ENERGY AND ENVIRONMENT

Methane Leaks from North American Natural Gas Systems

Methane emissions from U.S. and Canadian natural gas systems appear larger than official estimates.

A. R. Brandt,[1]*‡ G. A. Heath,[2] E. A. Kort,[3] F. O'Sullivan,[4] G. Pétron,[5,6] S. M. Jordaan,[7] P. Tans,[5]
J. Wilcox,[1] A. M. Gopstein,[8]† D. Arent,[2,9] S. Wofsy,[10] N. J. Brown,[11] R. Bradley,[12] G. D. Stucky,[13]
D. Eardley,[13] R. Harriss[14]

Natural gas (NG) is a potential "bridge fuel" during transition to a decarbonized energy system: It emits less carbon dioxide during combustion than other fossil fuels and can be used in many industries. However, because of the high global warming potential of methane (CH_4, the major component of NG), climate benefits from NG use depend on system leakage rates. Some recent estimates of leakage have challenged the benefits of switching from coal to NG, a large near-term greenhouse gas (GHG) reduction opportunity (1–3). Also, global atmospheric CH_4 concentrations are on the rise, with the causes still poorly understood (4).

To improve understanding of leakage rates for policy-makers, investors, and other decision-makers, we review 20 years of technical literature on NG emissions in the United States and Canada [see supplementary materials (SM) for details]. We find (i) measurements at all scales show that official inventories consistently underestimate actual CH_4 emissions, with the NG and oil sectors as important contributors; (ii) many independent experiments suggest that a small number of "supermitters" could be responsible for a large fraction of leakage; (iii) recent regional atmospheric studies with very high emissions rates are unlikely to be representative of typical NG system leakage rates; and (iv) assessments using 100-year impact indicators show system-wide leakage is unlikely to be large enough to negate climate benefits of coal-to-NG substitution.

Underestimation—Device to Continent

This study presents a first effort to systematically compare published CH_4 emissions estimates at scales ranging from device-level (>10^3 g/year) to continental-scale atmospheric studies (>10^{13} g/year). Studies known to us that (i) report measurement-based emissions estimates and (ii) compare those estimates with inventories or established emission factors (EFs) are shown in the first chart.

Studies that measure emissions directly from devices or facilities ("bottom-up" studies) typically compare results to emissions factors (EFs; e.g., emissions per device). Large-scale inventories are created by multiplying EFs by activity factors (e.g., number of devices).

Studies that estimate emissions after atmospheric mixing occurs ("atmospheric" studies) typically compare measurements to emissions inventories, such as the U.S. Envi-

[1]Stanford University, Stanford, CA. [2]National Renewable Energy Laboratory, Golden, CO. [3]University of Michigan, Ann Arbor, MI. [4]Massachusetts Institute of Technology, Cambridge, MA. [5]National Oceanic and Atmospheric Administration, Boulder, CO. [6]University of Colorado, Boulder, CO. [7]University of Calgary, Calgary, Canada. [8]U.S. Department of State, Washington, DC. [9]Joint Institute for Strategic Energy Analysis, Golden, CO. [10]Harvard University, Cambridge, MA. [11]Lawrence Berkeley National Laboratory, Berkeley, CA. [12]Independent consultant, Gaithersburg, MD. [13]University of California, Santa Barbara, Santa Barbara, CA. [14]Environmental Defense Fund, Boulder, CO.

*Full affiliations for all authors are included in the supplementary materials. †Views expressed in this article are those of the author and do not necessarily reflect those of the U.S. Department of State or the U.S. Government. ‡Corresponding author. abrandt@stanford.edu
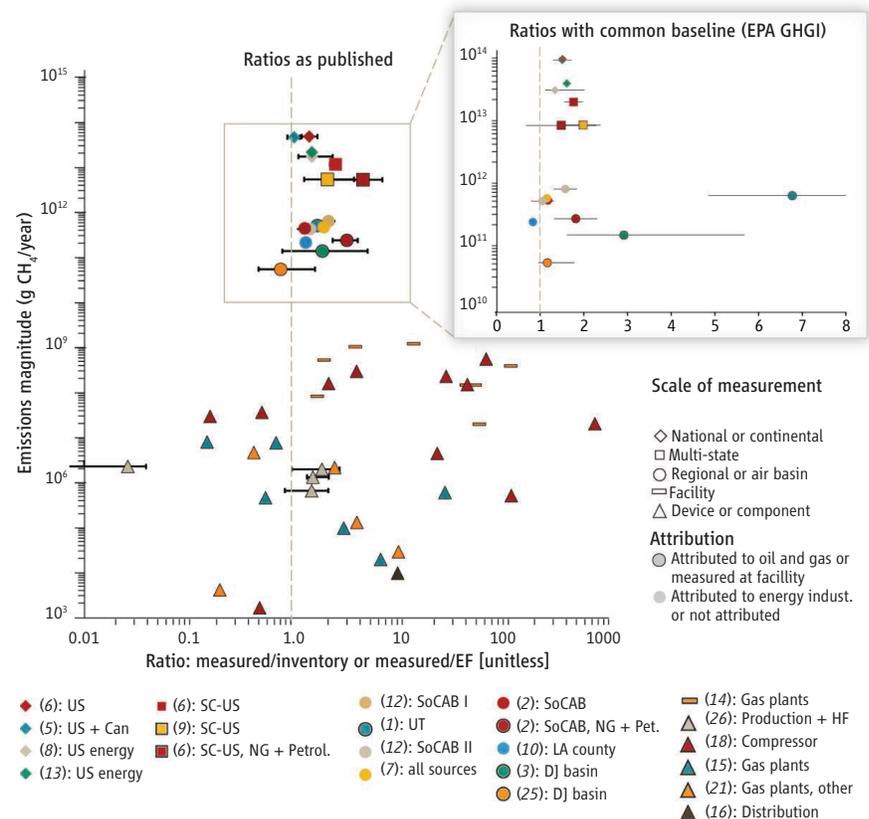
Inventories and emissions factors consistently underestimate actual measured CH_4 emissions across scales. Ratios >1 indicate measured emissions are larger than expected from EFs or inventory. Main graph compares results to the EF or inventory estimate chosen by each study author. Inset compares results to regionally scaled common denominator (17), scaled to region of study and (in some cases) the sector under examination. Multiple points for each study correspond to different device classes or different cases measured in a single study. Definitions of error bar bounds vary between studies. (US, United States; Can, Canada; SC, South Central; Petrol. and Pet., petroleum; SoCAB, South Coast Air Basin; LA, Los Angeles; DJ, Denver-Julesberg; UT, Utah; HF, hydraulic fracturing). See SM for figure construction details.
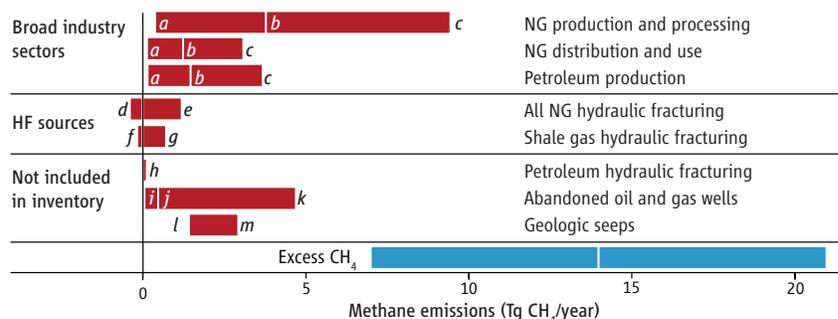
ronmental Protection Agency (EPA) national GHG inventory (GHGI). Atmospheric studies use aircraft (*1*, *5–8*), tower (*3*, *6*), and ground (*3*, *7–10*) sampling, as well as remote sensing (*7*, *11*, *12*). All such studies observe atmospheric concentrations and must infer fluxes by accounting for atmospheric transport. The various inference methods have strengths and weaknesses (see SM). The greatest challenge for atmospheric studies is attributing observed CH_4 concentrations to multiple potential sources (both anthropogenic and natural).

Results from bottom-up studies (generally <10^9 g CH_4/year) and atmospheric CH_4 studies at regional scale and larger (above 10^{10} g CH_4/year) are shown in the first chart. We also include studies that do not focus on NG systems, in order to place NG emissions in context with other CH_4 sources. Across years, scales, and methods, atmospheric studies systematically find larger CH_4 emissions than predicted by inventories. EFs were also found to underestimate bottom-up measured emissions, yet emissions ratios for bottom-up studies are more scattered than those observed in atmospheric studies (*13–16*).

Regional and multistate studies focusing on NG-producing (*1–3*, *9*) and NG-consuming regions (*2*, *7*, *10–12*) find larger excess CH_4 emissions than national-scale studies. This may be due to averaging effects of continental-scale atmospheric processes, to regional atmospheric studies focusing on areas with other air quality problems (*1*, *3*), or simply to methodological variation. Atmospheric measurements are constrained in spatial and temporal density: Regional studies cover 0.5 to 5% of NG production or consumption with dense measurements, although often limited to short-duration sampling "campaigns" (*3*, *7*); national studies cover wide areas with limited sample density (*6*) (table S5).

To facilitate comparison, the inset in the first chart normalizes atmospheric studies (>10^{10} g CH_4/year) to baselines computed from the most recent (2011) EPA GHGI estimates for the year and region in which study measurements were made (*17*). After normalization, the largest (e.g., national-scale) atmospheric studies (>10^{12} g CH_4/year) suggest typical measured emissions ~1.5 times those in the GHGI (*5*, *6*, *8*, *9*).

Why might emissions inventories be underpredicting what is observed in the atmosphere? Current inventory methods rely on key assumptions that are not generally satisfied. First, devices sampled are not likely to be representative of current technologies and practices (*18*). Production techniques

are being applied at scale (e.g., hydraulic fracturing and horizontal drilling) that were not widely used during sampling in the early 1990s, which underlies EPA EFs (*18*).

Second, measurements for generating EFs are expensive, which limits sample sizes and representativeness. Many EPA EFs have wide confidence intervals (*19*, *20*). And there are reasons to suspect sampling bias in EFs, as sampling has occurred at self-selected cooperating facilities.

Third, if emissions distributions have "heavy tails" (e.g., more high-emissions sources than would be expected in a normal distribution), small sample sizes are likely to underrepresent high-consequence emissions sources. Studies suggest that emissions are dominated by a small fraction of "superemitter" sources at well sites, gas-processing plants, coproduced liquids storage tanks, transmission compressor stations, and distribution systems (see table S6 and fig. S2). For example, one study measured ~75,000 components and found that 58% of emissions came from 0.06% of possible sources (*21*).

Last, activity and device counts used in inventories are contradictory, incomplete, and of unknown representativeness (*17*, *22*). Data should improve with increased reporting requirements enacted by EPA (*23*, *24*).

Source Attribution in Atmospheric Studies
Does evidence suggest possible sources of excess CH_4 emissions relative to official estimates within the NG sector? A key challenge is attribution of atmospheric observations to sources. Isotopic ratios (*7*, *11*) and prevalence signatures of non-CH_4 hydrocarbons (*3*, *6–8*) can be used to attribute emis-

sions to fossil sources rather than biogenic sources. Evidence from regional studies suggests that CH_4 emissions with fossil signatures are larger than expected (*3*, *6*, *7*, *9*, *11*), whereas national-scale evidence suggests a mix of biogenic and fossil sources (*6*). Atmospheric studies that control for biogenic CH_4 sources (*1*, *2*, *7*) are dependent on biogenic source estimation methods that also have high uncertainties (*6*). Natural geologic seeps could confound attribution (see the second chart and SM).

Studies can attribute emissions to liquid petroleum and NG sources rather than coal by sampling in places with little coal-sector activity (*2*, *3*, *6*, *7*, *9*). Attributing leakage to the NG system, as defined by EPA industry sector classifications, is more challenging. Alkane fingerprints may allow attribution to oil-associated NG (*9*), although NG processing changes gas composition, which may complicate efforts to pinpoint leakage sources. Geographic colocation of facilities and sampling, along with geographically isolating wind directions (*2*, *3*, *7*), can allow attribution of emissions to NG subsectors. Without spatial isolation, sector attribution can require assumptions about gas composition that introduce significant uncertainty (*2*, *3*, *25*).

We plotted results of a thought experiment (see the second chart) in which we estimated emissions ranges of selected possible sources within the NG sector, as well as sources that could be mistaken for NG emissions owing to chemical and isotopic signatures. Although such an analysis is speculative given current knowledge, it illustrates ranges of possible source magnitudes.

Potential contributions to total U.S. CH_4 emissions above EPA estimates. EPA estimate in blue, based on central estimate and uncertainty range from large-scale studies from the inset in the first chart. Both NG sources and possible confounding sectors are included. NG production, petroleum production, and NG distribution emissions are based on regional empirical studies (*1*, *2*, *6*), which estimate emissions rates from high-emitting sources but do not estimate prevalence. Scenarios (*a*) to (*c*) correspond to 1, 10, and 25% of gas production or consumption from such high-emitting sources. Ranges (*d*) to (*g*) correspond to estimates for flowback emissions rates during hydraulic fracturing (HF) of all gas wells and shale gas wells, relative to EPA estimates. Ranges (*h*) to (*m*) reflect sources not included in EPA CH_4 inventories but which could be mistaken for NG emissions by chemical or isotopic composition. See SM for details.

We include in the second chart a range of excess CH_4 from all sources (7 to 21 × 10^{12} g or Tg/year) based on normalized national-scale atmospheric studies from the inset in the first chart. This excess is conservatively defined as 1.25 to 1.75 times EPA GHGI estimates. This estimate is derived from national-scale atmospheric studies and includes all sources of CH_4 emissions: It should not be expected that NG sources are responsible for all excess CH_4.

The scenarios in the second chart for NG production and/or processing, distribution, and petroleum system emissions apply observed leakage rates from the literature that are higher than EPA GHGI estimates (*1*, *2*, *7*). The frequency of such high-emitting practices is unknown, so illustrative prevalence scenarios are plotted: 1, 10, or 25% of activity is represented by high-emitters; the remaining facilities emit at EPA GHGI rates. This evidence suggests that high leakage rates found in recent studies (*1*, *2*, *7*) are unlikely to be representative of the entire NG industry; if this were the case, associated emissions would exceed observed total excess atmospheric CH_4 from all sources.

In general, the wide ranges in the second chart suggest a poor understanding of sources of excess CH_4 and point to areas where improved science would reduce uncertainty. However, hydraulic fracturing for NG is unlikely to be a dominant contributor to total emissions (*26*). Also, some sources not included in the GHGI may contribute to measured excess CH_4, e.g., abandoned oil and gas wells and geologic seeps (see SM).

Policy Challenges and Opportunities

Leakage scenarios in the second chart have implications for decision-making and policy. A key tool for environmental decision-making is life-cycle assessment (LCA), which compares impacts associated with varying methods of supplying a useful product (e.g., kWh of electricity). A key challenge in LCA studies is attribution of emissions from systems that produce two products, such as "gas" wells that also produce hydrocarbon liquids, or "oil" wells that also produce NG. This challenge is complicated by incongruence between LCA methodology and EPA sector definitions (see SM).

Recent LCAs have estimated GHG emissions from NG use in power generation and transport (see SM). LCA studies generally agree that replacing coal with NG has climate benefits (*27*). However, LCAs have relied heavily on EPA GHGI results. Updating these assessments with uncertainty ranges from the second chart (see SM) still supports robust climate benefits from NG substitution for coal in the power sector over the typical 100-year assessment period. However, climate benefits from vehicle fuel substitution are uncertain (gasoline, light-duty) or improbable (diesel, heavy-duty) (*28*). These conclusions may undercount benefits of NG, as both EPA GHGI methods and many regionally focused top-down studies attribute CH_4 emissions from coproducing NG systems to the NG sector, rather than to a mixture of oil and NG sources.

How can management and policy help address the leakage problem? Opportunities abound: Many solutions are economically profitable at moderate NG prices, with some technologies already being adopted or to be required in regulation (*23*, *26*) (e.g., reduced emissions completions). Facility studies using existing technology have found leakage detection and repair programs to be profitable (*21*).

The heavy-tailed distribution of observed emissions rates presents an opportunity for large mitigation benefits if scientists and engineers can develop reliable (possibly remote) methods to rapidly identify and fix the small fraction of high-emitting sources.

However, this heterogeneity also creates challenges in formulating statistical distributions for use in inventories. Approaches that assume "typical" emissions rates for this industry are inherently challenged. Inventories can be improved through efforts to better characterize distributions and by incorporating flexibility to adapt to new knowledge.

Improved science would aid in generating cost-effective policy responses. Given the cost of direct measurements, emissions inventories will remain useful for tracking trends, highlighting sources with large potential for reductions, and making policy decisions. However, improved inventory validation is crucial to ensure that supplied information is timely and accurate. Device-level measurements can be performed at facilities of a variety of designs, vintages, and management practices to find low-cost mitigation options. These studies must be paired with additional atmospheric science to close the gap between top-down and bottom-up studies. One such large study is under way (*29*), but more work is required.

If natural gas is to be a "bridge" to a more sustainable energy future, it is a bridge that must be traversed carefully: Diligence will be required to ensure that leakage rates are low enough to achieve sustainability goals.

References and Notes

1. A. Karion *et al.*, *Geophys. Res. Lett.* **40**, 4393 (2013).
2. J. Peischl *et al.*, *J. Geophys. Res.* **118**, 4974 (2013).
3. G. Pétron *et al.*, *J. Geophys. Res.* **117**, (D4), D04304 (2012).
4. E. G. Nisbet *et al.*, *Science* **343**, 493 (2014).
5. E. A. Kort *et al.*, *Geophys. Res. Lett.* **35**, L18808 (2008).
6. S. M. Miller *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **110**, 20018 (2013).
7. P. O. Wennberg *et al.*, *Environ. Sci. Technol.* **46**, 9282 (2012).
8. Y. Xiao *et al.*, *J. Geophys. Res.* **113**, (D21), D21306 (2008).
9. A. S. Katzenstein *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **100**, 11975 (2003).
10. Y.-K. Hsu *et al.*, *Atmos. Environ.* **44**, 1 (2010).
11. A. Townsend-Small *et al.*, *J. Geophys. Res.* **117**, (D7), D07308 (2012).
12. D. Wunch *et al.*, *Geophys. Res. Lett.* **36**, 10.1029/2009 GL039825 (2009).
13. J. S. Wang *et al.*, *Global Biogeochem. Cycles* **18**, 10.129/2003GB0021680 (2004).
14. A. Chambers, *Optical Measurement Technology for Fugitive Emissions from Upstream Oil and Gas Facilities* (Alberta Research Council, Edmonton, AB, 2004).
15. Clearstone Engineering, *Identification and Evaluation of Opportunities to Reduce Methane Losses at Four Gas Processing Plants* (Gas Technology Institute, Des Plaines, IL, 2002).
16. Innovative Environmental Solutions, *Field Measurement Program to Improve Uncertainties for Key Greenhouse Gas Emissions Factors for Distribution Sources* (Gas Technology Institute, Des Plaines, IL, 2009).
17. EPA, "Inventory of U.S. greenhouse gas emissions and sinks: 1990–2011" (EPA, 2013).
18. M. R. Harrison *et al.*, *Natural Gas Industry Methane Emissions Factor Improvement Study* (EPA, 2011).
19. K. E. Hummel *et al.*, *Methane Emissions from the Natural Gas Industry*, vol. 8, *Equipment Leaks* (EPA, 1996).
20. H. J. Williamson *et al.*, *Methane Emissions from the Natural Gas Industry*, vol. 4, *Statistical Methodology* (EPA, 1996).
21. National Gas Machinery Laboratory, Clearstone Engineering, Innovative Environmental Solutions, *Cost-Effective Directed Inspection and Maintenance Control Opportunities at Five Gas Processing Plants and Upstream Gathering Compressor Stations and Well Sites* (EPA, 2006).
22. Office of Inspector General, EPA, *EPA Needs to Improve Air Emissions Data for the Oil and Natural Gas Production Sector* (EPA, 2013).
23. EPA, *Fed. Regist.* 77(159), 49490 (16 August 2012); 40 Code of Federal Regulations, Parts 60 and 63.
24. EPA, *Greenhouse Gas Reporting Program*, subpart W, *Petroleum and Natural Gas Systems* (EPA, 2013).
25. M. A. Levi, *J. Geophys. Res.* **117**, (D21), 16 (2012).
26. D. T. Allen *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **110**, 17768 (2013).
27. C. L. Weber, C. Clavin, *Environ. Sci. Technol.* **46**, 5688 (2012).
28. R. A. Alvarez *et al.*, *Proc. Natl. Acad. Sci. U.S.A.* **109**, 6435 (2012).
29. Environmental Defense Fund, Natural gas: EDF is fighting for tough rules and enforcement (EDF, 2013); www.edf.org/climate/natural-gas.

Supplementary Materials
www.sciencemag.org/content/343/6172/733/suppl/DC1

10.1126/science.1247045