



Atmospheric Constraints on Methane Inventories: How Much Do We Know and How do We Know It?

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with contributions from many, many colleagues. Please see our list of citations.



Stakeholder Workshop:

EPA GHG Data on Natural Gas and Petroleum Systems 7 November, 2019 Pittsburg, PA







Outline

Introduction to the challenges of complementary methods. Describe atmospheric methods for deriving regional methane emissions. How we can account for:

> Multiple regional sources Day / night emissions Background contamination Variations in emission over time?

Review some recent atmospheric studies of oil/gas methane emissions:

Airborne/ automobile site-based work synthesized by EDF

Princeton study

Penn State airborne work

Outline research needs moving forward



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What's the issue? Why use atmospheric methods? Why are there disagreements among methods?



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How do we know methane emissions? How well do we know methane emissions?

- Our understanding of the *global* methane budget comes from *atmospheric measurements*.
- While the total of global emissions is pretty well known, the uncertainty by source or by region can be quite large.

We know total global emissions because we know the total amount of methane in the atmosphere. *Not* because we added up all the pieces.



Notes: - Multiple significant

sources. None is dominant.

Large
uncertainty
bounds. (Why?)

Units are TgCH₄
per year

Global Carbon Project, 2017



And observations of methane averaged across the global network.







What's the issue? Why use atmospheric methods? Why are there disagreements among methods?



Method 1: Bottom-up Approach



		2014 EPA Inventory Values	
		National Emission Factor or Range of	Calculate
Activity	National Activity Data	Regional Values (Potential) ²²	Potential (Mg) ^b
Cas Wells			
Gas Wells	E02 972 weller 1 m	NAdd	0.0
Associated Gas Wells	503,873 Wells41.44	7 40 40 40	47.754
Non-associated Gas Wells (less tractured wells)	205,363 Wells41	7.43-42.49 SCI0/Well ^o	17,754.
Gas Wells with Hydraulic Fracturing	250,777 wells41	7.59-42.49 sctd/well ^e	35,085.
Well Pad Equipment			
Heaters	99,038 heaters ^{4,4,2}	14.87-67.29 sctd/heater®	23,953.4
Separators	306,377 separators ^{c,d,2}	0.94-142.27 scfd/separator ^b	118,591.
Dehydrators	17,126 dehydrators ^{c,d,2}	23.18-106.25 scfd/dehydrator ^b 9.43-61.68 scfd/meter ^b	8,417. 107,173.
Meters/Piping	523,885 meters ^{c,d,2}		
Compressors	48,518 compressors ^{c,a,2}	263.85-312.19 scfd/compressor ⁶	96,170.
Gathering and Boosting			
Gathering and Boosting Stations*	4,999 stations ^{e,2}	53,066 scfd CH4/station	1,864,870.
Pipeline Leaks	431,051 miles ⁽²	52.38-61.97 scfd/mileb	169,701.
Drilling, Well Completion, and Well Workover			
Gas Well Completions without Hydraulic			
Fracturing	767 completions/year9	707.23-854.65 scf/completion ^b	112
Gas Well Workovers without Hydraulic Fracturing	8,933 workovers/yeara2	2,367.7-2,861.3 scf/workoverb	445.
Hydraulic Fracturing Completions and Workovers	completions and	MT/(completion or	
that vent*	1,791 workovers/year	36.82 workover)*	65,940.
Flared Hydraulic Fracturing Completions and	completions and	MT/(completion or	
Workovers*	548 workovers/vear	4.91 workover)h	2,690.
Hydraulic Fracturing Completions and Workovers	completions and	MT/(completion or	
with RECs*	1.043 workovers/year	3.24 workover)h	3,379.
Hydraulic Fracturing Completions and Workovers	completions and	MT/(completion or	
with RECs that flare*	1.979 workovers/vear	4.88 workover)h	9.653.
Well Drilling	18.837 wells ¹	2,505,9-2,965.0 scf/well	971
Normal Operations			
Pneumatic Device Vents*	834 919 controllers ^{c,d,2}	176 74-209 12 scfd/devicesd	1,105,119
Pneumatic Device Vents - Low Rieed (LR)	226 280 controllers:42	22 52-26 64 scfd/devicesd	Aggregate
Pneumatic Device Vents - High Bleed (HB)	29 006 controllerse4.2	612 66-724 91 scfd/devices4	Aggregate
Pneumatic Device Vents - Intermittent Rieed (IR)	579 633 controllers ^{6,4,2}	215 13-254 55 scfd/devices4	Aggregate
Chemical Injection Pumps*	83 249 active numpsc42	208 89-252 30 scfd/numns4	128 876
Kimray Pumps	5 012 753 MMscf/vrb2	977 5-1 156 6 scf/MMscft	100.857
Dehydrator Vents	5 625 985 MMscf/vrb2	271 58-321 34 scf/MMscft	31 448
Condensate Tank Vents	0,020,000 minoolyi		01,110.
Condensate Tanks without Control Devices	120 MMbbl/urk1	21 87-302 75 cof/bbl	253 002
Condensate Tanks with Control Devices	120 MMbbl/wk1	4 27-60 55 cof/bbl	200,002.
Compressor Exhaust Vented	135 mmbbryts	4.37-00.33 SCI/DDF	30,010.
Compressor Exhaust Vented	E1 CA9 MMUDL-b2	0.027.0.080 ccf/UDbsb	240 756
Gas Engines	51,040 MMPP1I**	0.237-0.200 SCI/HP1II*	249,700.
Well Clean Ups	00.477	0.050 4 407 400 6 4 6	140 500
Liquids Unloading with Plunger Lifts"	22,477 venting weilsam2	2,856-1,137,406 scty/venting weim-	112,508.0
Liquids Unicading without Plunger Lifts*	37,912 venting wellsam2	77,891-2,002,960 scty/venting weim-	148,075.
Blowdowns			
Vessel Blowdowns	422,542 vessels ^{b,2}	76.86-90.94 scfy/vessel ^b	668.
Pipeline Blowdowns	431,051 miles (gathering) ^{b,2}	304.49-360.28 scfy/mile ^b	2,702.
Compressor Blowdowns	48,518 compressors ^{b,2}	3,719-4,400 scfy/compressor ^b	3,713.
Compressor Starts	48,518 compressors ^{b,2}	8,320-9,844 scfy/compressor ^b	8,308.
Upsets			
Pressure Relief Valves	1,015,507 PRV ^{b,2}	33.50-39.64 scfy/PRVb	700.
Mishaps	107,763 miles ⁽²	659.24-780.03 scf/mile ^b	1,463.
Produced Water from Coal Bed Methane Wells			
Black Warrior	5,480 wells•	0.0023 kt/wello1	12,790.
	-	kt/gal water	
Powder River	20,596,530,150 gal produced water	2.3E-09 drainage ^{1,1}	47,627.3
Offshore Platforms	,		
Shallow Water Gas Platforms (Gulf of Mexico and	shallow water cas		
Pacific)	1.973 platformsp3	8.899 scfd/platforms	123,460

Activity	Act	tivity Data	Emission Fa	ctor (Potential) ³³	Calculated Potential Emissions (Mg)
Fugitives				. ,	
Pipeline Leaks	301,748	miles ^a	1.55	scfd/mile ^b	3,296.3
Compressor Stations (Transmission)*					
Station Total Emissions	1,834	stations ^{c,d,2}	44,459	scfd/station ^{c,d,bb}	573,179.2
Station + Compressor Fugitive					
Emissions	5,221	compressorsc,d,2	9,104	scfd/station ^{c,d,bb}	117,370.9
Reciprocating Compressor	2,173	compressors ^{c,d,2}	9,246	scfd/compressored.bb	339,361.9
Centrifugal Compressor (wet seals)	869	compressorsc,d,2	9,673	scfd/compressored.bb	59,092.2
Centrifugal Compressor (dry seals)	1,304	compressors ^{c,d,2}	6,259	scfd/compressorcd.bb	57,354.2
Compressor Stations (Storage)*					
Station Total Emissions	356	stations ^{c,d,2}	52,604	scfd/station ^{c,d,bb}	131,647.9
Station + Compressor Fugitive					
Emissions	356	stations ^{c,d,2}	10,100	scfd/station ^{c,d,bb}	25,276.0
Reciprocating Compressor	1,520	compressorsc,d,2	9,957	scfd/compressorc.d.bb	106,371.9
Wells (Storage)	19,522	wells ^{b,2}	114.50	scfd/well ^b	15,714.0
M&R (Trans. Co. Interconnect)	2,686	stations ^{e,2}	3,984	scfd/station ^b	75,230.0
M&R (Farm Taps + Direct Sales)	79,646	stations ^{e,2}	31.20	scfd/station ^b	17,468.9
Normal Operation					
Dehydrator vents (Transmission)	1,169,007	MMscf/vrb,2	93.72	scf/MMscf ^e	2,110,1
Dehydrator vents (Storage)	2,169,267	MMscf/yrb,2	117.18	scf/MMscf ^b	4.895.8
Compressor Exhaust	-,,				
Engines (Transmission)	53,295	MMHPhrb.2	0.24	scf/HPhr ^b	246.351.2
Turbines (Transmission)	12,717	MMHPhrb,2	0.01	scf/HPhr ^b	1.396.1
	,				.,
F (0)	5 000				04.077.0
Engines (Storage)	5,339	MMHPnr ^{0,2}	0.24	sct/HPnrº	24,677.0
Turbines (Storage)	1,875	MMHPhr ^{6,2}	0.01	scf/HPhr ^b	205.9
Generators (Engines)	2,608	MMHPhr ^{6,2}	0.24	sct/HPhr ^b	12,055.2
Generators (Turbines)	31	MMHPhr ^{6,2}	0.01	sct/HPhr ^e	3.4
Pneumatic Devices Trans + Stor*					
Pneumatic Devices Transmission	47,140	devices ^{c,d,2}	30,611	scfy/device ^{c,d,bb}	27,792.1
(High Bleed)	4,129	devices ^{c,d,2}	151,969	scty/devicec,d,bb	12,085.2
(Intermittent Bleed)	39,216	devices ^{c,d,2}	19,712	scfy/devicec,d,bb	14,888.7
(Low Bleed)	3,795	devices ^{c,d,2}	11,196	scfy/devicec.d,bb	818.2
Pneumatic Devices Storage	23,964	devices ^{c,d,2}	63,622	scfy/device ^{c,d,bb}	29,364.5
(High Bleed)	8,379	devices ^{c,d,2}	147,983	scfy/devicec.d,bb	23,882.0
(Intermittent Bleed)	13,482	devices ^{c,d,2}	19,333	scfy/devicec,d,bb	5,020.1
(Low Bleed)	2,103	devices ^{c,d,2}	11,414	scfy/device ^{c,d,bb}	462.3
Routine Maintenance/Upsets					
Pipeline venting	301,748	miles ^{a,1}	31.65	Mscfy/mile ^b	183,939.2
Station venting Trans + Storage					
		compressor stations			
Station Venting Transmission	1,834	c,d,2	4,359	Mscfy/station ^b	153,965.5
		compressor stations			
Station Venting Storage	356	c,d,2	4,359	Mscfy/station ^b	29,887.7
LNG Storage					
LNG Stations	70	stations ^{(g,3}	21,507	scfd/station ^b	10,622.8
LNG Reciprocating Compressors	270	compressorsf.g.3	21,116	scfd/compressor ^b	40,146.5
LNG Centrifugal Compressors	64	compressors ^{f.g.3}	30,573	scfd/compressor ^b	13,766.0
LNG Compressor Exhaust					
LNG Engines	579	MMHPhr ^{(g,3}	0.24	scf/HPhr ^b	2.677.7
LNG Turbines	113	MMHPhr ^{f,g,3}	0.01	scf/HPhr ^b	12.4
LNG Station venting	70	stations ^(g,3)	4,359	Mscfv/station ^b	5,898.6
LNG Import Terminals			1,000	moorprotation	0,000.0
I NG Stations	8	stations(a3	21,507	scfd/station ^b	1 270 0
ING Reciprocating Compressors	A1	compressors(a)	21 116	scfd/compressorb	6,056,5
ING Centrifunal Compressors	7	compressoref.g.3	30 573	sofd/compressorb	1 5/17 5
ING Compressor Exhaust	· · ·	Compressors	50,575	aururunipi caaur-	1,047.0
ING Engines	303	MMHPhrta3	0.24	sof/HPhrb	1 401 7
LING LINGINGS	303	MUMPLE III dave	0.24	Somerie	1,401.7

Emissions are primarily from the production sector





Method 2: Atmospheric mass-balance $FLUX = \overline{U}cos(\overline{\theta}) \int_{-b}^{b} \Delta X \int_{z=0}^{z_{top}} n_{air} dz dx$



There are many ways to treat the data, but in the end all atmospheric methods boil down to an atmospheric mass balance problem.

Major studies reveal 60% more methane emissions

In Pennsylvania, Methane Emissions Higher Than EPA Estimates

EPA's Greenhouse Gas Inventory needs some fixing

U.S. Cities Might Release More Methane Than Previously Thought



What's the issue? Why use atmospheric methods? Why are there disagreements among methods?



What could be wrong with the top-down approach?



What could be wrong with the inventory approach?

What if one rare malfunction emits more than 100 working devices?



Emission per device



This well is emitting 25kg/hr There's 5 wells in the basin Total emissions in this region=5*35=125kg/hr



Bottom-Up Approach

F

Top-down Approach

The CH₄ enhancement is 34ppb with a wind speed of 3m/s and an ABL depth of 1.3km.

Total emissions in this

region= 241kg/hr

Ð



That top-down estimate is too high! They probably forgot to account for the cows



Bottom-Up Approach

F

Top-down Approach

That bottom-up

estimate is too low! They probably didn't

account for any

extreme emissions!

- Fe

Other possible sources of differences

Source category missing from the inventory

Incomplete sampling of emissions over time

- Can be an issue with either approach

Imperfect knowledge of atmospheric flow

- Can also be a problem with either approach

Other possible sources of differences

Source category missing from the inventory

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Extrapolation estimate: Pneumatic devices

Inventory: Allen et al (2013) sampled ~300 of them for about one hour each. Total: 60,000 of them operating for 5 years. Sample / Total = 300 device-hours / $60,000^*365^*24^*5$ device hours = $1x10^{-7}$. Extrapolation by a factor of 10,000,000.

Airborne work: Aircraft samples of 20,000 devices for 10 hours each (mixed in with many other devices, of course).

Sample / Total = 200,000 device-hours / large number above = 1×10^{-4} .

About 1,000 times more data coverage. (with associated complications of many colocated sources)



Outline

Introduction to the challenges of *complementary* methods.

My point of view: It is very difficult to measure total emissions of methane from a complex national network of small leaks.

We have a stronger understanding when we search for consistency across methods that have complementary strengths.

Our current national methane emissions inventory is NOT consistent with atmospheric measurements.



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Time to deploy the grad student



The Marcellus Study:



Objective: To quantify natural gas emissions from production in the northeastern Marcellus region

ADVANTAGES

- Despite having only 3000 wells, 10% of all natural gas in the US is produced in northeast Pennsylvania (NEPA)
- 1. There's nothing else nearby, making it easy to interpret what we're measuring (or is it).
- 1. Dad lives in region and is a source of cheap labor to fix science instrumentation (i.e. restart router)

Deriving Natural Gas Emissions: 3 Steps



Step 1: Measure methane in Northeast PA





















77.5[°] W

77.0 W 76.5[°] W 75.5° W

75.5[°] W

76.0[°] W

0.06

76.0 W



















05/21/2015

05/24/2015 (2)

77.0[°] W 76.5[°] W 76.0[°] W 75.5 W

77.5' W







05/24/2015 (1)

05/14/2015

42.0'1

41.5[°] N

42.0

41.5[°] N

42[°] N

41[°] N

40[°] N

81[°] W

80° W

79[°] W

78[°] W

77°

77.5° W 77.0[°] W 76.5[°] W 76.0[°] W 75.5[°] W



74° W

75[°] W

76[°] W



0.02 0.04 CH₄ Enhancement (ppm)

Barkley et al., ACP, 2017

10 flights

Step 2: Model Methane Enhancements

CH₄ Emissions Inventory







Coal mines/beds



Landfill / Industry





Step 2: Model Methane Enhancements

-Use Weather Research and Forecasting Model (WRF-Chem) to model methane emissions throughout region at 3 km resolution





Modeling domain to simulate the atmospheric conditions during the deployment period (2015-2017) WRF

Unconventional Production/Gathering Coal Mines May 24th 2015 Total Enhancement **NG Transmission/Distribution Enteric Fermentation** 0600Z **Conventional Wells** Landfills and Other 0.01 0.02 0.03 0.04 0.05 0 Modeled Methane Enhancement (in ppm) Barkley et al., ACP, 2017

Step 3: Optimize Natural Gas Emissions

May 29th 2015:



Aircraft emissions estimate on May 29th 2015





Observed CH₄ Enhancement measured during the flight (in ppm)

Aircraft emissions estimate on May 29th 2015





Observed and modeled Non-Natural Gas CH₄ enhancement for the May 29th flight (in ppm)

Aircraft emissions estimate on May 29th 2015





Barkley et al., ACP, 2017

Observation-derived natural gas CH₄ enhancement for the May 29th flight (in ppm)
Aircraft emissions estimate on May 29th 2015





Observed and modeled Natural Gas CH₄ Enhancement for the May 29th flight (in ppm)

Aircraft emissions estimate on May 29th 2015





Observed and optimized Natural Gas CH₄ enhancement for the May 29th flight (in ppm)

EXAMPLE 2: MAY 24th, 2015 The utility of a model-based approach

Aircraft emissions estimate on May 24th 2015



Observed CH₄ enhancement for the May 24th flight at 20z (in ppm)



Modeled CH₄ Enhancement for May 24th, 2015





Coal plume has a significant impact on the regional measurements



May 24th 2015: WRF vs Obs All sources



Optimized Natural Gas Emission Rate = 0.29%

Barkley et al., ACP, 2017

Best-guess upstream emission estimates





Optimal mean leakage rate based on 10 flights in May 2015: **0.39% of production** Barkley et al., ACP, 2017

Let's quantify natural gas emissions in Southwest Pennsylvania



In this region, both coal and UNG wells are major sources of methane emissions

6 flights (19 transects) in 2015-2016 performed by the University of Maryland



There's a lot more methane in SWPA



GOOD NEWS: Total flux is **easier to quantify** BAD NEWS: Total flux is **harder to attribute**



Duration of transect (minutes)



September 14, 2015





September 14, 2015 0.4 Obs CH, Enhancement 0.35 Modeled CH₄ Enhancement 0.3 0.25 Enhancement (ppm) 0.2 0.15 0.1 0.05 -0.05 -0.1 20 17 18 21 Hour (UTC) UNG Rate= 1.6%

Optimized Model vs Obs solution using:

Coal rate= 1.0 x EPA inventory

Continuous ethane measurements allow us to characterize the ethane/methane ratio of the mixed coal and gas plume



Ratios appear to be close to 3% ethane to methane.

Ratios of individual sources







SWPA Coal: 0.3% C₂H₆/CH₄

Kim 1973

SWPA Gas: 7.0% C₂H₆/CH₄

Colon-Roman 2016

Biogenic sources: 0% C₂H₆/CH₄ It is known

We can plug this information into the model to see what rates give us the observed ratio of the mixed plume

Replicating the ethane/methane signal

09/14/2017



UNG = 0.9% of production

Barkley et al., GRL, 2019A

Coal Rate = 1.3 x EPA Inventory



Find where solutions overlap across the 19 transects

Bottom up inventory projects on a construction of production!!!

Gridded EPA Inventory for 2012



What if we estimate emissions from all of the south-central U.S. at once?

Can this be done? Does it match up with inventories?



Fly downwind of gas production in southern US and use frontal transects to estimate emissions









- Five, six-week campaigns over 3 years, covering each season and summer twice. ~25 flights / campaign.
- Each campaign: 2 weeks in each of 3 regions across US (MidAtlantic, MidWest, SouthCentral).
- About 50% of the data in the atmospheric boundary layer (ABL).
- 1140 total flight hours. About 1,500 flasks and 1,000 vertical profiles.

Optimization of Methane Sources: Oct 18th









2017/10/26



2017/10/30



2017/11/02



We're really good at recreating the total methane plume



Figure 2. Observed vs. modelled CH_4 for each of the 7 flights using the optimized gas and animal ag emission rates for each flight.



...but knowing which source to attribute it to will take more information.

Optimization of Methane Sources: Oct 18th, 2017



CH₄ Enhancement (ppm)

Major methane sources in the South





Major ethane sources in the South







ID	Basin	C ₂ H ₆ /CH ₄
А	Anadarko	0.080
В	Woodford	0.070
С	Permian	0.125
D	Ft. Worth	0.067
E	East Texas	0.040
F	Gulf	0.051

10/21/2017



Methane Enhancement (ppm)





Ethane Optimization



Figure 4. Observed vs. modelled C_2H_6 for each of the 7 flights using the optimized gas and animal ag emission rates for each flight.



Best estimate of oil and gas emissions is roughly 2x inventory.

Animal agriculture emissions estimate is roughly equal to inventory.

Barkley et al., GRL, 2019B

Figure 5. Optimized EPA gas inventory multipliers and their 95% confidence intervals for each flight. Each color represents a different strategy used in the optimization. (blue) Both gas and animal ag inventories were optimized using CH₄ data. (red) Only gas inventories were optimized, keeping animal ag values constrained by their inventory data. (yellow) Gas inventories were optimized using C₂H₆ data. (purple?) Both gas and animal ag inventories were optimized using the joint CH₄-C₂H₆ technique.



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Outline research needs moving forward



Synthesis

Describe atmospheric methods for deriving regional methane emissions. How we can account for:

Multiple regional sources

Trace gases (in this case, ethane). Spatial attribution (gridded inventory).

Day / night emissions

Flight data that integrates over a couple of days of emissions (south-central US).

Background contamination

Gridded inventory / spatial attribution and atmospheric transport reanalysis.

Variations in emission over time?

Repeated flights over a region. Tower-deployments spanning months to years.



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Outline research needs moving forward



How can we work towards greater confidence in and understanding of atmospheric emissions estimates?

- Make atmospheric measurements using multiple methods.
- Compare these to each other (and to inventories).
- If these disagree...study, iterate, interrogate...until the results converge.


Atmospheric measurements: Site-level. Ground-based.



Omara et al., 2016; Caulton et al., 2019

Illustration by Omara and Presto, Carnegie Mellon University

PennState

Airborne atmospheric methane observations: Entire gas-basin



Barkley et al., 2017, Atmospheric Chemistry and Physics



Pennsylvania gas wells, among the most productive in the nation, have very low emissions as a percentage of production.

But atmospheric data suggests the emissions in Pennsylvania are 2-5 times higher than EPA inventories would suggest.

Environmental Defense Fund-led, nation-wide re-assessment of natural gas methane emissions.



Fig. 1. Comparison of this work's bottom-up (BU) estimates of methane emissions from oil and natural gas (O/NG) sources to topdown (TD) estimates in nine U.S. O/NG production areas. (A)

Alvarez et al., Science, 2018

Environmental Defense Fund-led, nation-wide re-assessment of natural gas methane emissions.

Table 1. Summary of this work's bottom-up estimates of CH₄ emissions from the U.S. oil and natural gas (O/NG) supply chain (95% confidence interval) and comparison to the EPA Greenhouse Gas Inventory (GHGI).

	2015 CH ₄ Emis		
Industry segment	This work (bottom-up)	EPA GHGI (17)	
Production	7.6 (+1.9/-1.6)	3.5	
Gathering	2.6 (+0.59/-0.18)	2.3	Those do not
Processing	0.72 (+0.20/-0.071)	0.44	These do not
Transmission and Storage	1.8 (+0.35/-0.22)	1.4	agree!
Local Distribution*	0.44 (+0.51/-0.22)	0.44	
Oil Refining and Transportation*	0.034 (+0.050/-0.008)	0.034	
U.S. O/NG total	13 (+2.1/-1.7)	8.1 (+2.1/-1.4) [†]	

*This work's emission estimates for these sources are taken directly from the GHGI. The local distribution estimate is expected to be a lower bound on actual emissions and does not include losses downstream of customer meters due to leaks or incomplete combustion (Section S1.5).

[†]The GHGI only reports industry-wide uncertainties.

Site-by-site data atmospheric data

EPA inventory

Alvarez et al., Science, 2018

Environmental Defense Fund-led, nation-wide re-assessment of natural gas methane emissions.

Methane emissions from the U.S. oil and natural gas supply chain were estimated using ground-based, facility-scale measurements and validated with aircraft observations in areas accounting for ~30% of U.S. gas production. When scaled up nationally, our facility-based estimate of 2015 supply chain emissions is $13 \pm 2 \text{ Tg/y}$, equivalent to 2.3% of gross U.S. gas production. This value is ~60% higher than the U.S. EPA inventory estimate, likely because existing inventory methods miss emissions released during abnormal operating conditions. Methane emissions of this magnitude, per unit of natural gas consumed, produce radiative forcing over a 20-year time horizon comparable to the CO₂ from natural gas combustion. Significant emission reductions are feasible through rapid detection of the root causes of high emissions and deployment of less failure-prone systems. Alvarez et al., Science, 2018

What's causing this discrepancy? A small number of large sources

higher than mean production site emissions estimated in this work). Emissions released from liquid storage tank hatches and vents represented 90% of these sightings. It appears that abnormal operating conditions must be largely responsible, because the observation frequency was too high to be attributed to routine operations like condensate flashing or liquid unloadings alone (24). All other observations were due to anomalous venting from dehydrators, separators, and flares. Notably, the two largest sources of aggregate emissions in the EPA GHGI-pneumatic controllers and equipment leaks—were never observed from these aerial surveys. Similarly, a national survey of gathering facilities found that emission rates were four times higher at the 20% of facilities where substantial tank venting emissions were observed, as compared to the 80% of facilities without such venting (25). In addition, very large emissions from leaking isolation valves

Princeton Marcellus study

-measures ~650 wellpads or 18% of all active unconventional wellpads in the state.
-Finds emission rate of 0.53%
-PA DEP inventory (using EPA methods) estimates emission rate of ~0.1%
-Factor of 5 different!

Abstract

A large-scale study of methane emissions from well pads was conducted in the Marcellus shale (Pennsylvania), the largest producing natural gas shale play in the United States, to better identify the prevalence and characteristics of superemitters. Roughly 2100 measurements were taken from 673 unique unconventional well pads corresponding to ~18% of the total population of active sites and ~32% of the total statewide unconventional natural gas production. A log-normal distribution with a geometric mean of 2.0 kg h⁻¹ and arithmetic mean of 5.5 kg h⁻¹ was observed, which agrees with other independent observations in this region. The geometric standard deviation (4.4 kg h⁻¹) compared well to other studies in the region, but the top 10% of emitters observed in this study contributed 77% of the total emissions, indicating an extremely skewed distribution. The integrated proportional loss of this representative sample was equal to 0.53% with a 95% confidence interval of 0.45–0.64% of the total production of the sites, which is greater than the U.S. Environmental Protection Agency inventory estimate (0.29%), but in the lower range of other mobile observations (0.09–3.3%). These results emphasize the need for a sufficiently large sample size when characterizing emissions distributions that contain superemitters.



Distribution of emissions per well pad



Caulton et al, Environmental Science and Technology, 2019

Oh wait, the x-axis extends further



Caulton et al, Environmental Science and Technology, 2019

Median vs Mean are a factor of 6 different.



Median may characterize what to expect at a given wellpad, but doesn't represent the total GHG emissions from the system

Cumulative distribution of emissions, site-by-site



Caulton et al, Environmental Science and Technology, 2019



Outline

Introduction to the challenges of complementary methods. Describe atmospheric methods for deriving regional methane emissions. How we can account for:

> Multiple regional sources Day / night emissions Background contamination

Variations in emission over time?

Review some recent atmospheric studies of oil/gas methane emissions:

Airborne/ automobile site-based work synthesized by EDF Princeton study

Penn State airborne work

Outline research needs moving forward



Synthesis

Review some recent atmospheric studies of oil/gas methane emissions:

Airborne/ automobile site-based work synthesized by EDF Princeton study Penn State airborne work

- All of these atmospheric data, spanning most of the unconventional gas production in the central and eastern United States, suggest that the EPA inventory currently underestimates emissions by roughly a factor of 2.

- Most of the emissions appear to be caused by a very small number of sites.
- What is missing within the inventory is not clear.

- Continuous monitoring of emissions is limited. Could we just be getting really unlucky with our time sampling?



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Outline research needs moving forward

Long-term, regional-scale atmospheric methane observations

Long-term data sets / analyses underway

Indianapolis. > 5 year record. Complex background conditions, but capacity to simulate this / filter data. Analyses underway. Also > 40 aircraft flights over > 5 years. Synthetic analysis underway. Similar data sets emerging from Boston, Salt Lake City, Los Angeles. Some published results.

Marcellus. 2 year record. Manuscript ready to be drafted. Results could be presented.

N. America - half(?) decade with reasonable CH4 coverage. PSU/NOAA project to perform continental inversions. NIST - 37 tower inversion for the NE US - 2016-2017 underway.

TROPOMI - experimental GEOCARB - to be launched

Deployment of calibrated CRDS instruments at the four identified tower locations



Definitive tower locations of the 4 towers called North (N), East (E), South (S), and Central (C). Unconventional wells are plotted in the background.

	Latitude	Longitude	Installation Date	Elevation (mASL)	Sampling height (mAGL)
Tower N- North	42.0159	-76.4333	05/08/15	476	46
Tower S- South	41.4662	-76.4188	05/07/15	591	61
Tower C- Central	41.7568	-76.3265	05/05/15	341	59
Tower E- East	41.7685	-75.6807	05/13/15	450	59

Coordinates, elevations, and sampling heights of the 4 towers

Barkley et al, in prep



Photo of temporary shed (upper) and tube inlet at tower N, 46m AGL (lower)

Afternoon Towers CH4: What we actually see.



CH₄ enhancement (ppb)





South as background

Recreate pdf of enhancements



Recreate pdf of enhancements



Enhancement (ppm)



Synthesis

Outline research needs moving forward

Continuous monitoring of emissions is happening. These results will be emerging in the data, and the results (to date) appear to be broadly consistent with the airborne studies.

What else is needed?



A call for collaborative research.

Need:

Field measurements designed to understand the difference between inventory and atmospheric methods at the level that allows the inventory to be updated.

Hypothesis: Inventory data are reasonably accurate *for what they include*. Abnormal operating conditions at a small number of sites are not included.

Hard problem. Once we have found sites with anomalously large emissions, how can we clearly identify the discrepancy with inventory, in a way that enables a more accurate inventory?

If we want an accurate national oil and gas methane emissions inventory, we need to solve this problem.





thanks for your attention







References

- Alvarez, Ramón A., Daniel Zavala-Araiza, David R. Lyon, David T. Allen, Zachary R. Barkley, Adam R. Brandt, Kenneth J. Davis, Scott C. Herndon, Daniel J. Jacob, Anna Karion, Eric A. Kort, Brian K. Lamb, Thomas Lauvaux, Joannes D. Maasakkers, Anthony J. Marchese, Mark Omara, Stephen W. Pacala, Jeff Peischl, Allen L. Robinson, Paul B. Shepson, Colm Sweeney, Amy Townsend-Small, Steven C. Wofsy, and Steven P. Hamburg, 2018. Assessment of Methane Emissions from the U.S. Oil and Gas Supply Chain, Science, 10.1126/science.aar7204 (2018).
- Barkley, Z. R., K. J. Davis, S. Feng, N. Balashov, A. Fried, J. DiGangi, 2019B, Forward Modelling and Optimization of Methane Emissions in the South Central United States Using Aircraft Transects Across Frontal Boundaries, in press, Geophysical Research Letters.
- Barkley, Z. R., T. Lauvaux, K. J. Davis, A. Deng, A. Fried, P. Weibring, D. Richter, J. G. Walega, J. DiGangi, S. H. Ehrman, X. Ren, R. R. Dickerson, 2019A., Estimating methane emissions from underground coal and natural gas production in southwestern Pennsylvania. *Geophysical Research Letters.* 46, https://doi.org/10.1029/2019GL082131.
- Barkley, Z. R., Lauvaux, T., Davis, K. J., Deng, A., Cao, Y., Sweeney, C., Martins, D., Miles, N. L., Richardson, S. J., Murphy, T., Cervone, G., Karion, A., Schwietzke, S., Smith, M., Kort, E. A., and Maasakkers, J. D., 2017. Quantifying methane emissions from natural gas production in northeastern Pennsylvania, Atmos. Chem. Phys., doi:10.5194/acp-2017-200, <u>https://www.atmos-chemphys.net/17/13941/2017/</u>
- Caulton D.R., Jessica M. Lu, Haley M. Lane, Bernhard Buchholz, Jeffrey P. Fitts, Levi M. Golston, Xuehui Guo, Qi Li, James McSpiritt, Da Pan, Lars Wendt, Elie Bou-Zeid, and Mark A. Zondlo, 2019. Importance of Superemitter Natural Gas Well Pads in the Marcellus Shale, Environ. Sci. Technol. 2019, 53, 4747–4754
- Omara, M., Sullivan, M. R., Li, X., Subramanian, R., Robinson, A. L., and Presto, A. A.: Methane Emissions from Conven- tional and Unconventional Natural Gas Production Sites in the Marcellus Shale Basin, Environ. Sci. Technol., 50, 2099–2107, <u>https://doi.org/10.1021/acs.est.5b05503</u>, 2016.
- Global Carbon Project, 2017 http://www.globalcarbonproject.org/methanebudget/index.htm