Electronic Supplementary Information

Leachate emissions of short- and long-chain per- and polyfluoralkyl

substances (PFASs) from various Norwegian landfills

Heidi Knutsen^{1*}, Trond Mæhlum², Ketil Haarstad², Gøril Aasen Slinde¹, Hans Peter H. Arp^{*,1,3}

¹Norwegian Geotechnical Institute (NGI), P.O. Box 3930 Ullevål Stadion, N-0806 Oslo,

Norway. E-mail: hkn@ngi.no; hpa@ngi.no; Tel: +47 478 51 733, +47 950 02 667

² Norwegian Institute of Bioeconomy Research (NIBIO), P.O. Box 115, N-1431 Ås, Norway

³Norwegian University of Science and Technology (NTNU), NO-7491 Trondheim, Norway

Table of Contents

Table S1 – List of landfills (I-X) in the present study. Main waste fractions for landfills are given. Due to confidentiality reasons, the identities of landfills are anonymized.

Table S2 – Leachate, pH, electrical conductivity (EC), total organic carbon (TOC), ammonium (NH_3/NH_4^+) , total nitrogen and chemical oxygen demand (COD) for studied landfills. 24h and 2-week precipitation as well as yearly volumes of leachate generation are provided.

Table S3 – Per- and Polyfluoralkyl substances analysed in the present study, as well as their acronyms and molecular formulas.

Table S4 - Measured concentrations of PFAS substances in leachate samples analysed at Eurofins Environment Testing Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentration values are given as ng/L. LOQ values are provided.

Table S5 - Measured concentrations of PFAS substances in leachate samples analysed at ALS Laboratory Group Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentrations are given as ng/L. LOQ values are provided.

Table S6 – Correlations (r-values) of box-cox transformed leachate PFAS concentrations. Bold values indicate significant (p < 0.05) correlations.

Table S7 - PFAS emission data (gram/year) for 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, and are thus excluded from the calculations. n.a. = not analysed. I-S to III-S and VI-S = emissions based on supplementary PFAS results obtained from the landfill operators at the same sampling points as in this study (I-III and VI). Emissions were calculated as average leachate concentrations (Table 1) times yearly leachate volume (Table S2).

Table S8 - Estimated national PFAS release (kg/year), calculated based on the overall concentration data given in Table 1 and the estimated total volume of landfill leachate emitted from Norwegian landfills¹ ($1.0x10^7 \text{ m}^3$). SD = standard deviation.

Table S9 - Measured concentrations of PFAS substances in sediment samples analysed at Eurofins Environment Testing Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentration values are given as μ g/kg dry matter, except for dry matter content (DM) which is reported as %. LOQ values are provided.

Table S10 - Log $Q_{sed/leachate}$ -values based on PFOA-, PFOS- and \sum PFAS-concentrationsmeasured in leachate and sediment from landfill I, IV, V-VII, IX and X.

Table S11 - Correlations (r-values) of box-cox transformed leachate PFAS concentrations with electrical conductivity (EC). Bold values indicate significant (p < 0.05) correlations.

Figure S1 - PCA biplot that combines loadings (potential correlations between different variables, i.e. the parameters \sum_{28} PFAS (ng/L), DOC (mg/L), pH and EC (μ S/cm)) and scores (potential correlations between different objects/samples: leachates from landfill I-X).

Figure S2 - PCA biplot that combines loadings (potential correlations between different variables, i.e. the parameters PFOS $Q_{sed/leachate}$ (L/kg), DOC (mg/L), pH, EC (μ S/cm), yearly leachate production volume (m³/year), 24 hour and 2 week precipitation (mm)) and scores (potential correlations between different objects/samples, i.e. landfills were both sediment and leachate was analysed).

Ten Norwegian landfills, including one site in Svalbard (Table S1), were considered. Due to confidentiality reasons, the identities of the landfills are anonymized. The main waste fractions for each landfill are listed in Table S1.

Landfill ID	Location	Waste disposal (years)	Waste
I		1970-	Residual waste from households, garden waste, sewage sludge, contaminated soil (categorised as hazardous) and concrete.
Π	_	1998-	Contaminated soil mostly
III	_	1971-	Contaminated soil and concrete, asbestos, oil slam and garden waste.
IV	_	1988-	Residual waste from households, plastic, cardboard, paper, glass, metal, wood, WEEE, textiles, tires and industrial waste (e.g. contaminated soil), sand and oil slam.
V	-	1962-1997	Mixed MSW with some industrial waste.
VI	– Norway	1960-	MSW, garden waste, demolition drift. From 2009: only waste categorized as inert.
VII	_	1962-	Residual waste from households, waste categorised as hazardous. WEEE, glass, batteries, garden waste and smaller amounts of ash for metal recycling and contaminated soil.
VIII	_	1973-2009	Residual waste from households and industrial waste, recyclable waste, waste categorised as hazardous, wood, garden waste, WEEE, freezers and refrigerators, tires and metal.
IX	_	1971-1995	Mixed MSW with some industrial waste.
X	Svalbard		Until 2007: residual waste from households, industrial waste (næringsavfall), waste from construction and mine industry, slag and ash from a utility company and impregnated wood. From 2007: inert or non-degradable waste such as gypsum, steel, concrete, insulation, plastic pipelines, ash and slag.

Table S1 List of landfills (I-X) in the present study. Main waste fractions for landfills are given. Due to confidentiality reasons, the identities of landfills are anonymized.

Leachate and sediment from landfill I was sampled from a manhole directly from the underground leachate culvert that drains the bottom of the landfill, after preliminary sedimentation in a retention pond. The sampling point was located just before discharge into a fjord. At the sampling point, the sediment is in direct contact with the leachate. The leachate could be influenced by storm water and groundwater from the older parts of the landfill, where the liner does not fulfil modern requirements.

Untreated leachate from landfill II was sampled from a pumping station which drains leachate through the leachate pumping pipeline from the bottom of the landfill. Thus, the leachate was sampled before off-site treatment in a municipal wastewater treatment plant. The leachate could be influenced by storm water and groundwater from the older part of the landfill, where the liner does not meet today's requirements.

Leachate from landfill III was sampled from a retention pond which collects leachate from the landfill via ditches and piping. The leachate could be influenced by storm water. However, in recent time, a number of measures have been taken to prevent this (e.g. the establishment of a larger retention pond), and the influence is therefore considered limited.

At landfill IV, the leachate is pumped to a lagoon with aeration to promote the biological oxidation. Between the lagoon outlet and the sedimentation pond, a coagulant agent is added. Overflow water from the sedimentation pond is lead directly to a sump and then pumped to a municipal treatment plant. Leachate is sampled before pumping to the off-site treatment plant. The leachate could be influenced by storm water and groundwater, as the older parts of the landfill has improper lining compared to today's requirements. Sediment is sampled from the sludge dewatering plant.

Leachate from landfill V was sampled from a manhole at the leachate pipe which is originating from the bottom of the landfill. The landfill has no liner, but it is placed in a ravine of clay which acts as natural lining. Therefore, some clean groundwater could be of influence, but this is assumed to be limited, based on the topography and the size of the landfill. Sediment was sampled from the bottom of the manhole which leads out of the landfill. Here, sediment will settle due to the oxidation of iron with precipitation as well as some sedimentation. The sediment is in direct contact with the leachate leading out of the landfill.

Landfill VI is located in a hydrologically isolated area, such that transport of storm water inand out of the landfill is limited. In the older part of the landfill, the leachate can infiltrate to the groundwater (approximately 40 m below the infiltration pond). However, the new part of the landfill is established with sufficient lining, and the untreated leachate that was sampled represents this newer part of the landfill, sampled from a inflow drain before water treatment and pumping to the infiltration pond. Sediment was sampled from the infiltration pond.

Leachate from landfill VII was sampled from the raw leachate pumping pipeline coming from a leachate collection dam. The dam however also collect drainage water from a small landfill for inert waste recently established, but most of the drainage (est. > 90%) comes from the old MSW landfill. The landfill is established on a geological barrier of natural clay, and the influence of groundwater is therefore limited. Sediment was sampled from a sedimentation area within the leachate collection dam.

Leachate from landfill VIII was sampled from a borehole which is in contact with the groundwater at the older part of the landfill. Sampling involves the upper part of the groundwater, and it is assumed that the leachate to some degree is influenced by clean groundwater.

Raw leachate from landfill IX was sampled directly from the inflow drain before pumping to a treatment plant. The landfill is established without liner. Thus, influence of groundwater is possible. Sediment was sampled from an evaporation pond where leachate is collected.

Leachate from landfill X was sampled at the outlet of the leachate pond. There could be some influence of storm water. Sediment was sampled from two stations: X-1: from the sedimentation pond and X-2: downstream the sedimentation pond.

Landfill	Sampling date	рН	EC (μS/cm)	DOC (mg/L)*	24 h precipitation (mm)	2 week precipitation (mm)	Yearly leachate volume (m ³ /year)	Ammonium- nitrogen NH ₃ /NH ₄ ⁺ (mg/L) ¹	Nitrogen, total (mg/L) ¹	COD (mg/L) ¹	Sampling point
Ι	23.01.2018	7	1129	73	6.5	44.7	169580	n.a.	n.a.	n.a.	а
II	09.04.2018	6.7	2120	n.a.	4.8	42.4	462278	n.a.	n.a.	n.a.	а
III	09.04.2018	6.9	1453	n.a.	1.2	28	180629	n.a.	n.a.	n.a.	b
IV	03.04.2018	7.6	3540	76	0	0	81044	n.a.	n.a.	n.a.	b
V	23.05.2018	6.4	1723	38	0	12.4	32000	54	57	112	а
VI	24.04.2018	7.3	7310	1322	0	4.9	46230	696	852	6046	а
VII	30.05.2018	7.2	12120	125	0	0.2	52000	251	332	705	а
VIII	29.05.2018	7.2	1533	54	0	0.5	59000	17	23	189	c
IX	29.05.2018	6.8	1578	22	0	1.4	22000	21	29	67	а
X	16.06.2018	7.8	2460	n.a.	n.a.	n.a.	24750	n.a.	n.a.	n.a.	d
n a = not avai	lable										

Table S2 Leachate, pH, electrical conductivity (EC), total organic carbon (TOC), ammonium (NH_3/NH_4^+), total nitrogen and chemical oxygen demand (COD) for studied landfills. 24h and 2-week precipitation as well as yearly volumes of leachate generation are provided.

n.a. = not available

¹Not measured in the sampled leachates due to cost constraints – the concentrations are provided by landfill operators and represent measurements from 2017-2018 at the same sampling point as this study. ^aUnderground culvert or leachate pipe as accessed through a manhole or drain; ^bPond; ^cBorehole; ^dStream

Compound class	Name	Acronym	Molecular formula
	Perfluorbutanoic acid	PFBA	C ₃ F ₇ CO ₂ H
	Perfluoropentanoic acid	PFPeA	C ₄ F ₉ CO ₂ H
	Perfluorohexanoic acid	PFHxA	C ₅ F ₁₁ CO ₂ H
	Perfluoroheptanoic acid	PFHpA	C ₆ F ₁₃ CO ₂ H
	7H-dodecafluoroheptanoic acid	HPFHpA	C ₆ HF ₁₂ CO ₂ H
	Perfluorooctanoic acid	PFOA	C ₇ F ₁₅ CO ₂ H
Day and naturfly aring tod	Perfluorononanoic acid	PFNA	C ₈ F ₁₇ CO ₂ H
Per- and polyfluorinated carboxylic acid (PFCA)	Perfluorodecanoic acid Perfluoro-3,7-dimethyloctanoic acid	PFDA PF-3,7-DMOA	C ₉ F ₁₉ CO ₂ H
	Perfluoroundecanoic acid	PFUnDA	$C_{10}F_{21}CO_2H$
	Perfluorododecanoic acid	PFDoA	C ₁₁ F ₂₃ CO ₂ H
	Perfluorotridecanoic acid	PFTrA	C ₁₂ F ₂₅ CO ₂ H
	Perfluorotetradecanoic acid	PFTA	C ₁₃ F ₂₇ CO ₂ H
	Perfluorohexadecanoic acid	PFHxDA	C ₁₅ F ₃₁ CO ₂ H
	Perfluorobutanesulfonic acid	PFBS	C ₄ F ₉ SO ₃ H
	Perfluorohexanesulfonic acid	PFHxS	C ₆ F ₁₃ SO ₃ H
Perfluorinated sulfonate	Perfluoroheptane sulfonate	PFHpS	C ₇ F ₁₅ O ₃ H
(PFSA)	Perfluorooctane sulfonic acid	PFOS	C ₈ F ₁₇ SO ₃ H
	Perfluorodecane sulfonic acid	PFDS	C ₁₀ F ₂₁ SO ₃ H
	Perfluorooctane sulfonamide	FOSA	C ₈ F ₁₇ SO ₂ NH ₂
Perfluorosulfonamide (FOSA)	N-methyl-perfluorooctan esulfonamide	MeFOSA ¹	C ₈ F ₁₇ SO ₂ NHCH ₃
	N-ethyl-perfluorooctane sulfonamide	EtFOSA ¹	C ₈ F ₁₇ SO ₂ NHCH ₂ CH ₃
Perfluorooctane	Perfluoroctane sulfonamido acetic acid	FOSAA	C ₈ F ₁₇ SO ₂ NHCH ₂ CO ₂ H
sulfonamido acetic acid (FOSAA)	N-methyl-perfluorooctane sulfonamido-acetic acid	MeFOSAA	C ₈ F ₁₇ SO ₂ NCH ₃ CHCO ₂ H
(105AA)	N-ethyl-perfluorooctane sulfonamido- acetic acid	EtFOSAA	$C_8F_{17}SO_2N(C_2H_5)_2CH_2CO_2H$
Perfluoralkylsulfonamide	N-methyl-perfluorooctane sulfonamido ethanol	MeFOSE ¹	C ₈ F ₁₇ SO ₂ N(CH ₂) ₂ CH ₃ OH
alcohol (FOSE)	N-ethyl-perfluorooctane sulfonamido ethanol	EtFOSE ¹	C ₈ F ₁₇ SO ₂ N(CH ₂ CH ₂ OH)(CH ₂ CH ₃)
Fluorotelomer sulfonate	4:2 fluorotelomer sulfonic acid	4:2 FTSA	$C_6H_4F_9SO_3H$
(FTSA)	6:2 fluorotelomer sulfonic acid	6:2 FTSA	$C_8H_4F_{13}SO_3H$
(FISA)	8:2 fluorotelomer sulfonic acid	8:2 FTSA	$C_{10}H_4F_{17}SO_3H$
Fluorotelomer alcohol (FTOH)	8:2 fluortelomer alcohol	8:2 FTOH	F(CF ₂) ₈ CH ₂ CH ₂ OH

Table S3 Per- and Polyfluoralkyl substances analysed in the present study, as well as their acronyms and molecular formulas.

(FTOH) (FTOH) (CFTOH) (CFTOH)

Table S4 Measured concentrations of PFAS substances in leachate samples analysed at Eurofins Environment Testing Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentration values are given as ng/L. LOQ values are provided.

ID	Ι	II	III	IV	VI	VII	IX	X	LOQ
PFAS acronym	1	11	111	1 V	V I	V 11	іл	Λ	LUQ
PFBA	<20	<20	72	<20	590	580	46	26	0.6
PFPeA	55	130	82	190	580	450	29	39	0.3
PFHxA	87	110	99	190	1400	900	50	61	0.3
PFHpA	19	50	45	40	770	280	34	27	0.3
HPFHpA	<10	<10	0.43	<10	<10	<10	0.79	1.7	0.3
PFOA	66	94	120	72	1800	660	170	120	0.3
PFNA	<10	<10	12	<10	80	46	3.8	32	0.3
PFDA	<10	<10	4.9	<10	38	12	0.89	3.2	0.3
PFUnDA	<10	<10	0.63	<10	<10	<10	0.39	11	0.3
PFDoA	<10	<10	< 0.30	<10	<10	0.48	< 0.30	< 0.30	0.3
PFBS	30	260	100	47	4200	850	7.3	7.1	0.3
PFHxS	22	20	33	11	76	150	26	39	0.3
PFHpS	<10	<10	5.9	<10	<10	<10	9	5.1	0.3
PFOS	29	70	70	15	120	65	36	160	0.2
FOSA	<10	<10	1.6	<10	<10	6	2.6	5.1	0.3
6:2 FTSA	12	48	17	20	320	160	3.2	0.56	0.3
8:2 FTSA	<10	<10	8.1	<10	1300	<10	0.48	0.49	0.3
∑PFAS	320	780	670	590	11000	4200	420	540	0.3

PF-3,7-DMOA, PFTrA, PFTA, PFHxDA, PFDS and 4:2 FTSA were not detected over the quantification limit in the leachate samples. However, it should be noted that, in contrast to the sediment samples (Table S9), MeFOSA, EtFOSA, MeFOSE, EtFOSE, EtFOSAA and MeFOSAA were not analysed in the leachates by Eurofins. Thus, it is unsure if these substances also could have been present in the leachates.

Table S5 Measured concentrations of PFAS substances in leachate samples analysed at ALS Laboratory Group Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentrations are given as ng/L. LOQ values are provided.

PFAS acronym/ID	V	VIII	LOQ
PFHxA	68	95	10
PFOA	98.4	203	10
PFNA	<10	21	10
PFDA	<10	17	10
PFHxS	36	39	10
PFOS	50.6	58.8	10
6:2 FTSA	14	15	10
8:2 FTSA	<10	13	10
∑PFAS	432	592	-

PFUnDA, PFDoA, PFTrA, PFTA, FOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE and 8:2 FTOH were not detected over the LOQ in the leachate samples. In contrast to the leachates analysed at Eurofins (Table S4), the following PFASs were not analysed at ALS: PFBS, PFHpS, PFDS, PFBA, PFPeA, PFHpA, PFHxDA, HPFHpA, PF-3,7-DMOA. Thereby, it is unsure if these substances were present in the leachates analysed at ALS (Table S5).

Variable	PFBS	PFHxS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	6:2 FTSA	8:2 FTSA	HPFHpA	PF-3,7- DMOA	Sum PFAS
PFBS	1.00	0.48	0.21	0.51	0.93	0.91	0.84	0.55	0.51	0.89	0.97	0.89	0.59	0.37	0.83
PFHxS	0.48	1.00	0.66	0.89	0.39	0.42	0.65	0.87	0.83	0.51	0.41	0.24	-0.01	-0.46	0.66
PFOS	0.21	0.66	1.00	0.50	0.09	0.06	0.35	0.56	0.76	0.35	0.01	0.08	-0.22	-0.32	0.48
PFBA	0.51	0.89	0.50	1.00	0.46	0.47	0.78	0.95	0.76	0.48	0.48	0.34	-0.15	-0.47	0.73
PFPeA	0.93	0.39	0.09	0.46	1.00	0.99	0.84	0.50	0.53	0.89	0.92	0.82	0.69	0.38	0.85
PFHxA	0.91	0.42	0.06	0.47	0.99	1.00	0.80	0.48	0.56	0.91	0.91	0.83	0.70	0.37	0.81
PFHpA	0.84	0.65	0.35	0.78	0.84	0.80	1.00	0.85	0.64	0.71	0.82	0.62	0.31	0.00	0.97
PFOA	0.55	0.87	0.56	0.95	0.50	0.48	0.85	1.00	0.74	0.49	0.52	0.32	-0.02	-0.35	0.80
PFNA	0.51	0.83	0.76	0.76	0.53	0.56	0.64	0.74	1.00	0.71	0.37	0.39	0.08	-0.27	0.75
PFDA	0.89	0.51	0.35	0.48	0.89	0.91	0.71	0.49	0.71	1.00	0.82	0.89	0.63	0.42	0.76
6:2 FTSA	0.97	0.41	0.01	0.48	0.92	0.91	0.82	0.52	0.37	0.82	1.00	0.87	0.65	0.42	0.76
8:2 FTSA	0.89	0.24	0.08	0.34	0.82	0.83	0.62	0.32	0.39	0.89	0.87	1.00	0.57	0.58	0.60
HPFHpA	0.59	-0.01	-0.22	-0.15	0.69	0.70	0.31	-0.02	0.08	0.63	0.65	0.57	1.00	0.76	0.32
PF-3,7-DMOA	0.37	-0.46	-0.32	-0.47	0.38	0.37	0.00	-0.35	-0.27	0.42	0.42	0.58	0.76	1.00	-0.02
Sum PFAS	0.83	0.66	0.48	0.73	0.85	0.81	0.97	0.80	0.75	0.76	0.76	0.60	0.32	-0.02	1.00

Table S6 Correlations (r-values) of box-cox transformed leachate PFAS concentrations. Bold values indicate significant (p < 0.05) correlations.

Table S7 PFAS emission data (gram/year) for 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, and are thus excluded from the calculations. n.a. = not analysed. I-S to III-S and VI-S = emissions based on supplementary PFAS results obtained from the landfill operators at the same sampling points as in this study (I-III and VI). Emissions were calculated as average leachate concentrations (Table 1) times yearly leachate volume (Table S2).

ID	Year, month (n)	∑Short- chain PFSAs (< C6) ¹	∑Long- chain PFSAs (≥ C6) ²	∑Short- chain PFCAs (< C7) ³	∑Long- chain PFCAs (≥ C7) ⁴	∑Short- chain PFAS	∑Long- chain PFAS	Short:long- chain ratio	PFOS	PFOA	∑PFAS
Ι	2018, Jan. (1)	5.1	8.6	24	14	29	23	1.3	4.9	11	54
I-S	2017, Apr., June, Aug., Nov. – 2018, May (5)	4.0 ± 1.7	26 ± 35	27 ± 4.0	19 ± 2.8	31 ± 4.4	44 ± 36	0.70 ± 0.94	17 ± 24	12 ± 1.6	79 ± 40
II	2018, April (1)	120	42	111	67	231	108	2.1	32	43	361
II-S	2016, April (1)	148	19	45	28	99	47	2.1	13	17	153
III	2018, April (1)	18	20	46	33	64	53	1.2	13	22	121
III-S	2018, April (1)	20	19	44	33	63	52	1.2	11	22	125
IV	2018, April (1)	3.8	2.1	31	9.1	35	11	3.1	1.2	5.8	48
V*	2018, May (1)	n.a.	2.8	2.2	3.1	2.2	5.9	0.37	1.6	3.1	14
VI	2018, April (1)	194	9.1	119	124	313	133	2.3	5.5	83	509
VI-S	2017, Sept. (1)	31	12	83	71	195	83	2.3	6.1	46	424
VII	2018, May (1)	44	11	100	52	145	63	2.3	3.4	34	218
VIII*	2018, May (1)	n.a.	5.8	5.6	14	5.6	20	0.28	3.5	12	35
IX	2018, May (1)	0.16	1.6	2.8	4.6	2.9	6.2	0.47	0.8	3.7	9.2
Х	2018, June (1)	0.18	5.1	3.1	4.8	3.3	10	0.33	4.0	3.0	13
Median		19	10	38	23	49	46	1.1	5.2	14	100
$Mean \pm SD$		49 ± 67	13 ± 11	46 ± 41	34 ± 34	87 ± 98	47 ± 39	1.8 ± 0.9	8.3 ± 8.5	23 ± 22	155 ± 164
Min-max	2016-2018 (18)	0.16 - 194	1.6 - 42	2.2 - 119	3.1 - 124	2.2 - 313	5.9 - 133	0.28 - 3.1	0.79 - 32	3.0 - 83	9.2 - 509
Most abundant	-	(PFBS)	PFOS	PFHxA	PFOA	PFBS	PFOA	-	-	-	PFBS

¹PFBS; ²PFHxS, PFHpS and PFOS; ³PFBA, PFPeA and PFHxA; ⁴PFHpA, HPFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoA and PFDoA

* 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

Table S8 Estimated national PFAS release (kg/year), calculated based on the overall concentration data given in Table 1 and the estimated total volume of landfill leachate emitted from Norwegian landfills¹ (1.0x10⁷ m³). SD = standard deviation.

Calculation based on	∑Short-chain PFSA (< C6)	∑Long- chain PFSA (≥ C6)	∑Short-chain PFCA (< C7)	∑Long-chain PFCA (≥ C7)	∑Short-chain PFAS	∑Long-chain PFAS	PFOS	PFOA	∑PFAS
Mean \pm SD	5.5 ± 12	1.1 ± 0.6	5.1 ± 7.6	4.2 ± 6.9	9.8 ± 18	5.3 ± 7.3	0.68 ± 0.38	2.8 ± 4.6	17 ± 29
Median	1.1	1.0	2.5	1.9	3.6	2.9	0.65	1.2	6.3
Min-max	0.071 - 42	0.26 - 2.2	0.68 - 26	0.85 - 27	0.68 - 68	1.4 - 29	0.15 - 1.6	0.66 - 18	3.2 - 110

Table S9 Measured concentrations of PFAS substances in sediment samples analysed at Eurofins Environment Testing Norway AS where there was at least one measurement above the limit of quantification (LOQ). All concentration values are given as $\mu g/kg$ dry matter, except for dry matter content (DM) which is reported as %. LOQ values are provided.

ID	I	IV	V	VI	VII	IX	X-1*	X-2*	LOQ
PFAS acronym	1	17	•	• 1	V 11	17	71-1	X-Z	LUQ
PFBA	< 0.21	< 0.20	0.49	< 0.20	1.7	< 0.91	< 0.27	< 0.20	0.2
PFPeA	< 0.21	< 0.20	< 0.42	< 0.20	1.2	< 0.91	< 0.27	< 0.20	0.2
PFHxA	< 0.21	0.25	< 0.42	< 0.20	2.0	< 0.91	< 0.27	< 0.20	0.2
PFHpA	< 0.21	< 0.20	< 0.42	< 0.20	0.86	< 0.91	< 0.27	< 0.20	0.2
PFOA	1.6	1.2	1.3	0.21	3.6	1.8	0.54	0.23	0.1
PFNA	1.1	0.54	< 0.42	< 0.20	0.53	< 0.91	0.76	0.28	0.2
PFDA	2.7	1.0	0.56	0.48	1.3	< 0.91	0.49	< 0.20	0.2
PFUnDA	0.72	0.32	< 0.42	< 0.20	0.57	< 0.91	16	0.89	0.2
PFDoA	0.52	0.32	< 0.42	< 0.20	0.65	< 0.91	< 0.27	< 0.20	0.2
PFTrA	< 0.21	< 0.20	< 0.42	< 0.20	< 0.25	< 0.91	2.2	< 0.20	0.2
PFBS	< 0.21	< 0.20	< 0.42	< 0.20	3.3	< 0.91	< 0.27	< 0.20	0.2
PFHxS	0.31	< 0.20	< 0.42	< 0.20	0.41	< 0.91	< 0.27	< 0.20	0.2
PFOS	25	11	14	1.3	9.0	15	24	3.9	0.1
PFDS	0.31	< 0.20	< 0.42	< 0.20	< 0.25	< 0.91	0.45	< 0.20	0.2
FOSA	2.6	1.1	1.4	< 0.20	1.2	1.8	1.7	< 0.20	0.2
FOSAA	0.66	0.29	< 0.42	< 0.20	0.65	< 0.91	0.76	< 0.20	0.2
MeFOSAA	5.2	5.7	< 0.42	0.22	31	1.2	0.80	< 0.20	0.2
EtFOSAA	24	13	12	0.52	41	6.7	6.7	0.59	0.2
MeFOSE	0.76	1.8	< 0.42	< 0.20	6.5	< 0.91	< 0.27	< 0.20	0.2
EtFOSE	2.4	3.5	< 0.42	< 0.20	4.9	< 0.91	< 0.27	< 0.20	0.2
6:2 FTSA	0.52	0.54	< 0.42	< 0.20	1.1	< 0.91	< 0.27	< 0.20	0.2
8:2 FTSA	2.3	3.1	< 0.42	2.8	2.4	< 0.91	< 0.27	< 0.20	0.2
Sum PFAS	72	46	37	8.8	120	40	42	8.5	0.2
DM	29	31.4	14.3	75.6	24.5	6.6	22.4	56.2	0.2

*At landfill X, two sediment samples were analysed: X-1: from the sedimentation pond and X-2: downstream the sedimentation pond.

HPFHpA, PF-3,7-DMOA, PFTA, PFHxDA, PFHpS, MeFOSA, EtFOSA and 4:2 FTSA were not detected over the LOQ in the sediment samples.

Table S10 Log $Q_{sed/leachate}$ values based on PFOA-, PFOS- and \sum PFAS-concentrations measured in leachate and sediment from landfill I, IV, V-VII, IX and X.

Landfill ID	PFOA	PFOS	∑PFAS
Ι	1.38	2.94	2.35
IV	1.22	2.87	1.89
V	1.12	2.44	1.93
VI	-0.93	1.03	-0.10
VII	0.74	2.14	1.46
IX	1.02	2.62	1.98
Х	0.65	2.18	1.89
Median	1.0	2.4	1.9
Mean ± SD	0.74 ± 0.78	2.3 ± 0.64	1.6 ± 0.80
Min-max	-0.93 - 1.38	1.0 - 2.9	-0.10 - 2.4

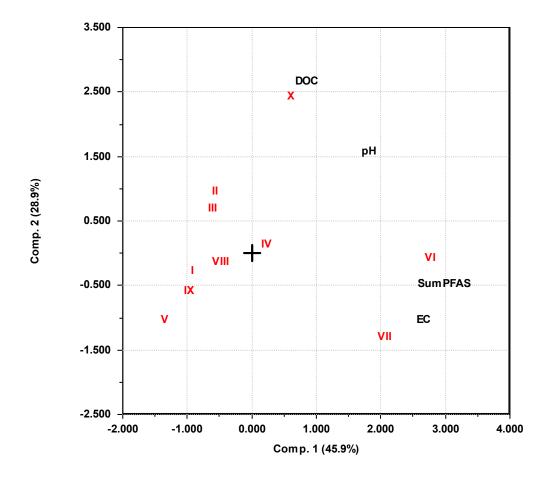


Figure S1 PCA biplot that combines loadings (potential correlations between different variables, i.e. the parameters \sum_{28} PFAS (ng/L), DOC (mg/L), pH and EC (μ S/cm)) and scores (potential correlations between different objects/samples: leachates from landfill I-X).

Table S11 Correlations (r-values) of box-cox transformed leachate PFAS concentrations with electrical conductivity (EC). Bold values indicate significant (p < 0.05) correlations.

Variable	PFBS	PFHxS	PFOS	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	6:2 FTSA	8:2 FTSA	HPFHpA	PR- 3.7- DMOA	Sum PFAS
EC (µs/cm)	0.67	0.74	0.22	0.72	0.78	0.79	0.82	0.75	0.68	0.65	0.68	0.40	0.47	-0.12	0.82

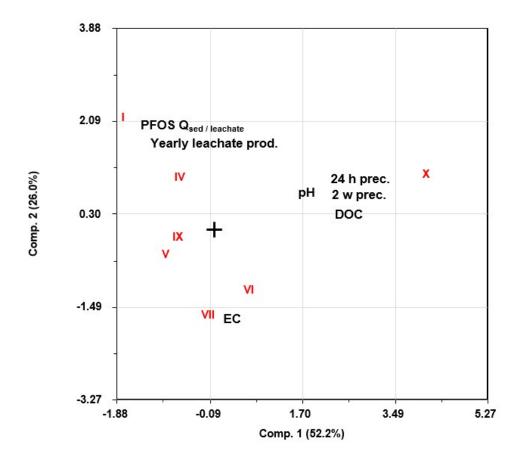


Figure S2 PCA biplot that combines loadings (potential correlations between different variables, i.e. the parameters PFOS $Q_{sed/leachate}$ (L/kg), DOC (mg/L), pH, EC (μ S/cm), yearly leachate production volume (m³/year), 24 hour and 2 week precipitation (mm)) and scores (potential correlations between different objects/samples, i.e. landfills were both sediment and leachate was analysed).

There were no significant correlations between leachate DOC and \sum_{28} PFAS or $Q_{sed/leachate}$ values for the landfill leachates investigated in this study. It is important to pinpoint that the DOC values used for correlation analysis were not measured in the sampled leachates, but were based on earlier results at the same sampling sites, provided by the landfill operators. As such, it cannot be ruled out that significant correlation could be found if the DOC levels had been measured in the samples in this study. In addition, it is possible that significant correlations between $Q_{sed/leachate}$ values and organic carbon content had been seen if the organic carbon levels in sediment had been measured. There were no significant correlations among PFAS with 24 h and 2 week precipitation. On the contrary, Benskin et al.² found significant correlations between log transformed concentrations of PFBA, PFPA and PFHxA with decreasing 24-h precipitation.

References to the Supplementary Information

- 1. H. P. H. Arp and G. Okkenhaug, *Miljøgifter i sigevann fra avfallsdeponier i Norge. Data fra perioden 2006-2010*, Report NGI report 20110546-00-5-R, Norwegian Geotechnical Institute, Oslo, <u>http://www.miljodirektoratet.no/old/klif/publikasjoner/2978/ta2978.pdf</u>, 2012.
- 2. J. P. Benskin, B. Li, M. G. Ikonomou, J. R. Grace and L. Y. Li, Per-and polyfluoroalkyl substances in landfill leachate: patterns, time trends, and sources, *Environmental science & technology*, 2012, **46**, 11532-11540.