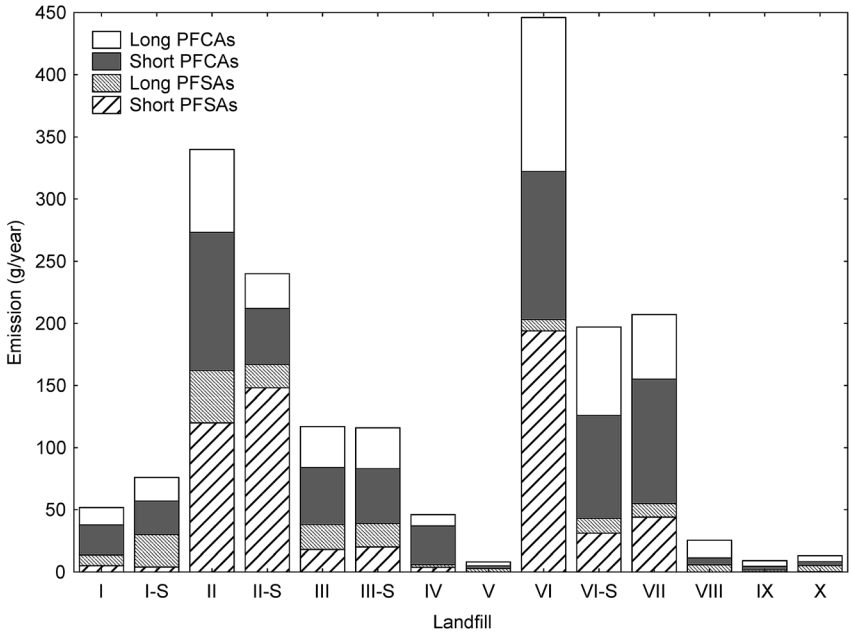

Fig. 1 Stacked bar chart of estimated yearly PFAS emissions (g per year, based on the amount of leachate generated at each landfill (Table S2†) and the concentrations in Table 1) of long-chain and short-chain perfluorinated sulfonates (PFSAs) and perfluoroalkyl carboxylic acids (PFCAs) for the Norwegian landfills in this study (Table S7†).

median: 49 g per year), and long-chain PFASs ranged from 5.9 to 130 g per year (mean: 47 ± 39; median: 46 g per year). One of the sites with the lowest PFAS emissions per year was in the high Arctic Site in Svalbard (landfill X), which is located in the Arctic, where the climate is relatively cold, causing relatively lower leachate production volumes of approximately 25 000 m^3 per year compared to other landfills in the present study (mean: 113000 ± 135000 m^3 per year, Table S2†); and therefore leaching from waste is also slowed.

It is important to bear in mind that there are several limitations to the emission estimates provided in the present study. These are (1) most sites had one sampling day, so seasonal, climate and hydrological factors could not be evaluated; (2) sampling occurred around April, which as discussed above is potentially when peak emissions for long-chain PFAS occur;[13] (3) discharge volume is provided by the operators and their accuracy may vary; and (4) the leachate samples could to some degree be influenced by storm water and groundwater.

Comparisons with emission estimates in the literature need to take into account different PFASs being considered, as well as annual leachate volumes, which can vary tremendously. Therefore comparisons with emissions in the literature presented below must be considered with these limitations in mind. The study on Norwegian landfills in 2006 (ref. 19) reported emissions of 2.1 kg per year, which is higher than any of the landfills in the present study, due to relatively higher PFAS concentrations (Table 2), as well as a relatively large annual loading of 345 000 m^3 leachate per year.[19] The study of the Canadian landfill in 2010[13] reported annual $\sum_{24}$PFAS emissions from 8.5 to 25 kg per year (mean: 16 kg per year) which is considerably higher than the landfills in this study. This is mainly due to the high annual volume of leachate produced at the Canadian site, of 2.2×10^6 m^3 per year,[13] compared to volumes from 2.2×10^4 to 4.6×10^5 m^3 per year in this study (Table S2†).

To extrapolate these results to the national level, which introduces new uncertainties, a previous report on leachate emissions in Norway concluded that a total volume of up to 1.0×10^7 m^3 is emitted from Norwegian landfills, with 55% of emissions being sent to wastewater treatment plants (WTP) and 45% to the environment.[30] Based on the mean concentrations provided in Table 1 multiplied by national leachate volume, this implies national $\sum_{28}$PFASs release from Norwegian landfills from 3.2 to 110 kg per year (mean: 17 ± 29; median: 6.3 kg per year) (Table S8†). Considering the current Norwegian population is approximately 5.3 million, this would correspond to a mean per capita emission factor of 3.2 ± 5.5 mg per year per person. For short-chain PFASs these were from 0.68 to 68 kg per year (mean: 9.8 ± 18; median: 3.6 kg per year), and long-chain from 1.4 to 29 kg per year (mean: 5.3 ± 7.3; median: 2.9 kg per year) (Table S8†).

National estimates have also been presented for other countries, such as China,[22] Germany,[17] and the U.S.[16] These were derived by multiplying average emission rates for landfills included in their study by the number of landfills in the country, rather than based on leachate volumes as here. For the purpose of comparison, the national emission levels are divided by population to give per capita emission factors. Yan *et al.*[22] estimated in 2015 the Chinese national emission of $\sum_{14}$PFAAs to groundwater from landfill leachate to be 3110 kg per year, based on the mean concentration of 82 100 ng L^{-1} from five municipal

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landfill sites in China, and the average amount of leachate generated per year (4.74×10^7 m^3 per year), assuming that 80% of the landfills were not lined. This would correspond to 2.2 mg per person per year (assuming a Chinese population of 1.39 billion), which is similar to this study (though this study considered all aqueous emissions from landfills, not just groundwater emissions). The German national landfill emission in 2009 of $\sum_{43}$PFASs was estimated at 88 kg per year,[17] corresponding to 1.1 mg per person per year (assuming population 81.8 million in 2009). A 2013 survey of U.S. landfills estimated $\sum_{19}$PFAS emissions from 563 to 638 kg per year,[16] or 1.8 to 2.0 mg per person per year. A study from Spain[24] estimated the annual discharge of $\sum_{16}$PFASs from four landfill sites serving 1.8 million people was 1.2 kg per year, which implies an emission rate of 0.7 mg per person per year.

Despite limitations, this comparison resulted in per capita emission factors that were very similar of those that can be derived in other regions, from 0.7 (Northern Spain), 1.1 (German), 1.8–2.0 (U.S.), 2.2 to groundwater (China), to 3.2 (Norway) mg per day per person.

Compared to total per capita emission factors from all sources of PFAS, not just landfills, based on river and water treatment plant data in the literature, the contributions of landfills are relatively low. For instance, considering just PFOS, total per capita emissions from all sources for the EU in 2009 were estimated at 9.9 mg per year per person.[31] In our 2018 study, Norwegian landfill emissions contribute roughly 0.35 mg per day per person emissions of PFOS, which would be 3.5% of the European per capita emissions in 2009. Total flux of sewage-derived PFOS from Japan was in 2008 estimated to be 3.6 tonnes per year, corresponding to 28 mg per year per person,[32] which is almost two orders of magnitude higher than the per capita emission factors derived here. Part of this may be attributable to PFOS emissions having declined in recent years, since these other per capita emissions were derived.

3.3 Sediment concentrations

PFASs were detected in all sediment samples ($n = 8$). The $\sum_{30}$PFASs concentration (dry weight) ranged from 8.5 to 120 μg kg^{-1} (mean: 47 ± 36; median: 41 μg kg^{-1}) (Table 3, with data for individual substances in Table S9†). The major contributors were long-chain PFASs, such as PFUnDA and PFOS. In addition, long-chain PFOS-precursors (especially EtFOSAA and MeFOSAA) contributed to a substantial amount of the $\sum_{30}$PFASs (46%). Long-chain PFASs are more likely to adsorb onto surfaces and partition into soils and sediments.[25,26]

Eggen *et al.*[19] reported that long-chain PFCAs, FOSA and EtFOSA were detected at relatively high concentrations in sediment samples from a Norwegian landfill sampled in 2006, with total PFAS concentrations (dry weight) ranging from 1.3 to 382 μg kg^{-1} (mean: 61 μg kg^{-1}), which is in the same range as this study (Table 3), indicating little change. According to

Table 3 Average PFAS concentrations (μg per kg dry weight) sediments from landfill I, IV–VII and IX–X (sampled in 2018). Values < LOQ are excluded from the calculations. n.a. = not analysed. PFHpS, HPFHpA, PF-3,7-DMOA, PFTA and PFHxDA, EtFOSA and MeFOSA were not detected > LOQ in any of the samples

ID	∑Short-chain PFSAs[a] (<C6)	∑Long-chain PFSAs[b] (≥C6)	∑Short-chain PFCAs[c] (<C7)	∑Long-chain PFCAs[d] (≥C7)	∑Short-chain PFAS	∑Long-chain PFAS	PFOS	PFOA	∑PFOS precursors[e]	∑PFAS
I	<LOQ	26	<LOQ	6.6	<LOQ	32	25	1.6	33	72
IV	<LOQ	11	0.25	3.4	0.25	14	11	1.2	24	46
V	<LOQ	14	0.49	1.9	0.49	16	14	1.3	12	37
VI	<LOQ	1.3	<LOQ	0.69	<LOQ	2.0	1.3	0.21	0.74	8.8
VII	3.3	9.4	4.9	7.5	8.2	17	9.0	3.6	84	120
IX	<LOQ	15	<LOQ	1.8	<LOQ	17	15	1.8	7.9	40
X-I[f]	<LOQ	24	<LOQ	20	<LOQ	44	24	0.54	8.26	42
X-II[f]	<LOQ	3.9	<LOQ	1.4	<LOQ	5.3	3.9	0.23	0.59	8.5
Median	3.3	13	0.49	2.6	0.49	16	13	1.3	10	41
Mean ± SD	3.3	13 ± 8.7	1.9 ± 2.6	5.4 ± 6.4	3.0 ± 4.5	18 ± 14	13 ± 9	1.3 ± 1.1	21 ± 28	47 ± 36
Min–max	3.3–3.3	1.3–26	0.25–4.9	0.69–20	0.25–8.2	2.0–44	1.3–25	0.21–3.6	0.59–84	8.5–120
Most abundant	(PFBS)	PFOS	PFHxA	PFUnDA	PFBS	PFOS	—	—	EtFOSAA	EtFOSAA

[a] PFBS. [b] PFHxS, PFOS and PFDS. [c] PFBA, PFPeA and PFHxA. [d] PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoA and PFTrA. [e] The PFOS precursors, EtFOSAA, EtFOSE, MeFOSAA, MeFOSE and FOSAA, were only analyzed in sediments and not in leachate, and hence are not included in the ∑Short-chain and ∑Long-chain data for consistency. [f] X-I was sampled from the outlet of a leachate pond, at the same place as the leachate sample from landfill X. X-II was sampled from a stream downstream the leachate pond.

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a literature survey by the Norwegian Environment Agency from 2008,[20] PFHxS and PFOS were detected in all sediment samples from 8 Norwegian landfills, wherein the $\sum_{13}$PFASs ranged from approximately 0.0013 to 0.024 μg per kg d.w.,[20] which is lower than the present study. However, it is noted that PFAS substances such as EtFOSAA and MeFOSAA were not measured in that study, whereas these were measured at relatively high concentrations in this study (Table 3).

3.4 Sediment-leachate distribution

The ratio of C_{sediment} to C_{leachate}, which will be referred to as $Q_{\text{sed/leachate}}$ (units L kg^{-1}), measured at the specific landfills is presented in Table S10.† It is noted that the $Q_{\text{sed/leachate}}$ value cannot be considered an equilibrium distribution coefficient, K_D, as it is uncertain if at each site C_{sediment} is in equilibrium with C_{leachate}, particularly for sites where sediments and leachates were sampled in different locations. Among the analysed PFASs, only PFOA and PFOS were detected > LOQ for both leachate and sediment samples at all landfills. The calculated, average log $Q_{\text{sed/leachate}}$ − values for PFOA and PFOS were 0.74 ± 0.78 and 2.3 ± 0.64, respectively, ranging from −0.93 to 1.38 and from 1.0 to 2.9 (Table S10†). These happen to be similar to log K_D (L kg^{-1}) reported in the literature for different soils and sediments, which cover broad ranges; *e.g.*: Schedin:[33] 2013: 0.3 ± 0.4 and 1.2 ± 0.6 for PFOA and PFOS; Kwadijk *et al.*:[28] 1.83 ± 0.40 and 2.35 ± 0.35 for PFOA and PFOS. According a literature review by Zareitalabad *et al.*,[29] log K_d-values for PFOA and PFOS ranged from −1.2 to 0.96 and from −0.7 to 1.9, respectively.

3.5 Correlation analysis

PFAS concentrations in landfill leachates are to some extent dependent on the leachate's water properties.[13,34] Principal component analysis on the variables $\sum_{28}$PFAS, DOC, pH and EC were conducted for all obtained samples (Fig. S1†). There was a clear correlation between $\sum_{28}$PFAS with increasing EC. Significant correlations between $\sum_{28}$PFASs and some individual PFASs (PFHxS, PFBA, PFPeA, PFHxA, PFHpA and PFOA) with EC were found (Table S11†), which is in agreement with a study by Benskin *et al.*,[13] as they observed significant correlations between PFBA, PFPeA and PFHxA with increasing EC. Possible explanations given in Benskin *et al.*[13] were the observation of decreased sorption to some clays with increasing EC due to competition effects with other anions, *e.g.* chloride, for the anionic sorption sites (this is discussed further below). However, because in this study a correlation with $\sum_{28}$PFASs and EC was evident, there may be some influence of the dilution of raw leachate with storm water or groundwater that helps account for this observation. There were no clear associations between PFAS-concentrations and pH or DOC in leachate. On the contrary, Gallen *et al.*[34] reported increasing concentrations of several PFAS compounds with increasing pH and DOC in leachate. In our case, this may be in part due to a limited range of pH values (from 6.4 to 7.8) and DOC values being relatively clustered between 28–125 mg L^{-1}, except for one outlier at 1322 mg L^{-1} (ESI, Table S2†).

In principle, factors such as pH, EC and DOC would influence the sorption of ionic PFASs. Thus, PCA biplots were made for the $Q_{\text{sed/leachate}}$ − values of PFOS and PFOA, where $Q_{\text{sed/leachate}}$ is considered a proxy for sorption. As their $Q_{\text{sed/leachate}}$ − values were strongly correlated ($r = 0.9$, $p = 0.001$), only the PCA-biplot with $Q_{\text{sed/leachate}}$ − values for PFOS is shown in Fig. S2.† $Q_{\text{sed/leachate}}$ and yearly leachate production volume were positively correlated, implying higher sorption to the sediment phase with increasing leachate volume. This could be accounted for by considering a Freundlich like sorption behaviour (*i.e.* sorption increases with increasing water to sediment ratios, leaving behind the more strongly sorbed residues). There were no significant correlations between $Q_{\text{sed/leachate}}$ and pH or DOC. However, there was a negative correlation between PFOS $Q_{\text{sed/leachate}}$ and EC, which indicates relatively less partitioning to the sediment phase with increasing EC. This could be accounted for by increasing competition for anionic sorption sites. It is not uncommon for leachate sediments to be rich in metal oxides,[35] which can be positively-charged and therefore contain anion exchange sites, depending on the pH and salt composition. Studies by Wang *et al.*[36,37] indicated that the sorption of PFOS and PFOA on the aluminium oxides boehmite and alumina decreased with increasing ionic strength. In contrast, negatively-charged clays like the phyllosilicate bentonite show negligible sorption of PFAS,[38] and therefore would be less influenced by EC than positively-charged metal oxides. Thus, the stronger sorption with higher flow volumes and lower EC seems to match well with mechanistic expectations for a Freundlich like behaviour: dilution would lower EC and PFAS concentrations, and therefore increase sorption *via* less competition for anion-exchange sites to *e.g.* metal oxide surfaces.

4. Environmental implications

This study estimated that the release of $\sum_{28}$PFASs from Norwegian landfills was in the range 3.2 to 110 kg per year (mean: 17 ± 29; median: 6.3 kg per year); though, it should be kept in mind there were several assumptions used to make this data, ranging from the (limited) sampling campaign to the assumption that the obtained data were representative of yearly emissions. Future sampling campaigns should address this. As hypothesized, due to the shift towards short-chain PFAS chemistry, emissions of short-chain PFASs appear to dominate over emissions of long-chain PFASs. Short-chain PFASs are generally more mobile, as made evident by the difficulty in this study in obtaining sediment concentrations of PFBS, despite it being the most dominating PFAS in leachate. Future studies could confirm this trend, and relate emission levels with current production levels, uses and disposal of specific PFASs. Landfills are one of the many sources of PFAS emissions. Here we presented a very rough estimation based on a literature comparison that landfills contribute approximately 3.5% of all

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European per capita emissions of PFOS. A similar estimation for other PFASs could not be made. Future studies could seek to quantify these values better through comparison of different emission sources, *e.g.* households, fire training facilities, airports, and industries, to their respective contributions to wastewater treatment plants, groundwater, rivers, and other recipients. Even though the composition of PFASs in commerce and in landfills change over time, landfills can continue to release contaminants like PFASs for years to come. If the emissions in Norway were consistent for 100 years at 17 kg per year, then there would be 1.7 tonnes emitted from one, relatively small country. To put this into context, a recent global emission inventory of PFHxS and PFDS has estimated that between 1958 and 2015, 120–1022 and 30–378 tons, respectively, have been emitted.[39] To prevent landfills from contributing to future PFAS emissions, proper management strategies are key, such as developing low cost leachate treatment facilities, including PFAS in leachate monitoring, better understanding of leaching mechanisms from waste, and evaluating how concentrations of PFAS in leachate changes with PFAS levels in deposited waste.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This work was supported by the Norwegian Research Council through FANTOM (NFR project # 231736/F20), basis funding for the NGI (GBV) and NIBIO. The authors thank all landfill operators for their cooperation in this study.

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