GREENHOUSE GASES

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

Ramón A. Alvarez[1]*, Daniel Zavala-Araiza[1], David R. Lyon[1], David T. Allen[2], Zachary R. Barkley[3], Adam R. Brandt[4], Kenneth J. Davis[3], Scott C. Herndon[5], Daniel J. Jacob[6], Anna Karion[7], Eric A. Kort[8], Brian K. Lamb[9], Thomas Lauvaux[3], Joannes D. Maasakkers[6], Anthony J. Marchese[10], Mark Omara[1], Stephen W. Pacala[11], Jeff Peischl[12,13], Allen L. Robinson[14], Paul B. Shepson[15], Colm Sweeney[13], Amy Townsend-Small[16], Steven C. Wofsy[6], Steven P. Hamburg[1]

Methane emissions from the U.S. oil and natural gas supply chain were estimated by 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 teragrams per year, equivalent to 2.3% of gross U.S. gas production. This value is ~60% higher than the U.S. Environmental Protection Agency 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. Substantial 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 preindustrial 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 markedly 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 O/NG production, processing, and transport (materials and methods, 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 (materials and methods). 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).

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/year (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 volume %]. Roughly 85% of national BU emissions are from production, gathering, and processing sources, which are concentrated in active O/NG production areas.

Our assessment does not update emissions from local distribution and end use of natural gas, owing to insufficient information addressing this portion of the supply chain. However, recent studies suggest that local distribution emissions exceed 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 (materials and methods, 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 and materials and methods, 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 twofold difference in the production segment (Table 1). The discrepancy in production sector emissions alone is ~4 Tg CH_4/year, 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/year for the lower bound of our BU estimate can be compared to ~10 Tg/year 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

[1]Environmental Defense Fund, Austin, TX, USA. [2]University of Texas at Austin, Austin, TX, USA. [3]The Pennsylvania State University, University Park, PA, USA. [4]Stanford University, Stanford, CA, USA. [5]Aerodyne Research Inc., Billerica, MA, USA. [6]Harvard University, Cambridge, MA, USA. [7]National Institute of Standards and Technology, Gaithersburg, MD, USA. [8]University of Michigan, Ann Arbor, MI, USA. [9]Washington State University, Pullman, WA, USA. [10]Colorado State University, Fort Collins, CO, USA. [11]Princeton University, Princeton, NJ, USA. [12]University of Colorado, CIRES, Boulder, CO, USA. [13]NOAA Earth System Research Laboratory, Boulder, CO, USA. [14]Carnegie Mellon University, Pittsburgh, PA, USA. [15]Purdue University, West Lafayette, IN, USA. [16]University of Cincinnati, Cincinnati, OH, USA.
*Corresponding author. Email: ralvarez@edf.org

undersample 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 ~8000 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 to 10 kg CH_4/hour was two to seven 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 by means of downwind measurement but could not be accurately (or safely) measured by 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 (materials and methods, 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 (materials and methods, section S1.6). Two other factors may lead to low bias in EPA GHGI and similar inventory

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).

Industry segment	2015 CH_4 emissions (Tg/year)	
	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
Processing	0.72 (+0.20/−0.071)	0.44
Transmission and storage	1.8 (+0.35/−0.22)	1.4
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 (materials and methods, section S1.5).
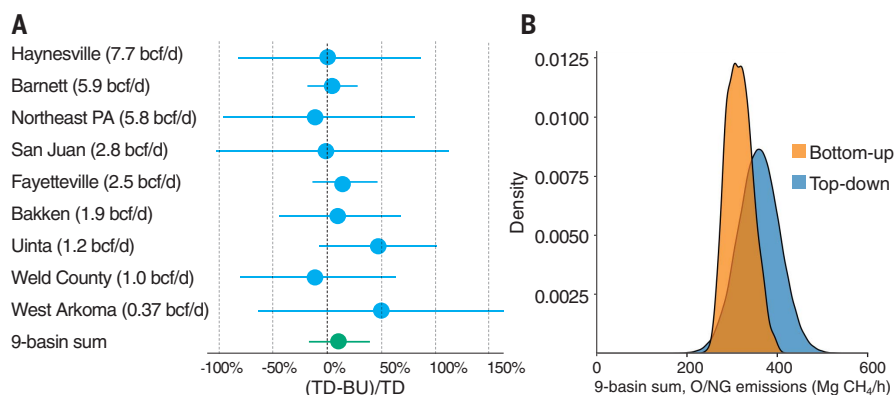†The GHGI only reports industry-wide uncertainties.

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 nine-basin sum of TD and BU mean estimates (blue and orange probability density, respectively). Neither the ensemble of TD-BU pairs (A) nor the nine-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%)].

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 more than twofold difference in production emissions (28).

The tail-heavy distribution for many O/NG CH_4 emission sources has important implications for mitigation because 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 (materials and methods, 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 ~2500 processing and compression facilities.

Our estimate of emissions from the U.S. O/NG supply chain (13 Tg CH_4/year) compares to the EPA estimate of 18 Tg CH_4/year for all other anthropogenic CH_4 sources (17). Natural gas losses are a waste of a limited natural resource (~\$2 billion/year), increase global levels of surface ozone pollution (30), and substantially 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/year 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) (materials and methods, 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 substantial 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.

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.
2. J. K. Shoemaker, D. P. Schrag, M. J. Molina, V. Ramanathan, Science 342, 1323–1324 (2013).
3. U.S. Energy Information Administration (EIA), "Annual Energy Outlook 2017" (EIA, 2017); www.eia.gov/outlooks/aeo/.
4. R. W. Howarth, R. Santoro, A. Ingraffea, Clim. Change 106, 679–690 (2011).
5. R. A. Alvarez, S. W. Pacala, J. J. Winebrake, W. L. Chameides, S. P. Hamburg, Proc. Natl. Acad. Sci. U.S.A. 109, 6435–6440 (2012).
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.
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.
8. A. R. Brandt et al., Science 343, 733–735 (2014).
9. D. T. Allen, J. Air Waste Manag. Assoc. 66, 549–575 (2016).
10. P. Balcombe, K. Anderson, J. Speirs, N. Brandon, A. Hawkes, ACS Sustain. Chem.& Eng. 5, 3–20 (2017).
11. J. A. Littlefield, J. Marriott, G. A. Schivley, T. J. Skone, J. Clean. Prod. 148, 118–126 (2017).
12. See supplementary materials.
13. Drillinginfo, Inc., Drillinginfo Production Query (2015); https://info.drillinginfo.com/.
14. K. McKain et al., Proc. Natl. Acad. Sci. U.S.A. 112, 1941–1946 (2015).
15. B. K. Lamb et al., Environ. Sci. Technol. 50, 8910–8917 (2016).
16. D. Wunch et al., Atmos. Chem. Phys. 16, 14091–14105 (2016).
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.
18. D. Zavala-Araiza et al., Proc. Natl. Acad. Sci. U.S.A. 112, 15597–15602 (2015).
19. C. W. Rella, T. R. Tsai, C. G. Botkin, E. R. Crosson, D. Steele, Environ. Sci. Technol. 49, 4742–4748 (2015).
20. M. Omara et al., Environ. Sci. Technol. 50, 2099–2107 (2016).
21. A. M. Robertson et al., Environ. Sci. Technol. 51, 8832–8840 (2017).
22. A. R. Brandt, G. A. Heath, D. Cooley, Environ. Sci. Technol. 50, 12512–12520 (2016).
23. D. Zavala-Araiza et al., Nat. Commun. 8, 14012 (2017).
24. D. R. Lyon et al., Environ. Sci. Technol. 50, 4877–4886 (2016).
25. A. L. Mitchell et al., Environ. Sci. Technol. 49, 3219–3227 (2015).
26. D. J. Zimmerle et al., Environ. Sci. Technol. 49, 9374–9383 (2015).
27. T. Howard, T. W. Ferrara, A. Townsend-Small, J. Air Waste Manag. Assoc. 65, 856–862 (2015).
28. R. A. Alvarez, D. R. Lyon, A. J. Marchese, A. L. Robinson, S. P. Hamburg, Elem. Sci. Anth. 4, 000137 (2016).
29. A. J. Marchese et al., Environ. Sci. Technol. 49, 10718–10727 (2015).
30. A. M. Fiore et al., Geophys. Res. Lett. 29, 21-1–25-4 (2002).
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.
32. A. P. Ravikumar, J. Wang, A. R. Brandt, Environ. Sci. Technol. 51, 718–724 (2017).
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.
34. Environmental Defense Fund (EDF), "Methane Detectors Challenge" (EDF, 2014); www.edf.org/energy/natural-gas-policy/methane-detectors-challenge.
35. J. D. Albertson et al., Environ. Sci. Technol. 50, 2487–2497 (2016).
36. D. J. Jacob et al., Atmos. Chem. Phys. 16, 14371–14396 (2016).

Science

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

Ramón A. Alvarez, 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

A leaky endeavor
Considerable amounts of the greenhouse gas methane leak from the U.S. oil and natural gas supply chain. Alvarez *et al.* reassessed the magnitude of this leakage and found that in 2015, supply chain emissions were ~60% higher than the U.S. Environmental Protection Agency inventory estimate. They suggest that this discrepancy exists because current inventory methods miss emissions that occur during abnormal operating conditions. These data, and the methodology used to obtain them, could improve and verify international inventories of greenhouse gases and provide a better understanding of mitigation efforts outlined by the Paris Agreement.
Science, this issue p. 186

ARTICLE TOOLS	http://science.sciencemag.org/content/361/6398/186
SUPPLEMENTARY MATERIALS	http://science.sciencemag.org/content/suppl/2018/06/20/science.aar7204.DC1
REFERENCES	This article cites 60 articles, 8 of which you can access for free http://science.sciencemag.org/content/361/6398/186#BIBL
PERMISSIONS	http://www.sciencemag.org/help/reprints-and-permissions

Use of this article is subject to the Terms of Service