PFAS Emissions From Landfill Sources

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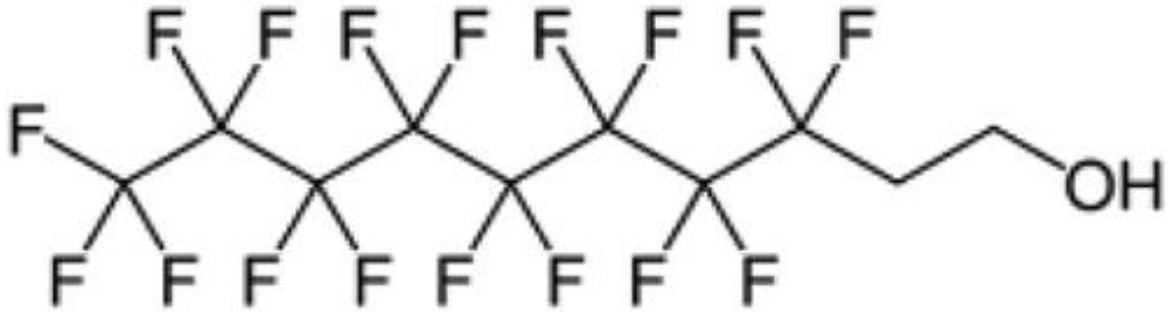
Outline

- Background
- PFAS Emission Sources at Landfills
- PFAS Impacts from Landfills



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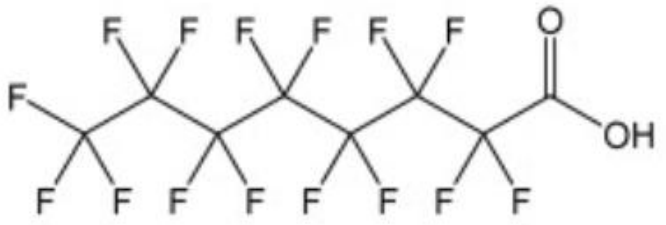
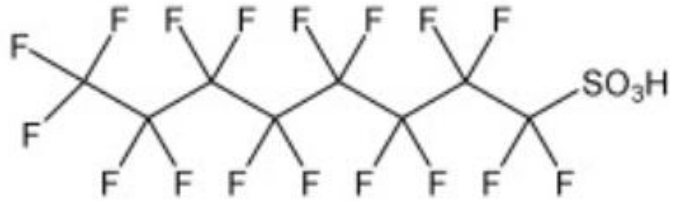
PFAS Characteristics

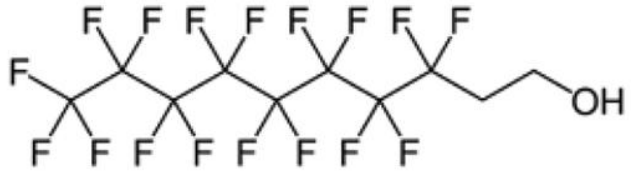
- PFAS subject to drinking water regulations have low volatility
  - Perfluorooctanoic acid (PFOA)
  - Perfluorooctane sulfonic acid (PFOS)
- Volatile PFAS also exist
  - Fluorotelomer alcohols (FTOHs)
  - Perfluorooctanesulfonamides (FOSAs)
  - Perfluorooctanesulfonamidoethanol (FOSEs)



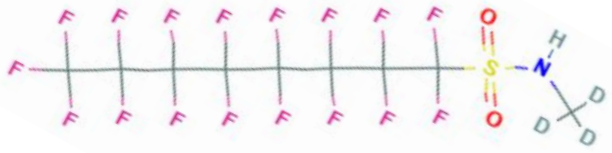
Perfluorooctanoic acid (PFOA)



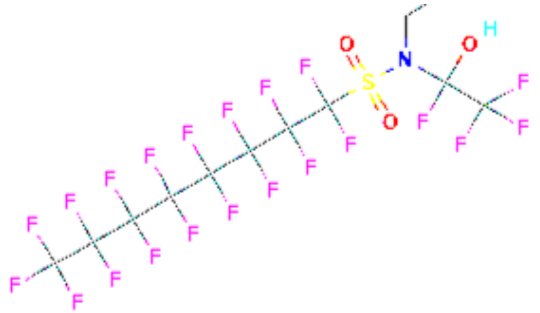
Perfluorooctane sulfonate (PFOS)



8:2-Fluorotelomer alcohol (8:2-FTOH)



n-Methylperfluorooctanesulfonamide N-MeFOSA)



n-Ethyl Perfluorooctanesulfonamidoethanol (N-EtFOSE)

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Barlaz *et al.* Study of PFAS in Landfill Gas 2023 Research Summary (USEPA Funded)

"We estimate that 21,862 million m^3 (mM^3) of LFG are treated by a flare or engine and 22,978 Mm^3 LFG are released as fugitive emissions. While preliminary, at an average total PFAS concentration of 5×10^7 pg/m^3, this would result in **1150 kg/yr of PFAS in fugitive LFG**, assuming 100% PFAS destruction efficiency in a flare or engine and 0% PFAS attenuation in soil. The volumes of LFG that are released by fugitive emissions and treated in a flare or engine are within 5%. Thus, estimated emissions will increase if PFAS destruction in a flare or engine is incomplete. To put this estimate into some context, we estimated that **660 kg/yr of PFAS are released to wastewater treatment plants** in 2017."

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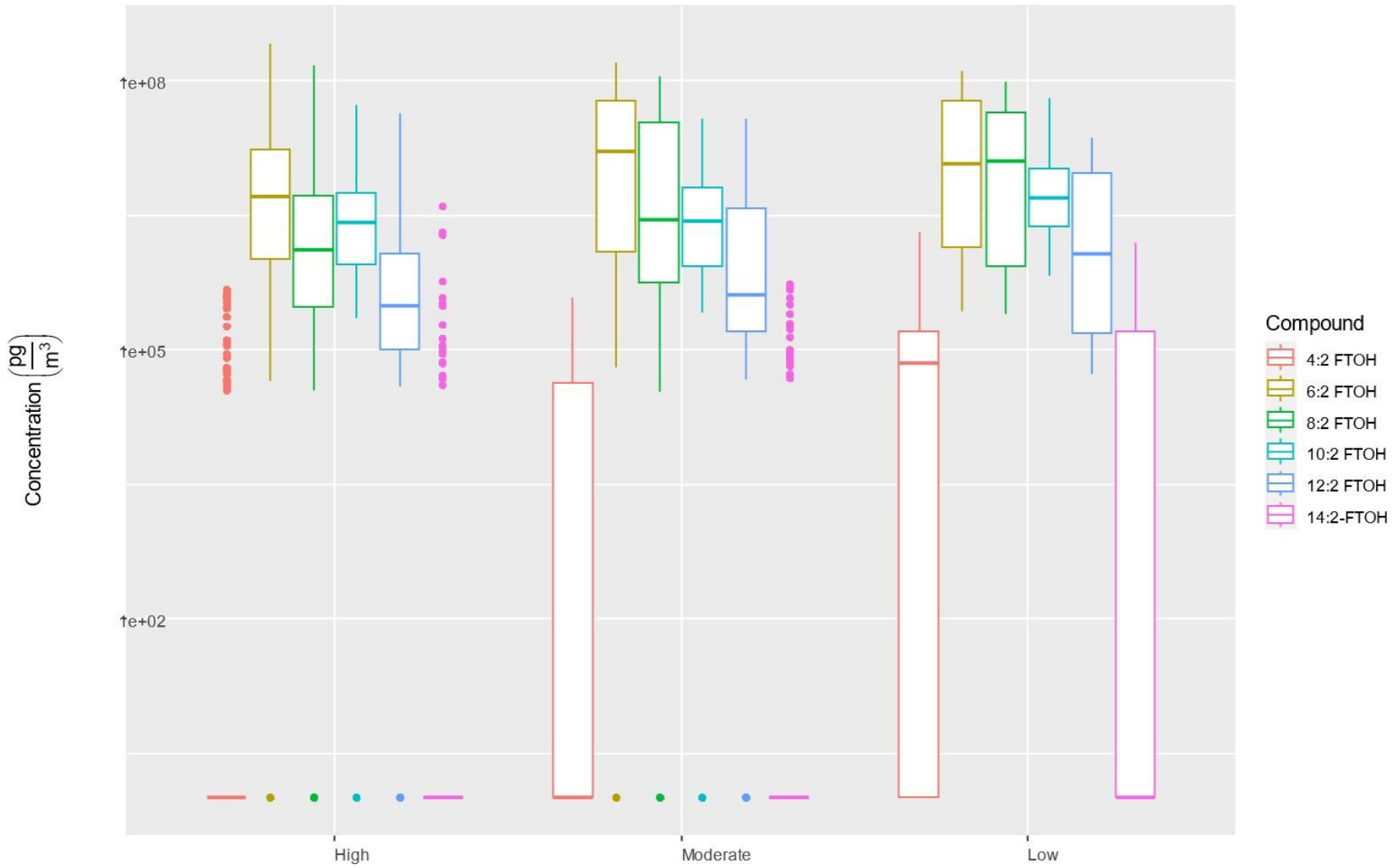
https://cfpub.epa.gov/ncer_abstracts/index.cfm/fuseaction/display.abstractDetail/abstract_id/10990/report/2023

PFAS in Landfill Gas

- Source: 2023 Progress Report of NC State Study (Barlaz *et al*.)
  - Volatile fluorotelomer alcohols (FTOHs) dominate
  - Data from 28 landfills
  - 10X higher than previous report
- Average concentration = 50,000,000 pg/m^3 = 50 µg/m^3
  - 10X higher than in 2022 Progress Report
  - Perspective: Compare to benzene at 6,000 µg/m^3
- Fugitive PFAS flux estimate = 1,150 kg/yr
  - Compare to leachate estimate of 563 to 638 kg/yr in leachate (2017 Lang *et al.* study)
- Variability important
  - More than order of magnitude difference between 25th and 75th percentile concentrations



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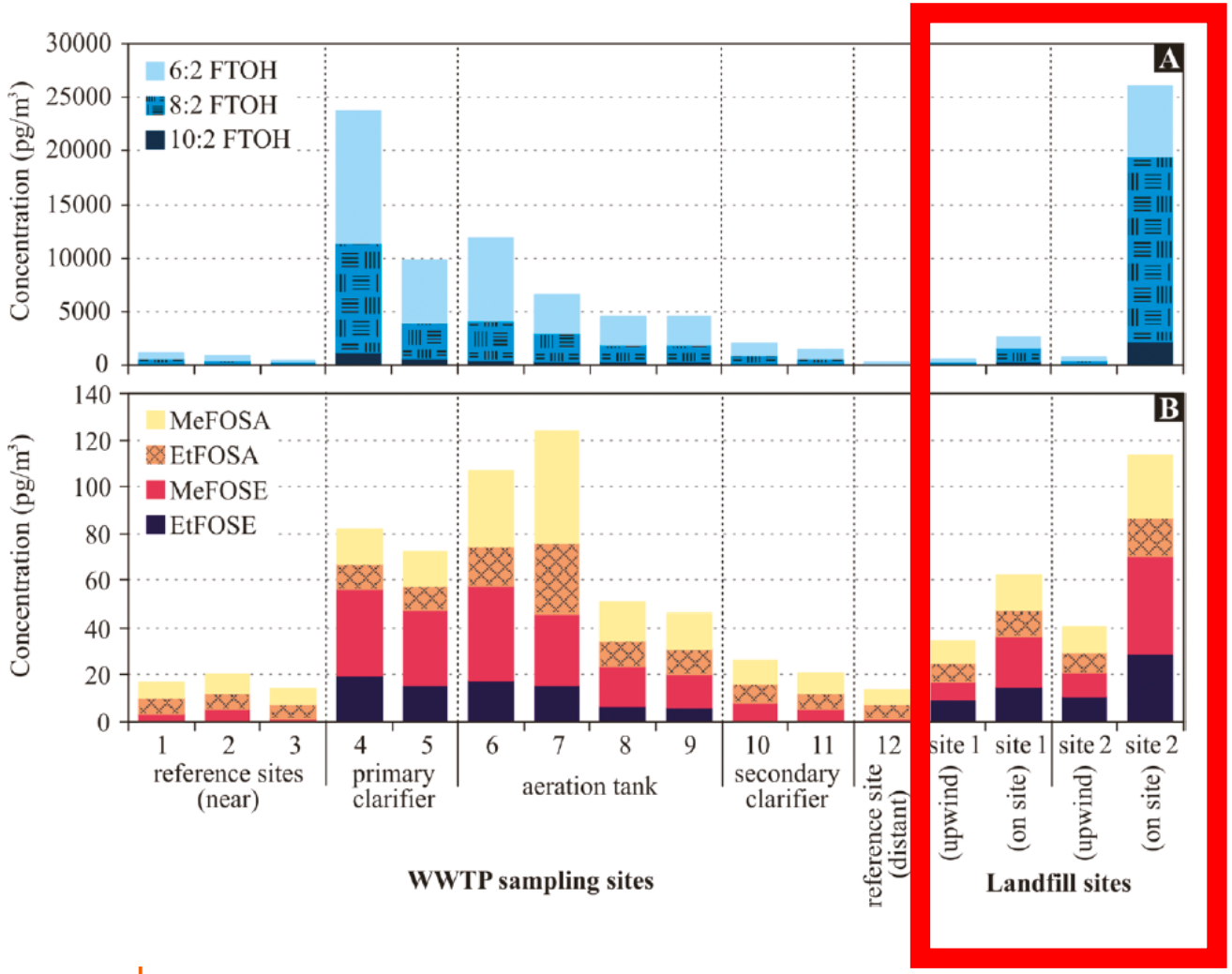
Barlaz et al. (2023): https://cfpub.epa.gov/ncer_abstracts/index.cfm/fuseaction/display.abstractDetail/abstract_id/10990/report/2023
Lang et al. (2017): https://pubs.acs.org/doi/10.1021/acs.est.6b05005

PFAS Measurements in Air Near Landfills (Canada)



- Outdoor air measurements (not raw landfill gas)
- Paired downwind v. upwind samples imply landfill emissions
- Excess (down v. up) FTOHs
  - 2,000 – 25,000 pg/m^3
- Excess Me/Et FOSA/FOSE
  - 20 – 70 pg/m^3
- Elevated levels of FTOHs also seen in other landfill studies conducted around 2010
- WWTPs are similar FTOH sources

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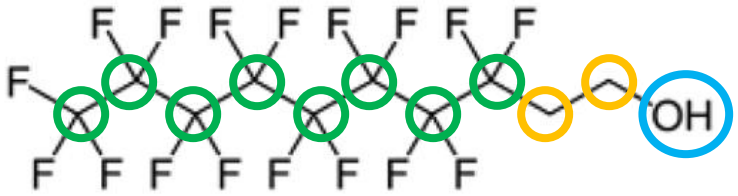
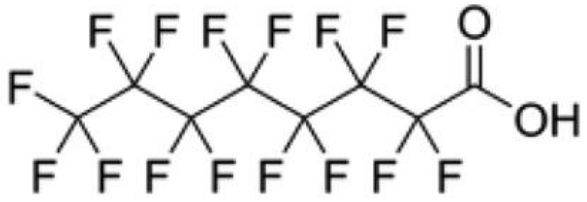
*Ahrens et al. (2011), Environ. Sci. Technol. **45:**8098–8105*

Structure and Properties of 8:2 FTOH

8:2 FTOH



8:2 fluorotelomer alcohol
(8:2 FTOH)



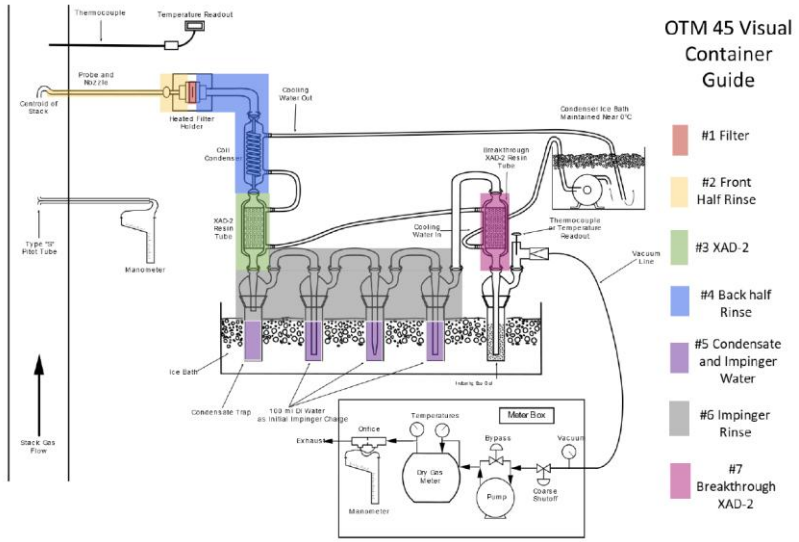
perfluorooctanic acid
(PFOA, C8)

- Experimental Henry's Law Constants (air/water) per ITRC for 8:2 FTOH reported at 0.0037 atm-m^3/mol and 0.05 atm-m^3/mol – compare to benzene at 0.0055 atm-m^3/mol and ethanol at ~0.000005 atm-m^3/mol
- Solubility appears limited – median about 0.2 mg/L (but reported values vary considerably)
- Log K_{oc} about 4 – some stickiness to solids
- **Overall: 8:2-FTOH is fairly volatile**
- C—H bonds allow 8:2 FTOH to be degraded; one of the stable breakdown products is PFOA
- Robust toxicity data for FTOHs have not been developed yet

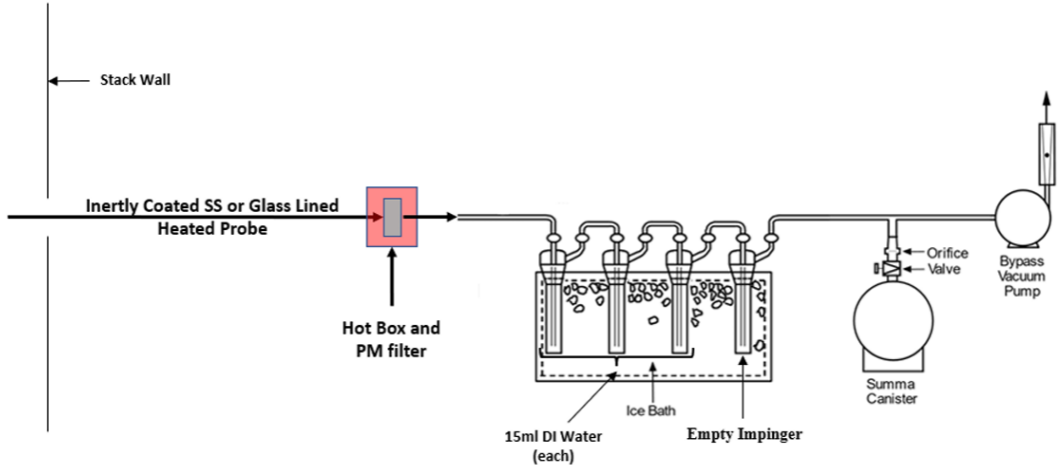
ITRC: https://pfas-1.itrcweb.org/wp-content/uploads/2022/01/PhysChemProp_Table_July2023-FINAL.xlsx

EPA Methods to Measure PFAS Emissions

- Stack Emissions Testing Methods – Current official EPA Methods for Stack Testing
  - OTM-45 – Focus on long-chain, polar compounds – includes FOSA and FOSE compounds
  - OTM-50 – Focus on short-chain products of incomplete combustion



<= OTM-45 / OTM-50 =>



- FTOHs not included in OTM-45 or OTM-50 *as published* (maybe OTM-55?)
- No official ambient air sampling methods – research and lab methods exist

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https://www.epa.gov/sites/default/files/2021-01/documents/otm_45_semivolatile_pfas_1-13-21.pdf
https://www.epa.gov/system/files/documents/2024-01/otm-50-release-1_0.pdf

Analyte Lists – No Overlap, No FTOHs

OTM-45 50 Analytes

PFAS Group	PFAS	CAS Registry Number
Perfluoroalkylcarboxylic acids (PFCAs)	PFBA	375-22-4
	PFPeA	2706-90-3
	PFHxA	307-24-4
	PFHpA	375-85-9
	PFOA	335-67-1
	PFNA	375-95-1
	PFDA	335-76-2
	PFUnDA	2058-94-8
	PFDoA	307-55-1
	PFTrDA	72629-94-8
	PFTeDA	376-06-7
	PFHxDA	67905-19-5
	PFODA	16517-11-6
Perfluorinated sulfonic acids (PFSAs)	PFBS	375-73-5
	PFPeS	2706-91-4
	PFHxS	355-46-4
	PFHpS	375-92-8
	PFOS	1763-23-1
	PFNS	68259-12-1
	PFDS	335-77-3
	PFDoS	79780-39-5
Perfluorinated sulfonamides (FOSAs)	FOSA	754-91-6
	MeFOSA	31506-32-8
	EtFOSA	4151-50-2
Perfluorinated sulfonamide ethanols (FOSEs)	N-MeFOSE	24448-09-7
	N-EtFOSE	1691-99-2
Perfluorinated sulfonamidoacetic acids (FOSAAs)	MeFOSAA	2355-31-9
	EtFOSAA	2991-50-6

PFAS Group	PFAS	CAS Registry Number
Fluorotelomer sulfonates (FTS)	4:2 FTS	757124-72-4
	6:2 FTS	27619-97-2
	8:2 FTS	39108-34-4
	10:2 FTS	120226-60-0
Fluorinated Replacement Chemicals	ADONA	919005-14-4
	GenX	13252-13-6
	9Cl-PF3ONS	756426-58-1
	11Cl-PF3OUdS	763051-92-9 83329-89-9
Additional Targets	NFDHA	151772-58-6
	PFEESA	113507-82-7
	PFDoS	1260224-54-1
	PFMBA	863090-89-5
	PFMPA	377-73-1
	PFecHS	67584-42-3
	8:2 FTUCA	70887-84-2
	10:2 FDEA	53826-13-4
	8:2 FTA	27854-31-5
	6:2 FHUEA	70887-88-6
	6:2 FTCA	53826-12-3
	3:3 FTCA	356-02-5
	5:3 FTCA	914637-49-3
	7:3 FTCA	812-70-4

OTM-50 30 Analytes

Table OTM-50-1. Volatile Fluorinated Compound Target List

Compound Name	CAS #	Chemical Formula
Carbon tetrafluoride	75-73-0	CF_4
Hexafluoroethane (FC-116)	76-16-4	C_2F_6
Tetrafluoroethene	116-14-3	C_2F_4
Trifluoromethane (HFC-23)	75-46-7	CHF_3
Octafluoropropane	76-19-7	C_3F_8
Difluoromethane (HFC-32)	75-10-5	CH_2F_2
Fluoromethane (HFC-41)	593-53-3	CH_3F
Pentafluoroethane (HFC-125)	354-33-6	C_2HF_5
Hexafluoropropene	116-15-4	C_3F_6
Hexafluoropropene oxide (HFPO)	428-59-1	C_3F_6O
Decafluorobutane	355-25-9	C_4F_{10}
Dodecafluoropentane	678-26-2	C_5F_{12}
Tetradecafluorohexane	355-42-0	C_6F_{14}
1H-Perfluoropentane	375-61-1	C_5HF_{11}
Hexadecafluoroheptane	335-57-9	C_7F_{16}
Heptafluoropropyl-1,2,2,2-tetrafluoroethyl ether (E1)	3330-15-2	$C_5HF_{11}O$
1H-Perfluorohexane	355-37-3	C_6HF_{13}
1H-Perfluoroheptane	375-83-7	C_7HF_{15}
2H-Perfluoro-5-methyl-3,6-dioxanonane (E2)	3330-14-1	$C_8HF_{17}O_2$
1H-Perfluorooctane	335-65-9	C_8HF_{17}
Octadecafluorooctane	307-34-6	C_8F_{18}
1H-Nonafluorobutane	375-17-7	C_4HF_9
1H-Heptafluoropropane	2252-84-8	C_3HF_7
1,1,1,2-Tetrafluoroethane (HFC-134a)	811-97-2	$C_2H_2F_4$
1,1,1-Trifluoroethane (HFC-143a)	420-46-2	$C_2H_3F_3$
Chlorodifluoromethane (HCFC-22)	75-45-6	$CHClF_2$
Chlorotrifluoromethane (CFC-13)	75-72-9	CF_3Cl
Octafluorocyclobutane (FC-C318)	115-25-3	C_4F_8
Octafluorocyclopentene (FC-C1418)	559-40-0	C_5F_8
Trichloromonofluoromethane (CFC-11)	75-69-4	CCl_3F

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https://www.epa.gov/sites/default/files/2021-01/documents/otm_45_semivolatile_pfas_1-13-21.pdf
https://www.epa.gov/system/files/documents/2024-01/otm-50-release-1_0.pdf

Potential PFAS Emission Sources at Landfills

- Fugitive Landfill Gas (LFG) Emissions
  - Barlaz et al. study reports no observed attenuation in soils
- LFG Combustion Devices
  - Energy Recovery Engines and Turbines
  - Flares
- Other Potential Sources
  - Leachate Evaporation Systems
  - Leachate Tanks
  - Stormwater Impoundments
  - Passive Landfill Gas Vents and Vent Flares
  - Leachate Treatment Systems

PFAS Behavior in Solid Waste Combustion Treatment at Landfills

- Study funded by EREF Environmental Research & Education Foundation
- UVM/PI A. Badireddy, collaborators F. De la Cruz, S. Zemba, W. Fritz
- Data collection complete, final report to be issued soon
- Study goal/scope
  - Measure PFAS emissions, destruction, and products of incomplete combustion in common landfill gas treatment (combustion) devices:
    - Internal combustion engines (energy recovery)
    - Flares
- Approach
  - Measure PFAS in landfill gas feeds
  - Test stack emissions in combustion exhaust

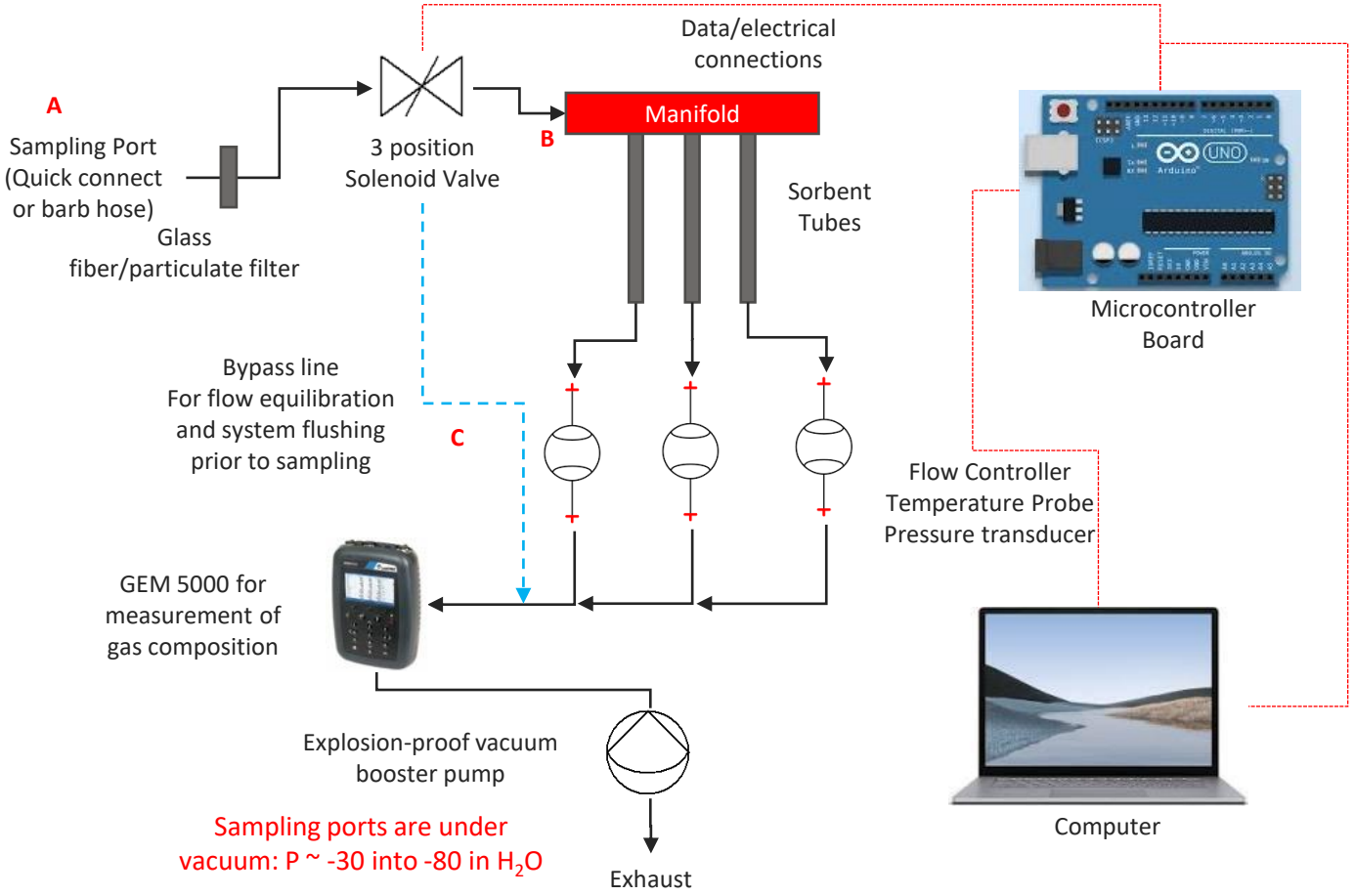
Study Description: https://erefdn.org/grantee/pfas-behavior-in-solid-waste-combustion-treatment-at-landfills/

Stack Emissions (Combustion Exhaust) Sampling

- Landfill 1: Energy Recovery (Internal Combustion) Engine
  - OTM 45
  - Value
    - Quantify feed rates of principal PFAS in feed to engine
    - Examine potential conversion of FTOHs to terminal non-volatile PFAS such as Perfluorooctanoic acid (PFOA)
- Landfill 2: Enclosed Flare
  - OTM 45 with dual extractions for FTOHs
  - OTM 50 for products of incomplete combustion
  - Value
    - Measure overall PFAS destruction and potential formation of PICs

Landfill Gas Sampling System (courtesy . De la Cruz)

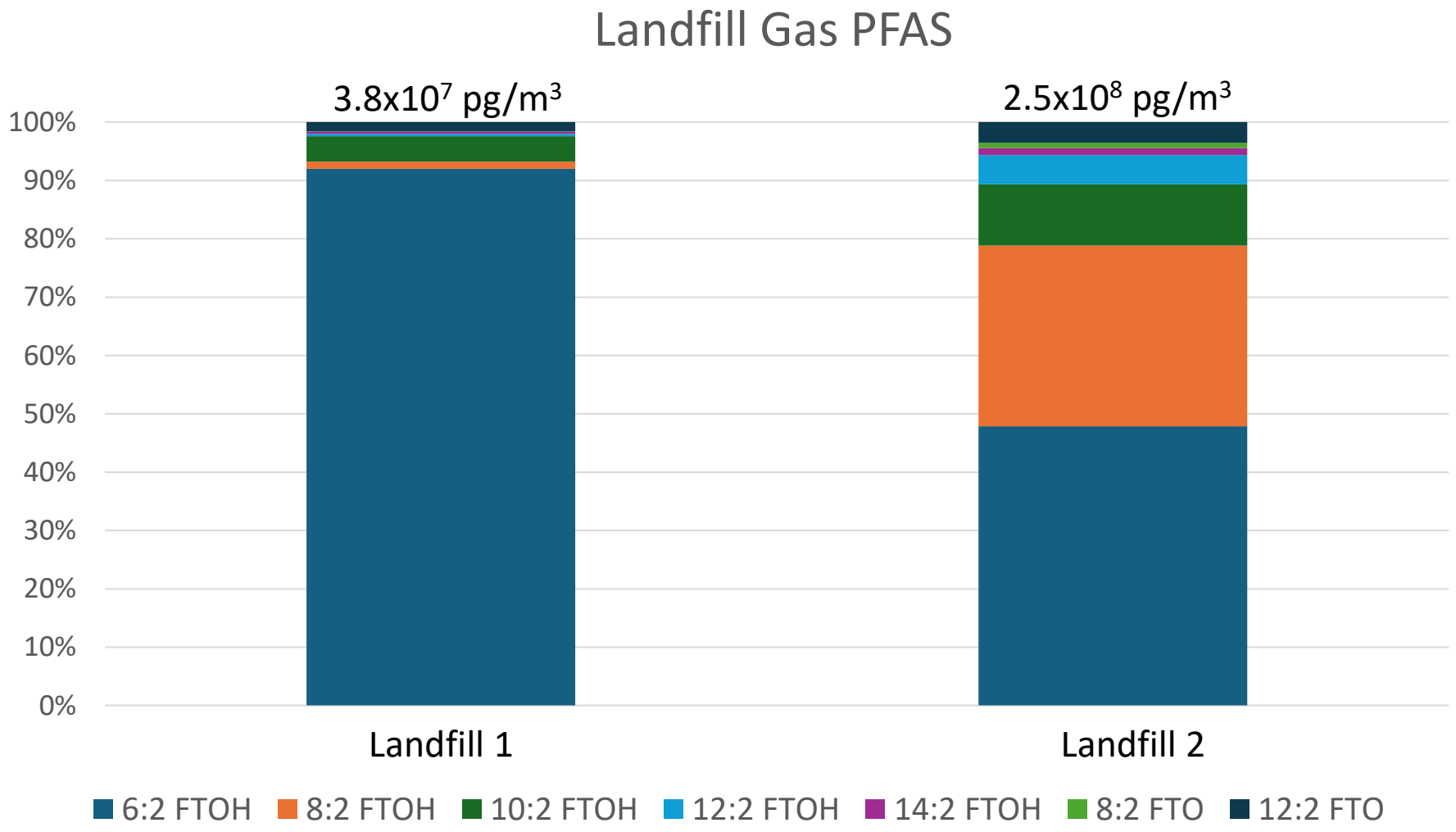
Main Header



Optimization:
How much volume to sample?
What flow rate?

NC STATE UNIVERSITY
Oregon State University
PURDUE UNIVERSITY
UNF UNIVERSITY of NORTH FLORIDA

Landfill Gas Testing
Preliminary Results (courtesy of A. Badireddy)



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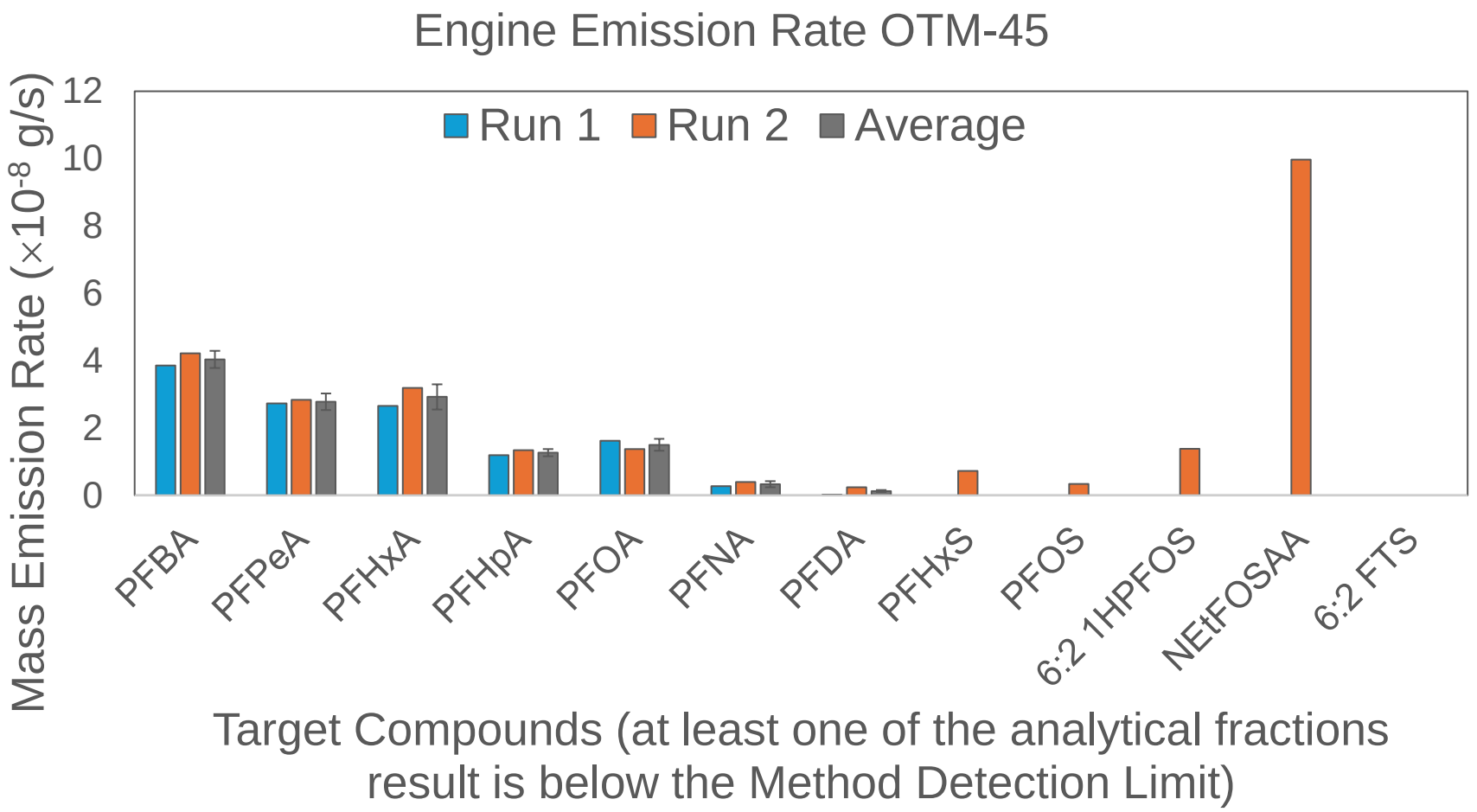
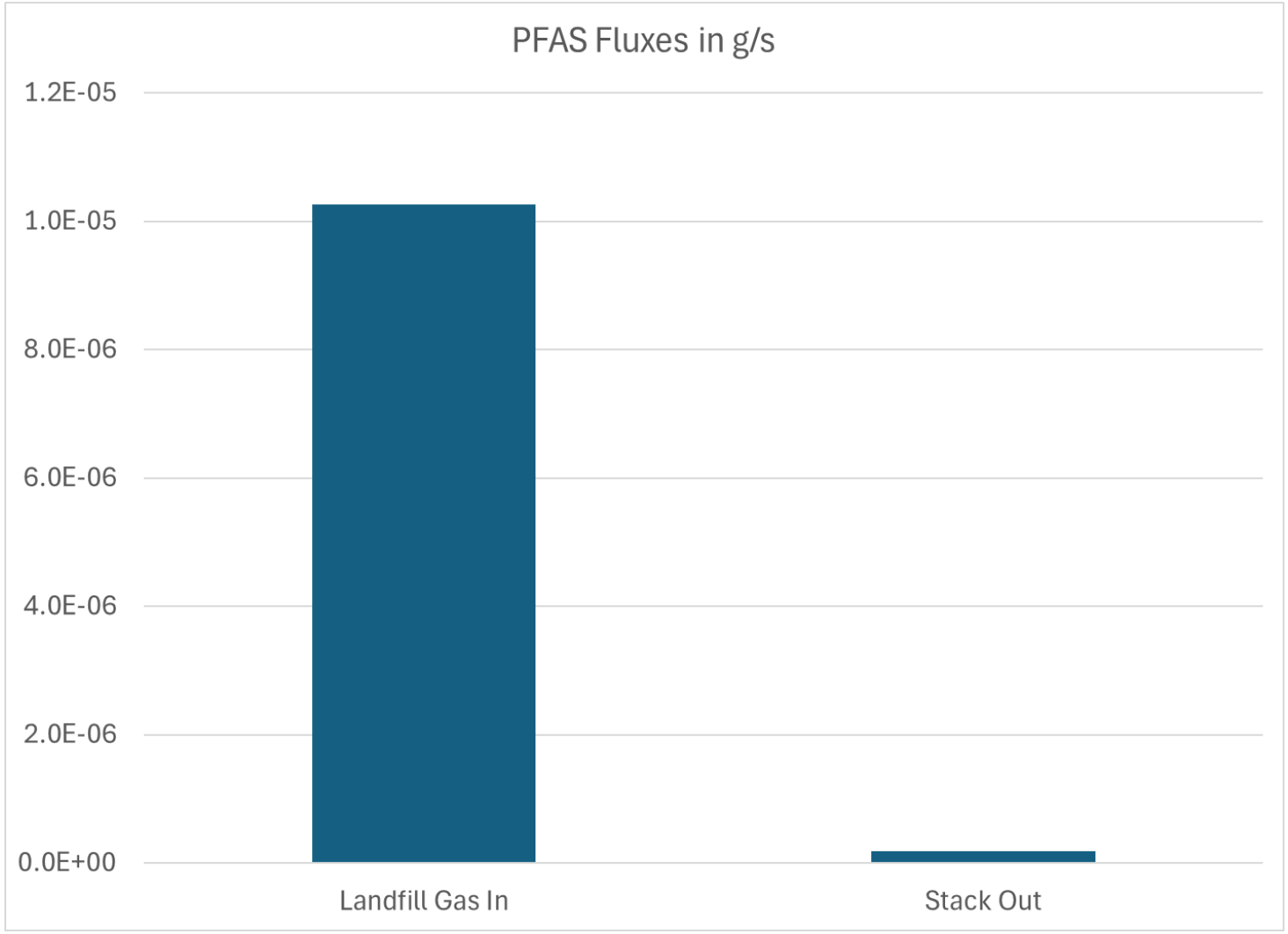
Landfill 1: PFAS Mass Emission Rates for IC Engine

Preliminary Results (courtesy of A. Badireddy)



Landfill 1: PFAS Mass Emission Rates for IC Engine

Preliminary Results (courtesy of A. Badireddy) – Detections Only



- 98% less PFAS in stack compared to gas feed
- FTOHs in stack not measured
- Select compound comparisons

Compound	Emission Rate (g/s)
6:2-FTOH	9.4E-06
PFHxA	2.9E-08
8:2-FTOH	1.2E-07
PFOA	1.5E-08

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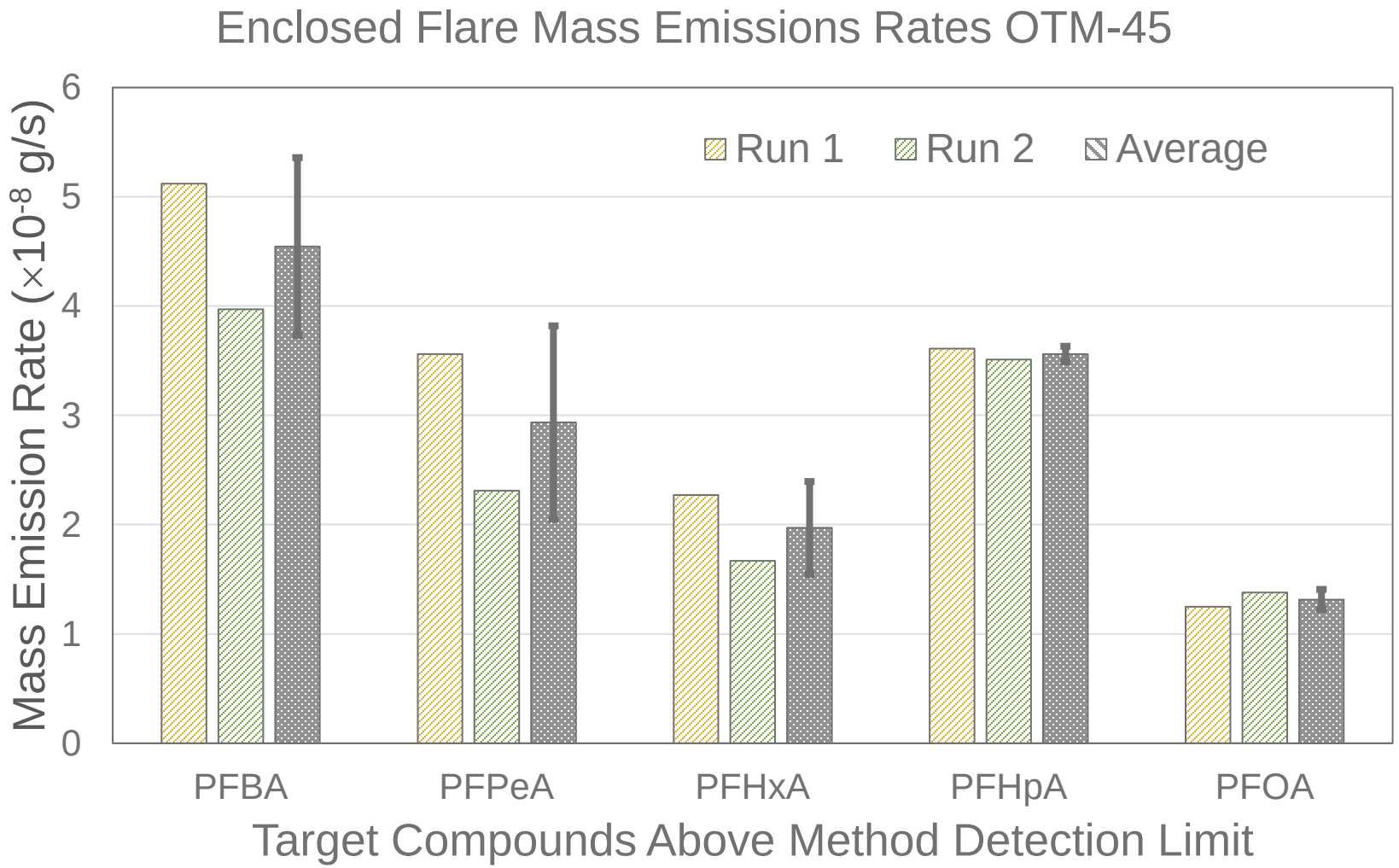
Landfill 2: PFAS Mass Emission Rates from Flare

Preliminary Results (courtesy of A. Badireddy)



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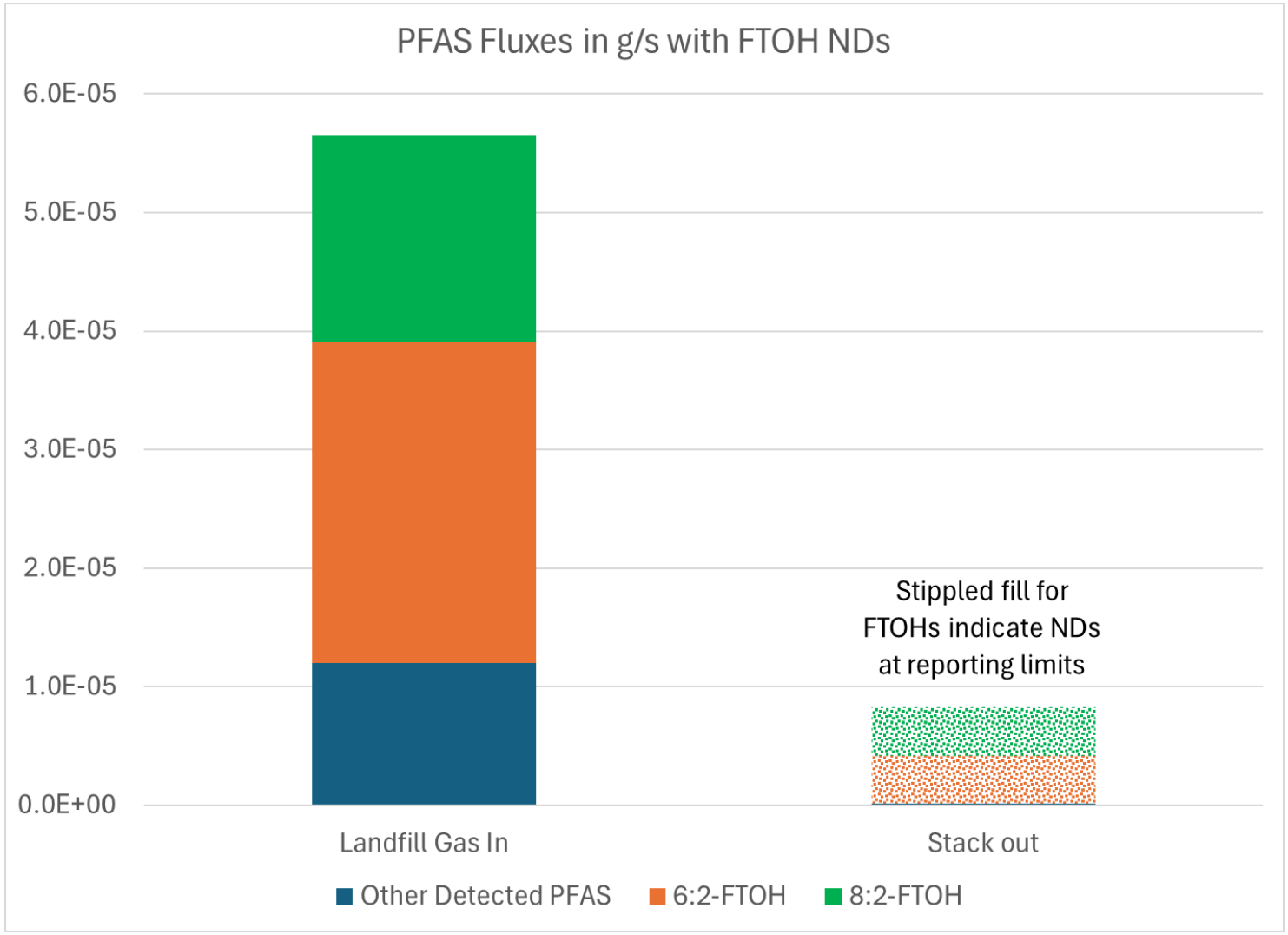
Landfill 2: PFAS Mass Emission Rates for Flare

Preliminary Results (courtesy of A. Badireddy) – Detections Only



- 99% less PFAS in stack compared to gas feed
- FTOHs in stack measured but not detected
  - >85% reduction in 6:2-FTOH
  - >77% reduction in 8:2-FTOH
- No detects in OTM 50
- Select compound comparisons

Compound	Emission Rate (g/s)
6:2-FTOH	8.2E-06
PFHxA	2.0E-08
8:2-FTOH	5.3E-06
PFOA	1.3E-08

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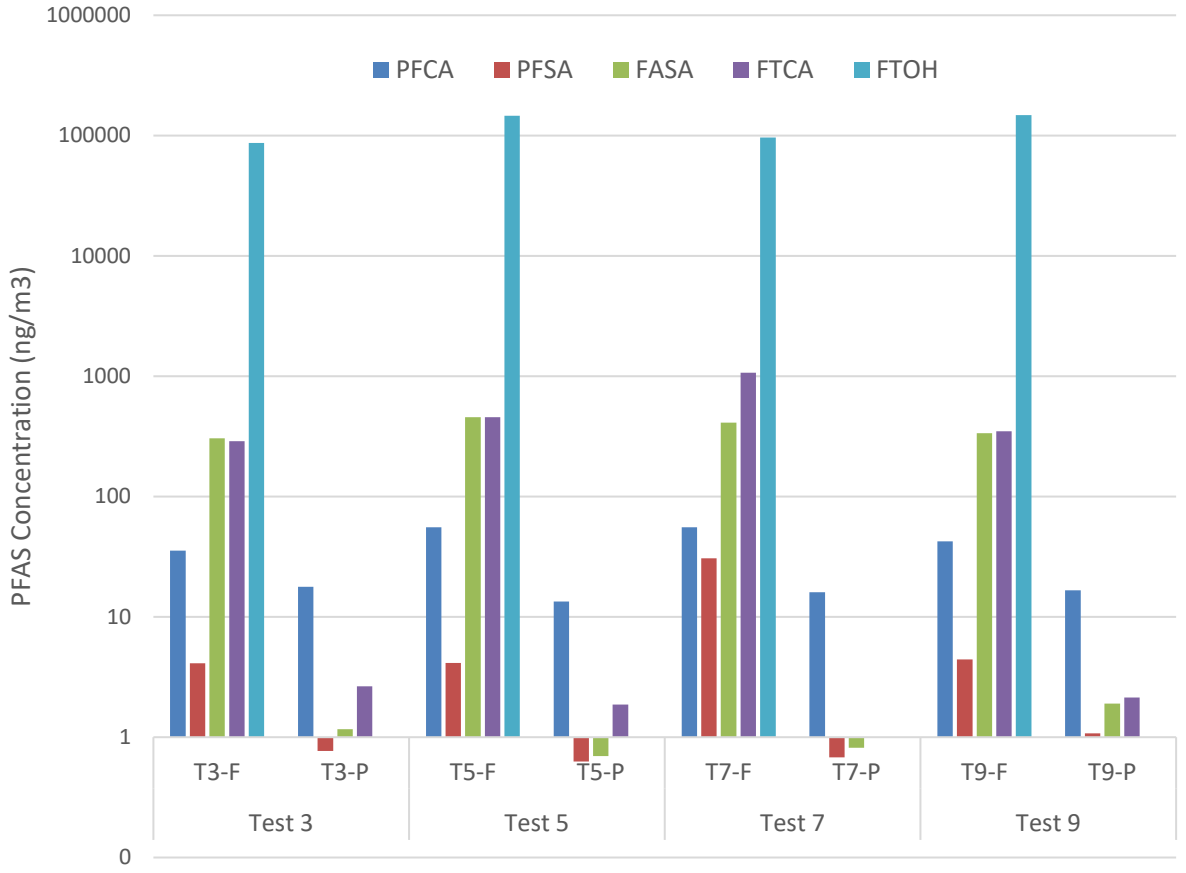
Observations re: Landfill Gas Combustion Testing

- Low levels of stable PFAS detected in both engine and flare emissions
  - Mostly carboxylic acids
  - Might reflect conversion of precursors
  - Might be the result of aerosols in landfill gas
- OTM 50 and FTOH methods are less sensitive than OTM 45
  - Catch weight reporting limits ~1,000 times higher
  - Limitations on destruction efficiency evaluation
- Sampling artifacts might be relevant
  - NEtFOSAA & 6:2-FTS detected in engine test
  - HPFO-DA detected in flare test

PFAS Emissions from Leachate Treatment Courtesy from L. Zeng – preliminary data

- Foam fractionation (FF) is a leading candidate technology to remove PFAS from landfill leachate
- FF systems bubble air through leachate and typically have air effluent
- Regulatory agencies are requesting characterization of emissions
- Some preliminary information from air measurements at a pilot-scale treatment study
  - Published EPA OTM 45 and OTM 50 not useful (only vent-scale)
  - Concentrated air effluent is odorous due to H_2S and NH_3
  - Volatile organic compounds (VOCs) consistent with leachate
  - PFAS emissions are measurable
    - Most prevalent compounds are FTOHs, concentrations higher than in typical landfill gas
    - Some nonpolar PFAS (e.g., PFOA) measurable, possibly the result of aerosols
  - Activated carbon effective at capturing PFAS (including FTOHs)



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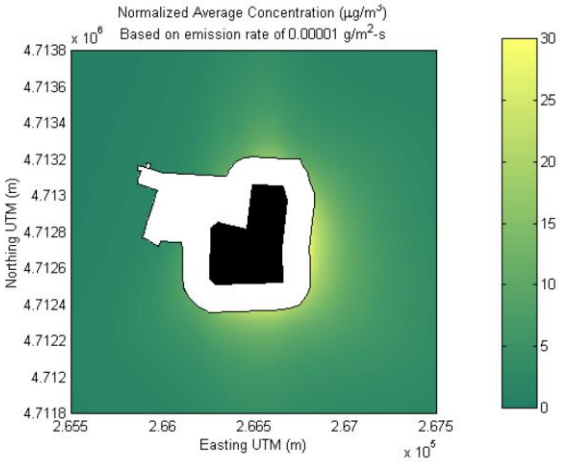
Ambient Air Impacts

$$DF = Dilution\ Factor = \frac{Concentration\ in\ Landfill\ Gas}{Concentration\ in\ Ambient\ Air}$$

- Relevant factors from dispersion modeling studies
  - Emission sources: Fugitive, flares, energy recovery IC engines and turbines, *etc.*
  - Gas system collection efficiency, combustion device destruction efficiency
  - Configuration of sources – fugitive area emissions, point stack emissions
  - Property boundaries and off-site receptor locations
  - Meteorological data
- *DF* Results from a survey of modeling studies *
  - Flare: Median *DF* = 2,000,000
  - IC Engine: Median *DF* = 190,000,000
  - Fugitive: Median *DF* = 71,000,000



- Preliminary EREF results indicate impacts comparable to published background levels
- Site-specific mix of sources and modeling inputs will be relevant to the overall *DF*
- Toxicity data are not available to evaluate health risks

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** See Zemba, Close and Estabrooks article "PFAS Releases from Landfills" in the March issue of the A&WMA em Magazine*

Example Estimation of Impacts

- Assume
  - FTOH concentration in landfill gas:
    5×10^7 pg/m^3 = 50,000 ng/m^3
  - 20 m^3/day inhalation volume
  - 70 kg adult
  - 30 years residential exposure
  - DF = 2,000,000
  - Possible 0.1% conversion to PFOA
- Calculate/estimate
  - FTOH exposure
    0.007 ng/kg-d = (50,000/2,000,000) × 20 / 70
  - Effective PFOA exposure (with 0.1% conversion)
    0.000007 ng/kg-d
- Toxicity data for PFOA (EPA RSL Tables)
  - Reference/safe dose = 0.03 ng/kg-d
  - Cancer potency = 0.0293 d-kg/ng
- Health risk estimates
  - Non-cancer HQ = 0.000007 / 0.03 = 0.0002
  - Cancer risk CR = 0.000007 × 0.0293 × (30/70) = 9×10^{-8}
- However, recognize some caveats
  - Assessment is a crude order-of-magnitude estimate
  - Not all FTOHs or PFAS have been considered
  - Alternate views exist on PFAS toxicity

$$DF = Dilution\ Factor = \frac{Concentration\ in\ Landfill\ Gas}{Concentration\ in\ Ambient\ Air}$$

Table 1. Dilution Factors (*DF*s) Obtained in Dispersion Modeling Studies.

Emission Source	Number of Studies	Dilution Factors Min.	Max.	Median
Fugitive	11	30,000	9,600,000	2,000,000
IC Engine	2	160,000,000	220,000,000	190,000,000
Flare	8	46,000,000	260,000,000	73,000,000

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8:2 fluorotelomer alcohol: A one-day nose-only inhalation toxicokinetic study in the Sprague-Dawley rat with application to risk assessment

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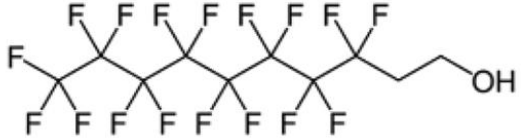
Table from the article "PFAS Releases from Landfills" in the March issue of the A&WMA em Magazine

Conclusions and Recommendations

- Fluorotelomer alcohols (FTOHs) are a category of PFAS that have been measured in air above background at locations downwind of landfills and wastewater treatment plants
  - FTOH presence confirmed in landfill gas



  - Nationwide, air emission of FTOHs via landfill gas may be comparable in magnitude to the discharge of PFAS via leachate
  - Multiple potential sources of emissions at landfills (including fugitive releases)
  - Preliminary data from the EREF-sponsored study suggest effective PFAS destruction by engines and flares and modeled impacts comparable to background levels in ambient air
- Recommendation: **Be prepared to address PFAS air emissions**
  - Facility-specific measurements in landfill gas could be beneficial
  - Stack emission measurements may be useful but could require method extensions
  - Modeling studies could be useful to assess potential impacts
    - Compare to published background levels
    - Air toxicity data have not been published by EPA, but reasonable worst-case assumptions to estimate exposure and compare to available data

Thank you!

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For more on PFAS

ITRC Technical Resources for PFAS

https://www.itrcweb.org/Team/Public?teamID=78

- Fact sheets
- Web-based Technical and Regulatory Guidance Document
- On-line Training Materials