REPORT

Assessment of methane emissions from the U.S. oil and gas supply chain

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Abstract

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 ± 2 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 CO_2 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.

Methane (CH_4) is a potent greenhouse gas, and CH_4 emissions from human activities since pre-industrial times are responsible for 0.97 W m^{-2} of radiative forcing, as compared to 1.7 W m^{-2} for carbon dioxide (CO_2) (*1*). CH_4 is removed from the atmosphere much more rapidly than CO_2, thus reducing CH_4 emissions can effectively reduce the near-term rate of warming (*2*). Sharp growth in U.S. oil and natural gas (O/NG) production beginning around 2005 (*3*) raised concerns about the climate impacts of increased natural gas use (*4*, *5*). By 2012, disagreement among published estimates of CH_4 emissions from U.S. natural gas operations led to a broad consensus that additional data were needed to better characterize emission rates (*4*–*7*). A large body of field measurements made between 2012 and 2016 (table S1) has dramatically improved understanding of the sources and magnitude of CH_4 emissions from the industry's operations. Brandt *et al*. summarized the early literature (*8*); other assessments incorporated elements of recent data (*9*–*11*). This work synthesizes recent studies to provide an improved overall assessment of emissions from the O/NG supply chain, which we define to include all operations associated with oil and natural gas production, processing and transport (Section S1.0) (*12*).

Measurements of O/NG CH_4 emissions can be classified as either top-down (TD) or bottom-up (BU). TD studies quantify ambient methane enhancements using aircraft, satellites or tower networks and infer aggregate emissions from all contributing sources across large geographies. TD estimates for nine O/NG production areas have been reported to date (table S2). These areas are distributed across the U.S. (fig. S1) and account for ~33% of natural gas, ~24% of oil production, and ~14% of all wells (*13*). Areas sampled in TD studies also span the range of hydrocarbon characteristics (predominantly gas, predominantly oil, or mixed), as well as a range of production characteristics such as well productivity and maturity. In contrast, BU studies generate regional, state, or national emission estimates by aggregating and extrapolating measured emissions from individual pieces of equipment, operations, or facilities, using measurements made directly at the emission point or, in the case of facilities, directly downwind.

Recent BU studies have been performed on equipment or facilities that are expected to represent the vast majority of emissions from the O/NG supply chain (table S1). In this work we integrate the results of recent facility-scale BU studies to estimate CH_4 emissions from the U.S. O/NG supply chain, and then we validate the results using TD studies (Section S1). The probability distributions of our BU methodology are based on observed facility-level emissions, in contrast to the component-by-component approach used for conventional inventories. We thus capture enhancements produced by all sources within a facility, including the heavy tail of the distribution. When the BU estimate is developed in this manner, direct comparison of BU and TD estimates of CH_4 emissions in the nine basins for which TD measurements have been reported indicates agreement between methods, within estimated uncertainty ranges (**Fig. 1**).

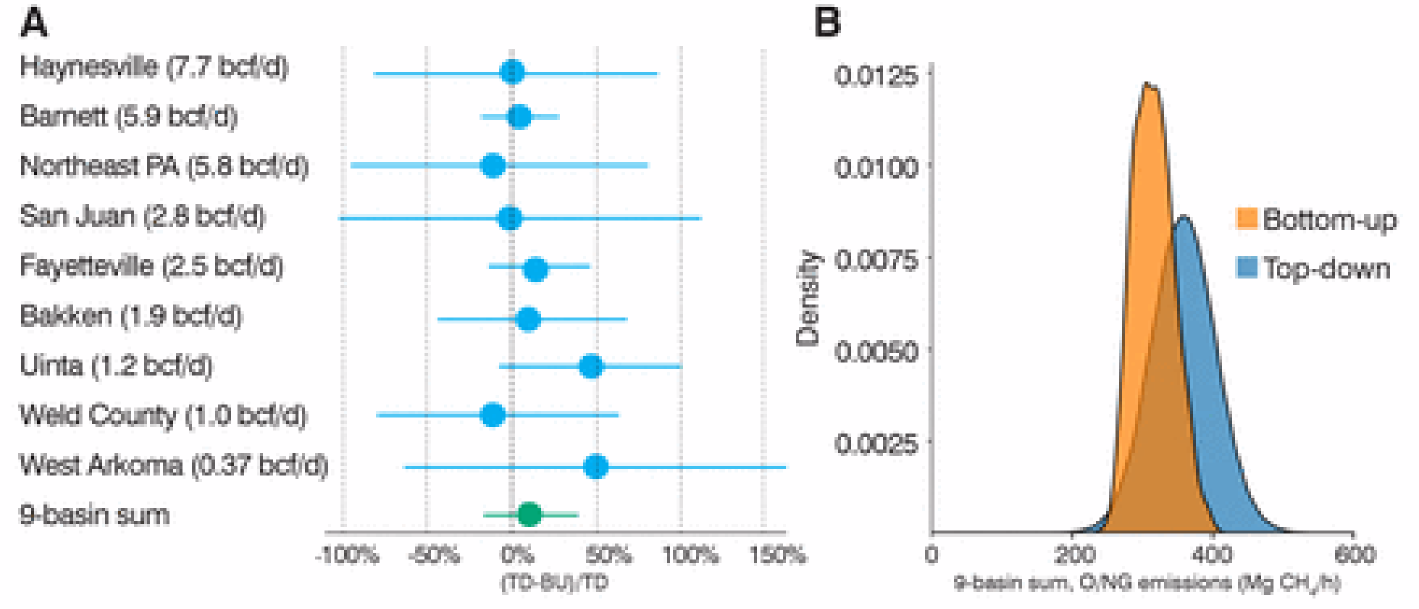

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Fig. 1 Comparison of this work's bottom-up (BU) estimates of methane emissions from oil and natural gas (O/NG) sources to top-down (TD) estimates in nine U.S. O/NG production areas.

(**A**) Relative differences of the TD and BU mean emissions, normalized by the TD value, rank ordered by natural gas production in billion cubic feet per day (bcf/d, where 1 bcf = 2.8×10^7 m^3). Error bars represent 95% confidence intervals. (**B**) Distributions of the 9-basin sum of TD and BU mean estimates (blue and orange probability density, respectively). Neither the ensemble of TD-BU pairs (A) nor the 9-basin sum of means (B) are statistically different (p=0.13 by a randomization test, and mean difference of 11% [95% confidence interval of -17% to 41%]).

Our national BU estimate of total CH_4 emissions in 2015 from the U.S. O/NG supply chain is 13 (+2.1/-1.6, 95% confidence interval) Tg CH_4/y (**Table 1**). This estimate of O/NG CH_4 emissions can also be expressed as a production-normalized emission rate of 2.3% (+0.4%/-0.3%) by normalizing by annual gross natural gas production (33 trillion cubic feet (*13*), with average CH_4 content of 90 vol%). Roughly 85% of national BU emissions are from production, gathering, and processing sources, which are concentrated in active O/NG production areas.

Table 1 Summary of this work's bottom-up estimates of CH_4 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).

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Our assessment does not update emissions from local distribution and end use of natural gas, due to insufficient information addressing this portion of the supply chain. However, recent studies suggest that local distribution emissions are significant, exceeding the current inventory estimate (*14*–*16*), and that end-user emissions might also be important. If these findings prove to be representative, overall emissions from the natural gas supply chain would increase relative to the value in **Table 1** (Section S1.5).

Our BU method and TD measurements yield similar estimates of U.S. O/NG CH_4 emissions in 2015, and both are significantly higher than the corresponding estimate in the U.S. Environmental Protection Agency's Greenhouse Gas Inventory (EPA GHGI) (**Table 1**, Section S1.3) (*17*). Discrepancies between TD estimates and the EPA GHGI have been reported previously (*8*, *18*). Our BU estimate is 63% higher than the EPA GHGI, largely due to a more than two-fold difference in the production segment (**Table 1**). The discrepancy in production sector emissions alone is ~4 Tg CH_4/y, an amount larger than the emissions from any other O/NG supply chain segment. Such a large difference cannot be attributed to expected uncertainty in either estimate: the extremal ends of the 95% confidence intervals for each estimate differ by 20% (i.e., ~12 Tg/y for the lower bound of our BU estimate can be compared to ~10 Tg/y for the upper bound of the EPA GHGI estimate).

We believe the reason for such large divergence is that sampling methods underlying conventional inventories systematically underestimate total emissions because they miss high emissions caused by abnormal operating conditions (e.g., malfunctions). Distributions of measured emissions from production sites in BU studies are invariably "tail-heavy", with large emission rates measured at a small subset of sites at any single point in time (*19*–*22*). Consequently, the most likely hypothesis for the difference between the EPA GHGI and BU estimates derived from facility-level measurements is that measurements used to develop GHGI emission factors under-sample abnormal operating conditions encountered during the BU work. Component-based inventory estimates like the GHGI have been shown to underestimate facility-level emissions (*23*), probably because of the technical difficulty and safety and liability risks associated with measuring large emissions from, for example, venting tanks such as those observed in aerial surveys (*24*).

Abnormal conditions causing high CH_4 emissions have been observed in studies across the O/NG supply chain. An analysis of site-scale emission measurements in the Barnett Shale concluded that equipment behaving as designed could not explain the number of high-emitting production sites in the region (*23*). An extensive aerial infrared camera survey of ~8,000 production sites in seven U.S. O/NG basins found that ~4% of surveyed sites had one or more observable high emission-rate plumes (*24*) (detection threshold of ~3-10 kg CH_4/h was 2-7 times 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 at transmission and storage facilities were quantified using downwind measurement but could not be accurately (or safely) measured using on-site methods (*26*). There is an urgent need to complete equipment-based measurement campaigns that capture these large emission events, so that their causes are better understood.

In contrast to abnormal operational conditions, alternative explanations such as outdated component emission factors are unlikely to explain the magnitude of the difference between our facility-based BU estimate and the GHGI. First, an equipment-level inventory analogous to the EPA GHGI but updated with recent direct measurements of component emissions (Section S1.4) predicts total production emissions that are within ~10% of the EPA GHGI, although the contributions of individual source categories differ significantly (table S3). Second, we consider unlikely an alternative hypothesis that systematically higher emissions during daytime sampling cause a high bias in TD methods (Section S1.6). Two other factors may lead to low bias in EPA GHGI and similar inventory estimates. Operator cooperation is required to obtain site access for emission measurements (*8*). Operators with lower-emitting sites are plausibly more likely to cooperate in such studies, and workers are likely to be more careful to avoid errors or fix problems when measurement teams are on site or about to arrive. The potential bias due to this "opt-in" study design is very challenging to determine. We therefore rely primarily on site-level, downwind measurement methods with limited or no operator forewarning to construct our BU estimate. Another possible source of bias is measurement error. It has been suggested that malfunction of a measurement instrument widely used in the O/NG industry contributes to underestimated emissions in inventories (*27*); however, this cannot explain the >2x difference in production emissions (*28*).

The tail-heavy distribution for many O/NG CH_4 emission sources has important implications for mitigation since it suggests that most sources – whether they represent whole facilities or individual pieces of equipment – can have lower emissions when they operate as designed. We anticipate that significant emissions reductions could be achieved by deploying well-designed emission detection and repair systems that are capable of identifying abnormally operating facilities or equipment. For example, pneumatic controllers and equipment leaks are the largest emission sources in the O/NG production segment exclusive of missing emission sources (38% and 21%, respectively; table S3) with malfunctioning controllers contributing 66% of total pneumatic controller emissions (Section S1.4) and equipment leaks 60% higher than the GHGI estimate.

Gathering operations, which transport unprocessed natural gas from production sites to processing plants or transmission pipelines, produce ~20% of total O/NG supply chain CH_4 emissions. Until the publication of recent measurements (*29*), these emissions were largely unaccounted by the EPA GHGI. Gas processing, transmission and storage together contribute another ~20% of total O/NG supply chain emissions, most of which come from ~2,500 processing and compression facilities.

Our estimate of emissions from the U.S. O/NG supply chain (13 Tg CH_4/y) compares to the EPA estimate of 18 Tg CH_4/y for all other anthropogenic CH_4 sources (*17*). Natural gas losses are a waste of a limited natural resource (~$2 billion/y), increase global levels of surface ozone pollution

(*30*), and significantly erode the potential climate benefits of natural gas use. Indeed, our estimate of CH_4 emissions across the supply chain, per unit of gas consumed, results in roughly the same radiative forcing as does the CO_2 from combustion of natural gas over a 20-year time horizon (31% over 100 years). Moreover, the climate impact of 13 Tg CH_4/y over a 20-year time horizon roughly equals that from the annual CO_2 emissions from all U.S. coal-fired power plants operating in 2015 (31% of the impact over a 100-year time horizon) (Section S1.7).

We suggest that inventory methods would be improved by including the substantial volume of missing O/NG CH_4 emissions evident from the large body of scientific work now available and synthesized here. Such empirical adjustments based on observed data have been previously used in air quality management (*31*).

The large spatial and temporal variability in CH_4 emissions for similar equipment and facilities (due to equipment malfunction and other abnormal operating conditions) reinforces the conclusion that significant emission reductions are feasible. Key aspects of effective mitigation include pairing well-established technologies and best practices for routine emission sources with economically viable systems to rapidly detect the root causes of high emissions arising from abnormal conditions. The latter could involve combinations of current technologies such as on-site leak surveys by company personnel using optical gas imaging (*32*), deployment of passive sensors at individual facilities (*33*, *34*) or mounted on ground-based work trucks (*35*), and in situ remote sensing approaches using tower networks, aircraft or satellites (*36*). Over time, the development of less failure-prone systems would be expected through repeated observation of and further research into common causes of abnormal emissions, followed by re-engineered design of individual components and processes.

Supplementary Materials

www.sciencemag.org/cgi/content/full/science.aar7204/DC1

Materials and methods

Additional author disclosures

Figs. S1 to S11

Tables S1 to S12

References (*37*–*77*)

Databases S1 and S2

References and Notes

1. ↵ G. Myhre *et al.*, in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (Cambridge Univ. Press,

Cambridge, UK, 2013); **www.ipcc.ch/pdf/assessment-report/ar5/wg1/WG1AR5_Chapter08_FINAL.pdf**. Google Scholar

2. ↵ J. K. Shoemaker, D. P. Schrag, M. J. Molina, V. Ramanathan, Climate change. What role for short-lived climate pollutants in mitigation policy? *Science* **342**, 1323–1324 (2013). doi:10.1126/science.1240162pmid:24337280 Abstract/FREE Full Text Google Scholar

3. ↵ U.S. Energy Information Administration (EIA), "Annual Energy Outlook 2017" (EIA, 2017); **www.eia.gov/outlooks/aeo/**. Google Scholar

4. ↵ R. W. Howarth, R. Santoro, A. Ingraffea, Methane and the greenhouse-gas footprint of natural gas from shale formations. *Clim. Change* **106**, 679–690 (2011). doi:10.1007/s10584-011-0061-5 CrossRef Web of Science Google Scholar

5. ↵ R. A. Alvarez, S. W. Pacala, J. J. Winebrake, W. L. Chameides, S. P. Hamburg, Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. U.S.A.* **109**, 6435–6440 (2012). doi:10.1073/pnas.1202407109pmid:22493226 Abstract/FREE Full Text Google Scholar

6. U.S. Department of Energy (DOE), "Ninety-day report of the Secretary of Energy Advisory Board's Shale Gas Subcommittee" (2011); **https://energy.gov/downloads/90-day-interim-report-shale-gas-production-secretary-energy-advisory-board**. Google Scholar

7. ↵ National Petroleum Council (NPC), "Prudent Development: Realizing the Potential of North America's Abundant Natural Gas and Oil Resources" (NPC, 2011); **www.npc.org**. Google Scholar

8. ↵ A. R. Brandt, G. A. Heath, E. A. Kort, F. O'Sullivan, G. Pétron, S. M. Jordaan, P. Tans, J. Wilcox, A. M. Gopstein, D. Arent, S. Wofsy, N. J. Brown, R. Bradley, G. D. Stucky, D. Eardley, R. Harriss, Energy and environment. Methane leaks from North American natural gas systems. *Science* **343**, 733–735 (2014). doi:10.1126/science.1247045pmid:24531957 Abstract/FREE Full Text Google Scholar

9. ↵ D. T. Allen, Emissions from oil and gas operations in the United States and their air quality implications. *J. Air Waste Manag. Assoc.* **66**, 549–575 (2016). doi:10.1080/10962247.2016.1171263pmid:27249104 CrossRef PubMed Google Scholar

10. P. Balcombe, K. Anderson, J. Speirs, N. Brandon, A. Hawkes, The natural gas supply chain: The importance of methane and carbon dioxide emissions. *ACS Sustain. Chem.& Eng.* **5**, 3–20 (2017). doi:10.1021/acssuschemeng.6b00144 CrossRef Google Scholar

11. ↵ J. A. Littlefield, J. Marriott, G. A. Schivley, T. J. Skone, Synthesis of recent ground-level methane emission measurements from the U.S. natural gas supply chain. *J. Clean. Prod.* **148**, 118–126 (2017). doi:10.1016/j.jclepro.2017.01.101 CrossRef Google Scholar

12. ↵ See supplementary materials.

13. ↵ Drillinginfo, Inc., Drillinginfo Production Query (2015); **https://info.drillinginfo.com/**. Google Scholar

14. ↵ K. McKain, A. Down, S. M. Raciti, J. Budney, L. R. Hutyra, C. Floerchinger, S. C. Herndon, T. Nehrkorn, M. S. Zahniser, R. B. Jackson, N. Phillips, S. C. Wofsy, Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proc. Natl. Acad. Sci. U.S.A.* **112**, 1941–1946 (2015). doi:10.1073/pnas.1416261112pmid:25617375 Abstract/FREE Full Text Google Scholar

15. B. K. Lamb, M. O. L. Cambaliza, K. J. Davis, S. L. Edburg, T. W. Ferrara, C. Floerchinger, A. M. F. Heimburger, S. Herndon, T. Lauvaux, T. Lavoie, D. R. Lyon, N. Miles, K. R. Prasad, S. Richardson, J. R. Roscioli, O. E. Salmon, P. B. Shepson, B. H. Stirm, J. Whetstone, Direct and indirect measurements and modeling of methane emissions in Indianapolis, Indiana. *Environ. Sci. Technol.* **50**, 8910–8917 (2016). doi:10.1021/acs.est.6b01198pmid:27487422 CrossRef PubMed Google Scholar

16. ↲ D. Wunch, G. C. Toon, J. K. Hedelius, N. Vizenor, C. M. Roehl, K. M. Saad, J.-F. L. Blavier, D. R. Blake, P. O. Wennberg, Quantifying the loss of processed natural gas within California's South Coast Air Basin using long-term measurements of ethane and methane. *Atmos. Chem. Phys.* **16**, 14091–14105 (2016). doi:10.5194/acp-16-14091-2016 CrossRef Google Scholar

17. ↲ U.S. Environmental Protection Agency (EPA), "Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2015" (EPA, 2017); **www.epa.gov/ghgemissions/inventory-us-greenhouse-gas-emissions-and-sinks-1990-2015**. Google Scholar

18. ↲ D. Zavala-Araiza, D. R. Lyon, R. A. Alvarez, K. J. Davis, R. Harriss, S. C. Herndon, A. Karion, E. A. Kort, B. K. Lamb, X. Lan, A. J. Marchese, S. W. Pacala, A. L. Robinson, P. B. Shepson, C. Sweeney, R. Talbot, A. Townsend-Small, T. I. Yacovitch, D. J. Zimmerle, S. P. Hamburg, Reconciling divergent estimates of oil and gas methane emissions. *Proc. Natl. Acad. Sci. U.S.A.* **112**, 15597–15602 (2015). pmid:26644584 Abstract/FREE Full Text Google Scholar

19. ↲ C. W. Rella, T. R. Tsai, C. G. Botkin, E. R. Crosson, D. Steele, Measuring emissions from oil and natural gas well pads using the mobile flux plane technique. *Environ. Sci. Technol.* **49**, 4742–4748 (2015). doi:10.1021/acs.est.5b00099pmid:25806837 CrossRef PubMed Google Scholar

20. M. Omara, M. R. Sullivan, X. Li, R. Subramanian, A. L. Robinson, A. A. Presto, Methane emissions from conventional and unconventional natural gas production sites in the Marcellus Shale Basin. *Environ. Sci. Technol.* **50**, 2099–2107 (2016). doi:10.1021/acs.est.5b05503pmid:26824407 CrossRef PubMed Google Scholar

21. A. M. Robertson, R. Edie, D. Snare, J. Soltis, R. A. Field, M. D. Burkhart, C. S. Bell, D. Zimmerle, S. M. Murphy, Variation in methane emission rates from well pads in four oil and gas basins with contrasting production volumes and compositions. *Environ. Sci. Technol.* **51**, 8832–8840 (2017). doi:10.1021/acs.est.7b00571pmid:28628305 CrossRef PubMed Google Scholar

22. ↲ A. R. Brandt, G. A. Heath, D. Cooley, Methane leaks from natural gas systems follow extreme distributions. *Environ. Sci. Technol.* **50**, 12512–12520 (2016). doi:10.1021/acs.est.6b04303pmid:27740745 CrossRef PubMed Google Scholar

23. ↲ D. Zavala-Araiza, R. A. Alvarez, D. R. Lyon, D. T. Allen, A. J. Marchese, D. J. Zimmerle, S. P. Hamburg, Super-emitters in natural gas infrastructure are caused by abnormal process conditions. *Nat. Commun.* **8**, 14012 (2017). doi:10.1038/ncomms14012pmid:28091528 CrossRef PubMed Google Scholar

24. ↲ D. R. Lyon, R. A. Alvarez, D. Zavala-Araiza, A. R. Brandt, R. B. Jackson, S. P. Hamburg, Aerial surveys of elevated hydrocarbon emissions from oil and gas production sites. *Environ. Sci. Technol.* **50**, 4877–4886 (2016). doi:10.1021/acs.est.6b00705pmid:27045743 CrossRef PubMed Google Scholar

25. ↲ A. L. Mitchell, D. S. Tkacik, J. R. Roscioli, S. C. Herndon, T. I. Yacovitch, D. M. Martinez, T. L. Vaughn, L. L. Williams, M. R. Sullivan, C. Floerchinger, M. Omara, R. Subramanian, D. Zimmerle, A. J. Marchese, A. L.

Robinson, Measurements of methane emissions from natural gas gathering facilities and processing plants: Measurement results. *Environ. Sci. Technol.* **49**, 3219–3227 (2015). doi:10.1021/es5052809pmid:25668106 CrossRef PubMed Google Scholar

26. ↵ D. J. Zimmerle, L. L. Williams, T. L. Vaughn, C. Quinn, R. Subramanian, G. P. Duggan, B. Willson, J. D. Opsomer, A. J. Marchese, D. M. Martinez, A. L. Robinson, Methane emissions from the natural gas transmission and storage system in the United States. *Environ. Sci. Technol.* **49**, 9374–9383 (2015). doi:10.1021/acs.est.5b01669pmid:26195284 CrossRef PubMed Google Scholar

27. ↵ T. Howard, T. W. Ferrara, A. Townsend-Small, Sensor transition failure in the high flow sampler: Implications for methane emission inventories of natural gas infrastructure. *J. Air Waste Manag. Assoc.* **65**, 856–862 (2015). doi:10.1080/10962247.2015.1025925pmid:26079559 CrossRef PubMed Google Scholar

28. ↵ R. A. Alvarez, D. R. Lyon, A. J. Marchese, A. L. Robinson, S. P. Hamburg, Possible malfunction in widely used methane sampler deserves attention but poses limited implications for supply chain emission estimates. *Elem. Sci. Anth.* **4**, 000137 (2016). doi:10.12952/journal.elementa.000137 CrossRef Google Scholar

29. ↵ A. J. Marchese, T. L. Vaughn, D. J. Zimmerle, D. M. Martinez, L. L. Williams, A. L. Robinson, A. L. Mitchell, R. Subramanian, D. S. Tkacik, J. R. Roscioli, S. C. Herndon, Methane emissions from United States natural gas gathering and processing. *Environ. Sci. Technol.* **49**, 10718–10727 (2015). doi:10.1021/acs.est.5b02275pmid:26281719 CrossRef PubMed Google Scholar

30. ↵ A. M. Fiore, D. J. Jacob, B. D. Field, D. G. Streets, S. D. Fernandes, C. Jang, Linking ozone pollution and climate change: The case for controlling methane. *Geophys. Res. Lett.* **29**, 21-1–25-4 (2002). doi:10.1029/2002GL015601 CrossRef Google Scholar

31. ↵ Texas Commission on Environmental Quality (TCEQ), "Houston-Galveston-Brazoria Attainment Demonstration State Implementation Plan Revision for the 1997 Eight-Hour Ozone Standard" (2010), pp. 3–18; **www.tceq.texas.gov/assets/public/implementation/air/sip/hgb/hgb_sip_2009/09017SIP_completeNarr_ado.pdf**. Google Scholar

32. ↵ A. P. Ravikumar, J. Wang, A. R. Brandt, Are optical gas imaging technologies effective for methane leak detection? *Environ. Sci. Technol.* **51**, 718–724 (2017). doi:10.1021/acs.est.6b03906pmid:27936621 CrossRef PubMed Google Scholar

33. ↵ U.S. Department of Energy (DOE) Advanced Research Projects Agency – Energy, (ARPA-E, 2014), "ARPA-E MONITOR Program" (ARPA-E); **https://arpa-e.energy.gov/?q=programs/monitor**. Google Scholar

34. ↵ Environmental Defense Fund (EDF), "Methane Detectors Challenge" (EDF, 2014); **www.edf.org/energy/natural-gas-policy/methane-detectors-challenge**. Google Scholar

35. ↵ J. D. Albertson, T. Harvey, G. Foderaro, P. Zhu, X. Zhou, S. Ferrari, M. S. Amin, M. Modrak, H. Brantley, E. D. Thoma, A mobile sensing approach for regional surveillance of fugitive methane emissions in oil and gas production. *Environ. Sci. Technol.* **50**, 2487–2497 (2016). doi:10.1021/acs.est.5b05059pmid:26807713 CrossRef PubMed Google Scholar

36. ↵ D. J. Jacob, A. J. Turner, J. D. Maasakkers, J. Sheng, K. Sun, X. Liu, K. Chance, I. Aben, J. McKeever, C. Frankenberg, Satellite observations of atmospheric methane and their value for quantifying methane

emissions. *Atmos. Chem. Phys.* **16**, 14371–14396 (2016). doi:10.5194/acp-16-14371-2016 CrossRef Google Scholar

37. ↲ Pipeline and Hazardous Materials Safety Administration (PHMSA), "Pipeline Data and Statistics" (PHMSA, 2017); **https://www.phmsa.dot.gov/data-and-statistics/pipeline/data-and-statistics-overview**. Google Scholar

38. H. L. Brantley, E. D. Thoma, W. C. Squier, B. B. Guven, D. Lyon, Assessment of methane emissions from oil and gas production pads using mobile measurements. *Environ. Sci. Technol.* **48**, 14508–14515 (2014). doi:10.1021/es503070qpmid:25375308 CrossRef PubMed Google Scholar

39. X. Lan, R. Talbot, P. Laine, A. Torres, Characterizing fugitive methane emissions in the Barnett Shale area using a mobile laboratory. *Environ. Sci. Technol.* **49**, 8139–8146 (2015). doi:10.1021/es5063055pmid:26148552 CrossRef PubMed Google Scholar

40. R. Subramanian, L. L. Williams, T. L. Vaughn, D. Zimmerle, J. R. Roscioli, S. C. Herndon, T. I. Yacovitch, C. Floerchinger, D. S. Tkacik, A. L. Mitchell, M. R. Sullivan, T. R. Dallmann, A. L. Robinson, Methane emissions from natural gas compressor stations in the transmission and storage sector: Measurements and comparisons with the EPA greenhouse gas reporting program protocol. *Environ. Sci. Technol.* **49**, 3252–3261 (2015). doi:10.1021/es5060258pmid:25668051 CrossRef PubMed Google Scholar

41. B. W. Yap, C. H. Sim, Comparisons of various types of normality tests. *J. Stat. Comput. Simul.* **81**, 2141–2155 (2011). doi:10.1080/00949655.2010.520163 CrossRef Google Scholar

42. N. M. Razali, Y. B. Wah, Power comparisons of Shapiro-Wilk, Kolmogorov-Smirnov, Lilliefors and Anderson-Darling tests. *J. Statist. Model. Anal.* **2**, 21–33 (2011). Google Scholar

43. U.S. Environmental Protection Agency (EPA), "Public Review of Draft U.S. Inventory of Greenhouse Gas Emissions and Sinks: 1990-2016" (EPA, 2018); **www.epa.gov/ghgemissions/inventory-us-greenhouse-gas-emissions-and-sinks**). Google Scholar

44. A. Townsend-Small, T. W. Ferrara, D. R. Lyon, A. E. Fries, B. K. Lamb, Emissions of coalbed and natural gas methane from abandoned oil and gas wells in the United States. *Geophys. Res. Lett.* **43**, 2283–2290 (2016). doi:10.1002/2015GL067623 CrossRef Google Scholar

45. M. Kang, C. M. Kanno, M. C. Reid, X. Zhang, D. L. Mauzerall, M. A. Celia, Y. Chen, T. C. Onstott, Direct measurements of methane emissions from abandoned oil and gas wells in Pennsylvania. *Proc. Natl. Acad. Sci. U.S.A.* **111**, 18173–18177 (2014). doi:10.1073/pnas.1408315111pmid:25489074 Abstract/FREE Full Text Google Scholar

46. U.S. Environmental Protection Agency (EPA), Greenhouse Gas Reporting Program, (EPA, 2017); **http://ghgdata.epa.gov/ghgp/main.do**. Google Scholar

47. S. Schwietzke, G. Pétron, S. Conley, C. Pickering, I. Mielke-Maday, E. J. Dlugokencky, P. P. Tans, T. Vaughn, C. Bell, D. Zimmerle, S. Wolter, C. W. King, A. B. White, T. Coleman, L. Bianco, R. C. Schnell, Improved mechanistic understanding of natural gas methane emissions from spatially resolved aircraft measurements. *Environ. Sci. Technol.* **51**, 7286–7294 (2017). doi:10.1021/acs.est.7b01810pmid:28548824 CrossRef PubMed Google Scholar

48. A. Gvakharia, E. A. Kort, A. Brandt, J. Peischl, T. B. Ryerson, J. P. Schwarz, M. L. Smith, C. Sweeney, Methane, black carbon, and ethane emissions from natural gas flares in the Bakken Shale, North Dakota. *Environ. Sci. Technol.* **51**, 5317–5325 (2017). doi:10.1021/acs.est.6b05183pmid:28401762 CrossRef PubMed Google Scholar

49. J. Peischl, A. Karion, C. Sweeney, E. A. Kort, M. L. Smith, A. R. Brandt, T. Yeskoo, K. C. Aikin, S. A. Conley, A. Gvakharia, M. Trainer, S. Wolter, T. B. Ryerson, Quantifying atmospheric methane emissions from oil and natural gas production in the Bakken shale region of North Dakota. *J. Geophys. Res. D Atmospheres* **121**, 6101–6111 (2016). doi:10.1002/2015JD024631 CrossRef Google Scholar

50. B. K. Lamb, S. L. Edburg, T. W. Ferrara, T. Howard, M. R. Harrison, C. E. Kolb, A. Townsend-Small, W. Dyck, A. Possolo, J. R. Whetstone, Direct measurements show decreasing methane emissions from natural gas local distribution systems in the United States. *Environ. Sci. Technol.* **49**, 5161–5169 (2015). doi:10.1021/es505116ppmid:25826444 CrossRef PubMed Google Scholar

51. J. Peischl, T. B. Ryerson, K. C. Aikin, J. A. de Gouw, J. B. Gilman, J. S. Holloway, B. M. Lerner, R. Nadkarni, J. A. Neuman, J. B. Nowak, M. Trainer, C. Warneke, D. D. Parrish, Quantifying atmospheric methane emissions from the Haynesville, Fayetteville, and northeastern Marcellus shale gas production regions. *J. Geophys. Res. D Atmospheres* **120**, 2119–2139 (2015). doi:10.1002/2014JD022697 CrossRef Google Scholar

52. M. L. Smith, A. Gvakharia, E. A. Kort, C. Sweeney, S. A. Conley, I. Faloona, T. Newberger, R. Schnell, S. Schwietzke, S. Wolter, Airborne quantification of methane emissions over the Four Corners region. *Environ. Sci. Technol.* **51**, 5832–5837 (2017). doi:10.1021/acs.est.6b06107pmid:28418663 CrossRef PubMed Google Scholar

53. J. D. Maasakkers, D. J. Jacob, M. P. Sulprizio, A. J. Turner, M. Weitz, T. Wirth, C. Hight, M. DeFigueiredo, M. Desai, R. Schmeltz, L. Hockstad, A. A. Bloom, K. W. Bowman, S. Jeong, M. L. Fischer, Gridded national inventory of U.S. methane emissions. *Environ. Sci. Technol.* **50**, 13123–13133 (2016). doi:10.1021/acs.est.6b02878pmid:27934278 CrossRef PubMed Google Scholar

54. D. T. Allen, V. M. Torres, J. Thomas, D. W. Sullivan, M. Harrison, A. Hendler, S. C. Herndon, C. E. Kolb, M. P. Fraser, A. D. Hill, B. K. Lamb, J. Miskimins, R. F. Sawyer, J. H. Seinfeld, Measurements of methane emissions at natural gas production sites in the United States. *Proc. Natl. Acad. Sci. U.S.A.* **110**, 17768–17773 (2013). doi:10.1073/pnas.1304880110pmid:24043804 Abstract/FREE Full Text Google Scholar

55. D. T. Allen, A. P. Pacsi, D. W. Sullivan, D. Zavala-Araiza, M. Harrison, K. Keen, M. P. Fraser, A. Daniel Hill, R. F. Sawyer, J. H. Seinfeld, Methane emissions from process equipment at natural gas production sites in the United States: Pneumatic controllers. *Environ. Sci. Technol.* **49**, 633–640 (2015). doi:10.1021/es5040156pmid:25488196 CrossRef PubMed Google Scholar

56. D. T. Allen, D. W. Sullivan, D. Zavala-Araiza, A. P. Pacsi, M. Harrison, K. Keen, M. P. Fraser, A. Daniel Hill, B. K. Lamb, R. F. Sawyer, J. H. Seinfeld, Methane emissions from process equipment at natural gas production sites in the United States: Liquid unloadings. *Environ. Sci. Technol.* **49**, 641–648 (2015). doi:10.1021/es504016rpmid:25488307 CrossRef PubMed Google Scholar

57. U.S. Environmental Protection Agency (EPA), “AP-42: Compilation of Air Emission Factors, Volume 1, Section 3.2, Natural Gas-fired Reciprocating Engines” (EPA, 2000); **www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emission-factors**. Google Scholar

58. J. Veil, "U.S. Produced Water Volumes and Management Practices in 2012" (Groundwater Protection Council, 2015); **http://www.gwpc.org/sites/default/files/Produced%20Water%20Report%202014-GWPC_0.pdf**. Google Scholar

59. U.S. Environmental Protection Agency (EPA), Oil and Gas Emission Estimation Tool 2014 (EPA, 2015); **ftp://ftp.epa.gov/EmisInventory/2011nei/doc/Tool_and_Report112614.zip**. Google Scholar

60. U.S. Energy Information Administration (EIA), "Natural Gas Consumption by End Use" (EIA, 2017); **www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm**. Google Scholar

61. N. N. Clark, D. L. McKain, D. R. Johnson, W. S. Wayne, H. Li, V. Akkerman, C. Sandoval, A. N. Covington, R. A. Mongold, J. T. Hailer, O. J. Ugarte, Pump-to-wheels methane emissions from the heavy-duty transportation sector. *Environ. Sci. Technol.* **51**, 968–976 (2017). doi:10.1021/acs.est.5b06059pmid:28005343 CrossRef PubMed Google Scholar

62. T. N. Lavoie, P. B. Shepson, C. A. Gore, B. H. Stirm, R. Kaeser, B. Wulle, D. Lyon, J. Rudek, Assessing the methane emissions from natural gas-fired power plants and oil refineries. *Environ. Sci. Technol.* **51**, 3373–3381 (2017). doi:10.1021/acs.est.6b05531pmid:28221780 CrossRef PubMed Google Scholar

63. C. Frankenberg, A. K. Thorpe, D. R. Thompson, G. Hulley, E. A. Kort, N. Vance, J. Borchardt, T. Krings, K. Gerilowski, C. Sweeney, S. Conley, B. D. Bue, A. D. Aubrey, S. Hook, R. O. Green, Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners region. *Proc. Natl. Acad. Sci. U.S.A.* **113**, 9734–9739 (2016). doi:10.1073/pnas.1605617113pmid:27528660 Abstract/FREE Full Text Google Scholar

64. T. N. Lavoie, P. B. Shepson, M. O. L. Cambaliza, B. H. Stirm, A. Karion, C. Sweeney, T. I. Yacovitch, S. C. Herndon, X. Lan, D. Lyon, Aircraft-based measurements of point source methane emissions in the Barnett Shale basin. *Environ. Sci. Technol.* **49**, 7904–7913 (2015). doi:10.1021/acs.est.5b00410pmid:26148549 CrossRef PubMed Google Scholar

65. T. I. Yacovitch, S. C. Herndon, G. Pétron, J. Kofler, D. Lyon, M. S. Zahniser, C. E. Kolb, Mobile laboratory observations of methane emissions in the Barnett Shale region. *Environ. Sci. Technol.* **49**, 7889–7895 (2015). doi:10.1021/es506352jpmid:25751617 CrossRef PubMed Google Scholar

66. M. Etminan, G. Myhre, E. J. Highwood, K. P. Shine, Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing. *Geophys. Res. Lett.* **43**, 12614–12623 (2016). doi:10.1002/2016GL071930 CrossRef Google Scholar

67. Z. R. Barkley, T. Lauvaux, K. J. Davis, A. Deng, N. L. Miles, S. J. Richardson, Y. Cao, C. Sweeney, A. Karion, M. K. Smith, E. A. Kort, S. Schwietzke, T. Murphy, G. Cervone, D. Martins, J. D. Maasakkers, Quantifying methane emissions from natural gas production in north-eastern Pennsylvania. *Atmos. Chem. Phys.* **17**, 13941–13966 (2017). doi:10.5194/acp-17-13941-2017 CrossRef Google Scholar

68. C. S. Foster, E. T. Crosman, L. Holland, D. V. Mallia, B. Fasoli, R. Bares, J. Horel, J. C. Lin, Confirmation of elevated methane emissions in Utah's Uintah Basin with ground-based observations and a high-resolution transport model: Methane emissions in Utah's Uintah Basin. *J. Geophys. Res. D Atmospheres* **122**, 13026–13044 (2017). Google Scholar

69. A. Karion, C. Sweeney, G. Pétron, G. Frost, R. Michael Hardesty, J. Kofler, B. R. Miller, T. Newberger, S. Wolter, R. Banta, A. Brewer, E. Dlugokencky, P. Lang, S. A. Montzka, R. Schnell, P. Tans, M. Trainer, R. Zamora, S.

Conley, Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* **40**, 4393–4397 (2013). doi:10.1002/grl.50811 CrossRef Web of Science Google Scholar

70. G. Pétron, A. Karion, C. Sweeney, B. R. Miller, S. A. Montzka, G. J. Frost, M. Trainer, P. Tans, A. Andrews, J. Kofler, D. Helmig, D. Guenther, E. Dlugokencky, P. Lang, T. Newberger, S. Wolter, B. Hall, P. Novelli, A. Brewer, S. Conley, M. Hardesty, R. Banta, A. White, D. Noone, D. Wolfe, R. Schnell, A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin. *J. Geophys. Res. D Atmospheres* **119**, 6836–6852 (2014). doi:10.1002/2013JD021272 CrossRef Google Scholar

71. A. Karion, C. Sweeney, E. A. Kort, P. B. Shepson, A. Brewer, M. Cambaliza, S. A. Conley, K. Davis, A. Deng, M. Hardesty, S. C. Herndon, T. Lauvaux, T. Lavoie, D. Lyon, T. Newberger, G. Pétron, C. Rella, M. Smith, S. Wolter, T. I. Yacovitch, P. Tans, Aircraft-Based Estimate of Total Methane Emissions from the Barnett Shale Region. *Environ. Sci. Technol.* **49**, 8124–8131 (2015). doi:10.1021/acs.est.5b00217pmid:26148550 CrossRef PubMed Google Scholar

72. M. L. Smith, E. A. Kort, A. Karion, C. Sweeney, S. C. Herndon, T. I. Yacovitch, Airborne ethane observations in the Barnett Shale: Quantification of ethane flux and attribution of methane emissions. *Environ. Sci. Technol.* **49**, 8158–8166 (2015). doi:10.1021/acs.est.5b00219pmid:26148554 CrossRef PubMed Google Scholar

73. J. Garratt, Review: The atmospheric boundary layer. *Earth Sci. Rev.* **37**, 89–134 (1994). doi:10.1016/0012-8252(94)90026-4 CrossRef Google Scholar

74. A. Townsend-Small, E. C. Botner, K. L. Jimenez, J. R. Schroeder, N. J. Blake, S. Meinardi, D. R. Blake, B. C. Sive, D. Bon, J. H. Crawford, G. Pfister, F. M. Flocke, Using stable isotopes of hydrogen to quantify biogenic and thermogenic atmospheric methane sources: A case study from the Colorado Front Range. *Geophys. Res. Lett.* **43**, 11462–11471 (2016). doi:10.1002/2016GL071438 CrossRef Google Scholar

75. T. N. Lavoie, P. B. Shepson, M. O. L. Cambaliza, B. H. Stirm, S. Conley, S. Mehrotra, I. C. Faloona, D. Lyon, Spatiotemporal variability of methane emissions at oil and natural gas operations in the Eagle Ford Basin. *Environ. Sci. Technol.* **51**, 8001–8009 (2017). doi:10.1021/acs.est.7b00814pmid:28678487 CrossRef PubMed Google Scholar

76. J. D. Goetz *et al*., Analysis of local-scale background concentrations of methane and other gas-phase species in the Marcellus Shale. *Elem. Sci. Anth.* **5**, 1 (2017). doi:10.1525/elementa.182 CrossRef Google Scholar

77. ↲ M. F. Hendrick, R. Ackley, B. Sanaie-Movahed, X. Tang, N. G. Phillips, Fugitive methane emissions from leak-prone natural gas distribution infrastructure in urban environments. *Environ. Pollut.* **213**, 710–716 (2016). doi:10.1016/j.envpol.2016.01.094pmid:27023280 CrossRef PubMed Google Scholar

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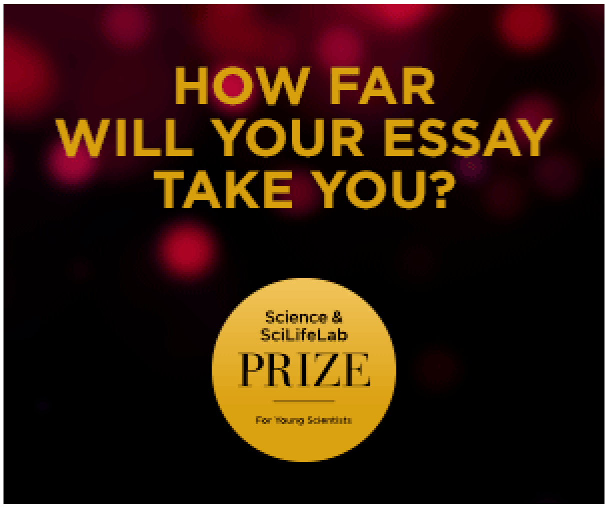

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