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Supplementary Materials for

Methane Leaks from North American Natural Gas Systems

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Published 14 February 2014, *Science* **343**, 733 (2014)
DOI: 10.1126/science.1247045

This PDF file includes

Materials and Methods
Supplementary Text
Figs. S1 to S3
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Other Supplementary Material for this manuscript includes the following:
(available at www.sciencemag.org/content/343/6172/733/suppl/DC1)

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1 Materials and Methods

1.1 Study goals, scope, and methods

This study synthesizes the state of knowledge of leakage of gas from natural gas (NG) systems in North America. Gas leakage is a concern for greenhouse gas (GHG) emissions because the primary component of NG is methane (CH_4), which has a significant global warming potential (GWP). Implications of this state of knowledge for policy are also discussed. More than 200 technical publications were reviewed for this study.

The scientific subquestions we examine include the following:

1. What do we know about device- or facility-level leakage rates? (e.g., what do we know about NG leakage from hydraulic fracturing?)
2. What uncertainties exist in building inventories of CH_4 emissions?
3. What do top-down atmospheric observations of CH_4 concentrations at various scales tell us about NG leakage rates?
4. What are the key drivers behind disparity between top-down and bottom-up estimates of leakage?
5. Can we specifically define and estimate excess emissions to sources within the NG sector?

The policy subquestions we address include the following:

1. What are the policy implications of the current state of scientific understanding of methane emissions from the petroleum sector and, specifically, NG?
2. Does methane leakage affect the "cutoffs" for appropriate NG fuel-switching options for climate mitigation? When is coal substitution favorable? When is diesel or gasoline substitution favorable?

This study assesses all NG segments where CH_4 is emitted to the atmosphere. We assess fugitive emissions (unintentional) and vented emissions (intentional), both routine and nonroutine. We also examine the extent to which other CH_4 sources (e.g., petroleum operations and solid waste facilities) may interfere with attribution of CH_4 concentrations to NG systems.

All types of NG are analyzed. To the extent that coproduced, oil-associated gas (associated gas) supplies ~20% of U.S. gross NG withdrawals (30), we examine gas coproduction at oil fields. We pay particular attention to changes in emission rates resulting from shifts from conventional resources to shale gas. We focus primarily on the U.S. gas system, with some studies including data on Canadian emissions.

1.2 Definitions

Bottom-up studies: These studies use direct measurements of emissions of gas at the device or facility level. Also, they refer to emissions inventories and life-cycle assessment (LCA) studies based on bottom-up data.

Top-down studies: Studies that use measurements of emissions at facility to national scales, typically taken at a location remote from individual pieces of equipment, such that atmospheric effects integrate emissions from multiple devices. Such studies require interpretation or modeling to assign or attribute emissions to sources such as the NG system. Facilities can be considered measurable via both top-down and bottom-up approaches, as estimates for facility level emissions can be made through exhaustive bottom-up measurement of all emission sources in a facility or via down-wind atmospheric studies by using tracer-based or plume transect approaches.

Hydrocarbons: Liquid and gaseous fossil fuels comprised of (primarily) C and H. These fluids are produced from hydrocarbon production wells, generally in conjunction with each other (e.g., "gas wells" generally produce some liquid hydrocarbons, while "oil wells" produce some gaseous hydrocarbons). Unless specifically noted, coal is not included in the current study.

1.3 Units, constants, and conversion factors

The studies examined in this review report emissions in a variety of units. In order to allow comparisons between studies, all estimates are converted to grams (g) of CH_4 emitted (e.g., Tg CH_4/year). In some cases, results will be discussed in "field units" [e.g., standard cubic foot (SCF)]. Standard conditions in this report are defined as 14.73 psia and 60 °F, resulting in 1.198 moles of ideal gas per SCF.

For clarity, the meaning of field units and their form used in this study (main text, SI) are listed in Table S1. Conversion factors used in converting field units to g of CH_4 are taken from EPA average compositions, listed in Table S2, Table S3, and Table S4.

Table S1. Field units from NG industry

Unit	Meaning	Numerical defn.
MSCF	Thousand standard cubic feet	10^3 ft^3 at standard conditions
MMSCF	Million standard cubic feet	10^6 ft^3 at standard conditions
BCF	Billion standard cubic feet	10^9 ft^3 at standard conditions
TCF	Trillion standard cubic feet	10^{12} ft^3 at standard conditions

Table S2. Conversion factors and constants used

Quantity	Value	Units
Moles per SCF (ideal gas)	1.1980	mol/SCF
Molecular weight CH_4	16	g/mol
Mass of CH_4 per SCF CH_4	19.17	g CH_4/SCF CH_4
SCF per m^3	35.315	SCF/m^3

Table S3. EPA gas composition by NG production stage. Source: EPA 2011 inventory (*17*), annex 3, table A-128, and pp. A178–A179.

Stage - Quantity	Value	Unit
Production	0.841	mol CH_4/mol gas
Processing	0.870	mol CH_4/mol gas
Transmission	0.934	mol CH_4/mol gas
Distribution	0.934	mol CH_4/mol gas

Table S4. EPA mass-to-volume conversion factors. Source: computed from above.

Stage - Quantity	Value	Unit
Production	62.03	MMSCF NG/Gg CH_4
Processing	59.96	MMSCF NG/Gg CH_4
Transmission	55.85	MMSCF NG/Gg CH_4
Distribution	55.85	MMSCF NG/Gg CH_4

1.4 Calculations

1.4.1 First text chart: Comparison of measured emissions rates to inventories or emissions factors

The first text chart compiles data from all available studies where a measured emissions rate from a given device, facility, air basin, or region, is compared to an inventory-based estimate or emissions factor (EF). The main part of the chart plots only the comparisons made by study authors themselves to the inventory or emissions factor that the authors deemed most comparable. We compute this comparison as a ratio of observed emissions over inventoried emissions (or over EF values) although not all authors express their own results using this ratio. The inventory or emissions factor used for comparison differs between studies (e.g., California Air Resources Board inventory, as compared to EPA GHG inventory, etc.). Study results are not corrected or modified in the main part of the first text chart to account for interpretation or attribution errors made by the study authors. We note instances where challenges in interpretation or comparison exist with a given study.

Measurements in these studies occurred across orders of magnitude of spatial scale (compressor seals to continents) and temporal scale (seconds to decades). We display this variability on the *y* axis of the first text chart by plotting results by measured rate of emissions.

The following data were collected for each study:

- Author;
- Institution;
- Study region or boundary;
- Location of study;
- Measurement methods;
- Modeling methods;
- Data vintage;
- Noted limitations;
- Natural gas stage(s) studied;
- Type(s) of NG;
- Gas species measured;
- Units reported;
- Emissions mechanism(s) studied (if applicable);
- Measured quantity of emissions (with specified high and low ranges). In studies where multiple cases were constructed, each case was recorded with its appropriate ranges;
- Type of uncertainty range, if specified (e.g., 95% confidence interval);
- Other notes;
- Emissions estimate from inventory or EF approach that was used for comparison by study author;
- Notes on comparable inventory or EF figure.

Some studies include multiple cases. Cases can differ based on the region included, the sources included, or the assumptions underlying the results.. All cases examined were included as long as they met the study criteria listed in the previous section.

These data are recorded in Data File S1 in the "Methods comparison" and "Calc – Figure 1 - Main" worksheets.

For each study and case, we compute the ratio of measured emissions to the inventory or emissions factor that the author uses for comparison (see first chart in the main text). This ratio is plotted as the *x*-axis variable of the first chart for each study. If uncertainty ranges are available for both the measured emissions and the comparable inventory or EF, three ratios are constructed:

- Central ratio: central estimate from study is compared to central estimate from the comparable inventory or EF.
- High ratio: High estimate from study is compared to low estimate of the inventory or EF.

- Low ratio: Low emissions result from study is compared to high emissions estimate of comparable inventory or EF.

If comparable inventory or EF estimate is presented as a point value, than this single point value is used as the denominator of all three ratios. If both the measured value and the comparable inventory or EF are presented as point estimates, then a simple point estimate of the ratio is computed.

1.4.1.1 Sources of data and calculation methods for each study

For each study, we outline the source of data and any calculation methods or assumptions below, as required to generate the results shown in first text chart.

These ratios are plotted in the main part of the first chart where the *y*-axis plots the scale of the measured emissions magnitude, in g CH_4 per year. Because the scale of measured emissions rates ranges from kg to Tg, a logarithmic scale is used. Similarly, a logarithmic scale is used on the *x* axis because of very large variation in ratios (0.01 to 1000). These data and calculations are recorded in Data File S1 in the "Calc – Figure 1 - Main" worksheet.

Miller et al.

Miller *et al*. (*6*) create three cases that we include in the first text chart. See below for discussion of the experimental and analytical methods used in Miller *et al*.

Case 1 estimates emissions for all sources across the entire United States. For this case, Miller *et al*. estimate emissions of 33.4 (±1.5) TgC as CH_4/year, as gathered from figure 1 [BPO1]in the original paper (and author communication). This is equivalent to to 45 (±1.9) Tg CH_4/year. Miller *et al*. compare their emissions estimate to both the EPA GHG inventory (GHGI) and the EDGAR spatial emissions database for the time and region of interest. We only utilize the EPA estimate in this study. The comparable estimate from the EPA inventory is 22.1 (±2.9) Tg CH_4/year. This leads to a central estimate for the ratio of measured/estimated to be 1.5 (+0.3, –0.23) g/g.

Case 2 estimates emissions for the South-Central United States (SC-US). For this case, Miller *et al*. estimate emissions of 8.1 (±1) TgC as CH_4/year, as gathered from article text (and author communication) in the original paper. This is equivalent to 11 (±1.3) Tg CH_4/year. Miller *et al*. compare their emissions estimate to the EDGAR spatial emissions database for the region of interest. The comparable estimate from the EDGAR case is 3 Tg CH_4/year. This leads to a central estimate for the ratio of measured/estimated to be 2.7 (+0.3, –0.3) g/g.

Case 3 estimates oil- and gas-related emissions for the South-Central United States (SC-US: Fossil). For this case, Miller *et al*. estimate emissions of 3.7 (±2) TgC as CH_4/year, as gathered from article text (and author communication) in the original paper. This is

equivalent to 4.9 (±2.6) Tg CH_4/year. Miller *et al*. compare their emissions estimate to the EDGAR spatial emissions database for the region of interest for oil and gas sources. The comparable estimate from the EDGAR case is 0.75 Tg CH_4/year. This leads to a central estimate for the ratio of measured/estimated to be 4.9 (±2.6) g/g.

Kort et al.

Kort *et al*. (*5*) create one case that we include in the first text chart. See below for discussion of the methods used in Kort *et al*. The Kort *et al*. study estimates CH_4 emissions for North America (United States and Canada). Kort *et al*. report emissions of 1.08 (±0.15) g/g, measured as a multiple of EDGAR v3.2 estimates for the region. These results are gathered from p. 4 in the original source. The comparable estimate from EDGAR v3.2 is 30 TgC as CH_4/year. This estimate is equivalent to 43.2 (±6) Tg CH_4/year, given the EDGAR baseline emissions for the year of data collected.

It should be noted that due to the large variability in EDGAR emissions estimates, and between EDGAR and EPA baseline emissions estimates, the multiple of 1.08 g/g found by Kort *et al*. with EDGAR 3.2, becomes significantly poorer agreement when compared to EDGAR v4.2 or recent EPA estimates. This is seen in the inset in the first text chart and is described below.

Katzenstein et al.

Katzenstein *et al*. (*9*) create one case that we include in the first text chart. See below for discussion of the methods used in Katzenstein *et al*. The Katzenstein *et al*. case estimates emissions for the South-Central United States, defined as Texas, Oklahoma, and Kansas. They report emissions of 4 (±1) Tg CH_4/year, as gathered from p. 11978 in the original source. Katzenstein *et al*. compare their emissions estimate to emissions from the U.S. oil and gas industries. The comparable estimate, implied by their assumptions, is 2.2 ±0.7 Tg CH_4/year. This leads to a central estimate for the ratio of measured/estimated to be 2.3 +1.8, -0.9 g/g.

Katzenstein *et al*. did not compute the ratio above, but instead they imply the comparison in text on p. 11978 of the original source (*9*). The article text leaves ambiguous whether their CH_4 emissions estimate should be compared to NG emissions, NG and petroleum emissions, or all emissions in the region of interest. Our interpretation of the text suggests that Katzenstein *et al*. meant their result to be comparable to NG emissions from the region of interest.

Wang et al., cited in Xiao et al.

Wang *et al*. (*13*) create one case that we include in the first text chart. Xiao *et al*. cite an estimate from Wang *et al*. of emissions for the U.S. energy industries. They estimate emissions of 20 TgCH_4/year, as gathered from paragraph 5 in Xiao *et al* (*8*). Xiao *et al*.

compare the emissions estimate from Wang *et al*. to the EPA GHGI from 2007/2008 for the time and region of interest. The comparable emissions quantity is estimated by Xiao *et al*. to be ~10 $TgCH_4$/year (see Xiao *et al*. para. 5 and 40) (*8*). This leads to a central estimate for the ratio of measured/estimated emissions of 1.9 g/g.

The Wang *et al*. estimate for U.S. energy industry emissions is cited as a personal communication in Xiao *et al*. (*8*). The original source of the emissions estimate was not found in the Wang *et al*. paper. Therefore, it is likely that the regional result presented here was computed as part of the global analysis in the cited paper, but not originally published as a region-specific result.

Xiao et al.

Xiao *et al*. (*8*) create three cases that we include in the first text chart as a median case with high-low ranges. See below for discussion of the methods used in Xiao *et al*. Each case estimates emissions for total CH_4 emissions from energy industries in the United States, using C_2H_6 emissions as modeled in their study. They report emissions of 2.4 Tg C_2H_6 /year, which can be converted to three estimates of CH_4 emissions using three CH_4/C_2H_6 ratios presented in table 4 and paragraph 40 in the original source. We convert these emissions to 13.3, 16.0, and 24.0 Tg CH_4/year (see Data file S1 for details). Xiao *et al*. compare their emissions estimate to a round estimate of ~10 Tg CH_4/year for the time and region of interest, using results for the year being modeled from two different EPA GHGIs. This leads to a central estimate for the ratio of measured/estimated emissions of 1.6 (+0.9, –0.4) g/g.

This study is an important piece of corroborating evidence from other studies, as it is a study of C_2H_6 (rather than a direct CH_4 measurement). Whereas there is uncertainty in the CH_4/C_2H_6 ratios used to model CH_4 emissions, the use of C_2H_6 has some advantages in that it rules out biogenic sources of CH_4.

Pétron et al.

Pétron *et al*. (*3*) create three cases that we include in the first text chart as a median case with high-low ranges (as recommended by study author G. Pétron through personal communication). See below for discussion of the methods used in Pétron *et al*. This study estimates emissions from the NG and petroleum industry in the Denver-Julesberg basin. They report emissions of 71.6 to 251.9 Gg CH_4/year, as gathered from table 4 in the original source. Using suggested cases from original study author (Pétron) we choose low, med, and high case estimates of 0.071, 0.129, and 0.259 Tg CH_4/year, respectively. Pétron *et al*. compare their emissions estimate to emissions estimated in the Western Regional Air Partnership (WRAP) air pollutant inventory for the time and region of interest. The comparable estimate is 64.3 (46–86) Gg CH_4. This leads to a central estimate for the ratio of measured/estimated of 2.0 g/g.

Levi

Levi (*25*) creates a number of cases in his assessment that we include in the first text chart. See below for discussion of the methods used in Levi. In general, Levi reinterprets the results from the Pétron *et al*. study to estimate emissions for the Denver Julesberg basin. He reports a range of emissions estimates in table 2 in the original source, which range from 46.4 to 58.8 Gg CH_4/year. For our comparison, we construct ranges as follows. Our central estimate is given by the average of the four "expected" values [min and max ranges for both Boulder Atmospheric Observatory (BAO) and mobile lab]. Our low estimate is given by lowest "expected" value less the lower bound "conservative error" quantity (mobile lab minimum). Our high estimate is highest expected value plus the "conservative error" upper bound (BAO maximum). Levi compares their[BPO2] emissions estimate to the same WRAP emissions inventory for the time and region of interest. This leads to a central estimate for the ratio of measured/estimated to be 0.8 (– 0.3, + 0.9) g/g.

Wunch et al.

Wunch *et al*. (*12*) create two cases that we include in the first text chart. See below for discussion of the methods used in Wunch *et al*. Both cases estimate emissions for the South Coast Air Basin (SoCAB). The two cases differ in the gas species that they use to scale observations of CH_4 concentrations to obtain fluxes of CH_4. In one case, they use the ratio of CO_2 to CH_4 in well-mixed air along with an average of EDGAR and CARB CO_2 emissions estimates to provide an estimate of CH_4 fluxes. In the other case, they use the CARB CO inventory to estimate CH_4 fluxes. They report emissions of 0.6 (±1) Tg CH_4/year and 0.4 (±1) Tg CH_4/year in these two cases, as presented in table 2 in the original source. Wunch *et al*. compare their emissions estimate to an urban-region-focused, population-scaled modification of the CARB CH_4 inventory for the time and region of interest. The comparable estimate is 0.26 Tg CH_4/year. This leads to a central estimate for the ratio of measured/estimated to be 2.3 g/g and 1.5 g/g in the CO_2-scaled and CO-scaled cases, respectively.

Hsu et al.

Hsu *et al*. (*10*) create one case that we include in the first text chart. See below for discussion of the methods used in Hsu *et al*. This case estimates emissions for Los Angeles County within SoCAB. They report emissions of 4.2 (±0.12) Mt CO_2 eq./year, as gathered from table 3 in the original source. We convert this estimate to 0.2 (±0.01) Tg CH_4/year using the same GWP used by Hsu *et al*. (21 g CO_2 eq./g CH_4). Hsu *et al*. compare their emissions estimate to the CARB CH_4 inventory for the time and region of interest. The comparable estimate is 3 Mt CO_2 eq/year. This leads to a central estimate for the ratio of measured/estimated of 1.4 g/g.

Wennberg et al.

Wennberg *et al*. (*7*) create one case that we include in the first text chart. See below for discussion of the methods used in Wennberg *et al*. This cases estimates emissions in the SoCAB for all sources of CH_4. They report emissions of 0.44 Tg CH_4/year, as gathered from table 3 in the original source. Wennberg *et al*. compare their emissions estimate to a modified version of the CALGEM spatial inventory for the time and region of interest. The comparable estimate is 0.212 Tg CH_4/year, respectively. This leads to a central estimate for the ratio of measured/estimated of 2.1 g/g.

Wennberg *et al*. also compute a "maximum possible" emissions rate from the NG industry, which is not significantly less than their total emissions rate (0.39 Tg CH_4/year). Because this is meant to represent an upper-bound constraint on possible emissions from NG, rather than a best estimate of the magnitude, we do not include this result in our comparison.

Peischl et al.

Peischl *et al*. (*2*) create two cases that we include in the first text chart. See below for discussion of the methods used in Peischl *et al*. Their cases estimate emissions of CH_4 into the SoCAB from all CH_4 sources, as well as from NG and petroleum sources. In an advance from previous work, they create estimates for subsources of gas within the NG and petroleum sectors, including pipeline-quality dry gas, and unprocessed raw gas. They report emissions of 411 (±37) Gg CH_4/year for all sources and 223 (±61) Gg CH_4/year for NG emissions, respectively. Because Peischl *et al*. estimate a variety of subsources, we include the following sources in our estimate of NG emissions: "pipeline quality dry NG/Local seeps" and "Local NG." These results are gathered from table 4 in the original source. Peischl *et al*. compare their emissions estimate to the CARB 2009 GHG inventory for the region of interest. The comparable estimates from the CARB inventory are 301 Gg CH_4/year and 64.6 Gg CH_4/year, respectively. This leads to a central estimate for the ratio of measured/estimated of 1.4 g/g for the case of all emissions sources and 3.5 g/g for the NG sources. We refer to this case as a "NG + petroleum" case, as the nature of NG production in SoCAB is almost entirely associated with oil production, which EPA would classify then as a "petroleum" sector source.

Karion et al.

Karion *et al*. (*1*) create one case that we include in the first text chart. See below for discussion of the methods used in Karion *et al*. The case of interest estimates emissions for NG operations in the Uintah basin, UT. They report CH_4 emissions of 54,600 (±15500) kg/h, as gathered from paragraph 15 in the original source, equal to 0.48 (±0.14) Tg CH_4/year. Karion *et al*. compare their emissions estimate to the WRAP

inventory for the time and region of interest. The comparable estimate is 30,333 kg/hour. This leads to a central estimate for the ratio of measured/estimated to be 1.80 g/g.

Chambers

Chambers (*14*) creates eight cases that we include in the first text chart. See below for discussion of the methods used in Chambers. Chambers estimates emissions for tanks, flares, fugitive emissions, and glycol dehydrators. He reports emissions of 19.3 to 1165 t CH_4/year as gathered from tables 18 and 21 in the original source. Chambers compares his emissions estimate to Canadian Association of Petroleum Producers (CAPP) detailed EF estimates for the technologies of interest. The comparable estimates using CAPP EF methods range from 0.3 to 240 tCH_4/year. This leads to a central estimate for the ratio of measured/estimated which range from 2.1 g/g to 128.6 g/g.

The full details for Chamber's eight estimates can be found in Data File S1.

Harrison et al.

Harrison *et al*. (*18*) create fifteen cases that we include in the first text chart. These cases include estimates for the following sources. We also report the total number of components screened.

- Valves: 1634 components screened
- Flanges: 1244 components screened
- Centrifugal compressor - Average BD vent for run: 3 components screened
- Centrifugal compressor - Average BD vent for idle + run: 11 components screened
- Centrifugal compressor - Wet seal (run): 9 components screened
- Reciprocating transmission compressor - Average BD vent for idle + pressurized: 10 components screened
- Reciprocating transmission compressor - Average BD vent for run: 6 components screened
- Reciprocating transmission compressor - Average BD vent for idle + depressurized: 15 components screened
- Reciprocating transmission compressor - Average rod packing for idle + pressurized: 5 components screened
- Reciprocating transmission compressor - Average rod packing for run: 2 components screened
- Reciprocating boosting compressors - Average BD vent for run: 16 components screened
- Reciprocating boosting compressors - Average BD vent for idle + depressurized: 8 components screened
- Reciprocating boosting compressors - Average PRV vent for run: 12 components screened

- Reciprocating boosting compressors - Average PRV vent for idle + depressurized: 6 components screened
- Reciprocating boosting compressors - Average rod packing – Run: 15 components screened

See below for discussion of the methods used in Harrison *et al*. They report emissions ranging from 0 to 29693 MSCF/year, as gathered from tables 3-2, 3-3, 3-4, and 3-5 in the original source. These emissions factors are for unspecified gas composition, but it appears that the Hi-Flow sampler is used to estimate emissions of CH_4 on a volumetric basis (e.g. discussion of CH_4 concentration calibration in report p. 35 and units reported in headings in table 3.3). However, this is not entirely clear, as some Harrison *et al*. results tables do not specify whether units are MSCF/year of CH_4 or are MSCF/year of total hydrocarbons. Harrison *et al*. compare their emissions estimate to relevant EPA/GRI emissions factors for the technology of interest. The comparable volumetric EPA EFs range from 0 to 9352 MSCF/year.

The comparable EPA emissions factors are presented by Harrison *et al*. in identical volumetric units (MSCF/year). As the Hi-Flow sampler results appear to be reporting the volume of CH_4 leakage (e.g., MSCF CH_4/year rather than MSCF total hydrocarbons/year), Harrison *et al*. should be comparing their results to MSCF/year of CH_4 emissions, not MSCF of total hydrocarbon emissions. It appears that their comparable EPA EFs are also reported on a CH_4 volumetric basis (e.g., Harrison *et al*. cite EPA/GRI 1996, Vol. 8, table 4-15, which adjusts total leakage volume to account for 93.4 mol% CH_4 in transmission quality gas). In this case, comparing standard volumes of CH_4 emissions allow for either volume/volume or mass/mass (g/g) ratios to be computed.

Clearstone

Clearstone Engineering Ltd. (*15*) create 12 cases that we include in the first text chart. See below for discussion of the methods used in the Clearstone report. The Clearstone report estimates emissions for the following devices:

- Connector: 82146 components screened
- Block valves: 15136 components screened
- Control valves: 1240 components screened
- Pressure relief valves: 385 components screened
- Pressure regulators: 169 components screened
- Orifice meters: 167 components screened
- Other flow meters: 7 components screened
- Crank case vents: 36 components screened
- Open ended lines: 1610 components screened
- Pump seals: 83 components screened
- Compressor seals: 206 components screened
- Blowdowns: 6 components screened

The Clearstone report estimate emissions factors ranging from 0.002 to 0.883 kg total hydrocarbons/h/source, as gathered from table 5 (p. 28) in the original source. Clearstone compares their emissions estimate to EPA/GRI (1996[BPO3]) emissions factors, as well as EPA (1995[BPO4]) emissions factors. The Clearstone report notes that EPA EFs are presented in CH_4 basis only, and so are not exactly comparable to the reported total hydrocarbon EFs (e.g., an unknown factor of 0.8–0.95 likely needed to convert between these two EFs). We assume comparison to the EPA/GRI (1996[BPO5]) EFs is more appropriate, as these form the major basis of the current EPA GHGI. The comparable estimates range from 0 to 5.53 kg CH_4/h/source. This leads to a central estimate for the ratio of measured/estimated to range from 0.2 to 30.1 g/g. Because Clearstone does not report gas composition and measures total hydrocarbon emissions, these ratios are likely high by an (uncertain) factor of ~ 1/0.9. In deference to our goal of using directly reported data in the main part of the first chart, we do not correct this discrepancy.

NGML

National Gas Machinery Laboratory (NGML), Clearstone Engineering Ltd., and Innovative Environmental Solutions Inc. (*21*) create nine cases that we include in the first text chart. See below for discussion of the methods used in NGML *et al*. This report estimates emissions for:

- Connectors: 64369 components screened
- Block valves: 7692 components screened
- Control valves: 495 components screened
- Pressure relief valves: 124 components screened
- Pressure regulators: 320 components screened
- Orifice meters: 57 components screened
- Crank case vents: 27 components screened
- Open ended lines: 1055 components screened
- Compressor seals: 299 components screened

NGML *et al*. report emissions of 0 to 0.52 kg total hydrocarbons/h/source, as gathered from table 4 (p. 31) in the original source. NGML *et al*. compare their emissions estimate to the EPA/GRI (1996) EFs for the technology of interest. The NGML report notes that EPA EFs are presented in CH_4 basis only, and so are not exactly comparable to reported total hydrocarbon EFs (e.g., an unknown factor of 0.8–0.95 likely needed to convert between these two EFs). The comparable EFs range from 0 to 1.17 kg CH_4/source/h. This leads to a range of central estimates for the ratio of measured/estimated to be of 0.2 to 10.8 g/g. Because NGML *et al*. do not report gas composition and instead measure total hydrocarbon emissions, these ratios are likely high by an (uncertain) factor of approx. 1/0.9. In deference to our goal of using directly reported data in the main part of the first text chart, we do not correct this discrepancy.

GTI

The Gas Technology Institute (GTI) study (*16*) create three estimates for emissions from distribution equipment that we include in the first text chart. See below for discussion of the methods used in GTI. The GTI study estimates emissions from the following sources:

- Commercial meters: 836 components screened
- Industrial meters: 46 components screened
- Residential meters: 2400 components screened
- District regulator stations: 77 components screened
- Pressure limiting stations: 11 components screened
- Custody transfer stations: 37 components screened
- Vehicle compressor stations: 10 components screened

Because comparisons were not broadly made in the GTI study to EPA EFs, figure 1 only includes residential meters, commercial meters, and industrial meters. They report emissions ranging between 2 and 8602 lbs CH_4/h, as gathered from table 3 in the original source. GTI compares their emissions estimate to EPA/GRI Tier 3 EFs for the devices of interest. The comparable estimates are 2.0 to 5.8 lbs CH_4/h. This leads to a range of central estimate for the ratio of measured/estimated of 0.4 to 4254 g/g.

Allen et al.

Allen *et al*. (*26*) create four estimates that we include in the first text chart. See below for discussion of the methods used in Allen *et al*. They estimate emissions, as useful for our purposes, for the following sources:

- hydraulic fracturing flowback emissions,
- chemical pumps,
- pneumatic controllers, and
- general equipment leaks.

These estimates were made by sampling 27 well completions, 9 gas well unloading events, 4 well workovers, and 489 wells in normal operation (see table 1 in original source). Allen *et al*. report emissions ranging from 0.6 to 2.2 Mg CH_4/unit per year for these sources. These results are gathered from table 2 in the main text of the original source, as central estimates plus uncertainty ranges in Gg CH_4/year. The results presented in Allen at al. main text are large in magnitude because they have been scaled to a national level using activity factors from the EPA inventory (generally region specific activity factors). Using the activity data from table S5-2 in the original source Supporting Information, we can compute ranges for low, central, and high emissions per unit. These per unit emissions can then be compared to the appropriate EPA inventory EF, as given in table S5-3 in the original source supporting information. Allen *et al*. compare their emissions estimate to the EPA inventory EFs (net of controls and regulatory reductions) for the technology of interest. The comparable estimates range from 0.3 to 8 Mg CH_4/device/year. This leads to central estimates for the ratio of measured/estimated emissions that range from 0.03 to 2 g/g.

1.4.2 The first text chart (inset): Adjustment of atmospheric estimates to scaled EPA GHGI

In order to make estimates in the main part of the first text chart more comparable, we normalize all ratios for studies at the regional atmospheric scale or larger. We normalize all these studies to the most recent EPA greenhouse gas inventory (GHGI). This inventory covers the years up to 2011, but was published in 2013, so we henceforth refer to it as the 2011/2013 GHGI (*17*). If a study already uses the EPA GHGI emissions factors for its comparison to inventory or EF, we do not make any adjustment to the ratio as reported in the study.

For each study, we normalize the GHGI estimate to the appropriate study spatial and sectoral boundary. In essence, we multiply the estimated national emissions from the GHGI by a scaling factor representing the fraction of activity in the region and sectors of interest (e.g., 11% of population, or 16% of NG end use consumption).
Some top-down studies use measurements sensitive to Canadian emissions [e.g., (*5*,*6*)]. These samples represent a small fraction of the observations considered in these respective analyses. These studies calculate a scaling factor, that when applied to the spatially-resolved EDGAR inventory, best matches observations. This scaling factor is applied uniformly to EDGAR (including Canada), but is most robust where observations are densest over the United States. The observationally derived scaling factor (e.g., Kort *et al*. ratio of 1.08 g/g relative to EDGAR v3.2), can then be applied to EDGAR U.S. emissions to estimate total U.S. emissions (e.g., 32.4 TgC/year in Kort *et al*.). These overall U.S. emissions can then be compared to EPA inventory. Some of the largest-scale atmospheric studies will show some influence of Canadian emissions, but any distortion due to this effect is likely to be small, owing to relative paucity of Canadian samples used in these studies and due to similarities in Canadian and U.S. drilling and production practice.

The following steps were taken for each study to generate a comparably scaled EPA GHGI emissions estimate:

1.4.2.1 Step 1: Determine study year

The study year is defined as the year in which measurements were taken or a key year of study measurements if multiple years of data are included.

1.4.2.2 Step 2: Collect emissions estimate for study year

EPA 2011/2013 GHGI estimates for anthropogenic emissions of CH_4 from all sources are collected for the study year.

1.4.2.3 Step 3: Determine appropriate sectoral and regional scaling

The appropriate scaling boundary for each study is then determined. In some cases, this scaling factor represents a regional boundary (e.g., South Coast air basin), in which case the scaling might be performed using the fraction of population present in the study's spatial boundary to the total U.S. population. In other cases, our scaling of the EPA GHGI is developed by matching to the same sector(s) that the study intended to measure CH_4 emissions from (e.g., estimate is of oil and gas emissions). If an author makes a quantitative attribution of CH_4 emissions to a particular sector or sub-sector (e.g., the author estimates leakage of pipeline quality NG) we utilize that judgment to create an appropriately scaled inventory for comparison.

1.4.2.4 Step 4: Generate scaling factors for included sources.

Scaling factors for each study were generated as described below.

1.4.2.4.1 Natural gas emissions by industry sector

Natural gas production sector emissions are scaled by the fraction of total United States non-associated gross gas production in the study area of interest. Oil-associated gas production is removed from both the numerator (production in the basin) and denominator (total U.S. production) because oil associated gas production emissions are included within the EPA GHGI petroleum inventory, not within the EPA GHGI NG inventory (*31*). EIA data sources are used in general to scale production emissions. For regional (e.g., sub-state scale) studies, regional production statistics are used from state reporting agencies (See Data File S1 for detailed listing of sources).

Natural gas processing sector emissions are scaled by the fraction of U.S. gas processed in the study region of interest. For regions where gas processing volumes are not available, volumes of gas processed are estimated. First, total gas production (associated and non-associated) volumes are computed as the feedstock for gas processing. We then assume that the fraction of gas processed is equal to the U.S. average processing fraction for 2011. This fraction is computed on either a net or gross output basis, depending on nature of reported gas production statistics. Using either reported or estimated processed gas, the share of U.S. gas processing that occurred in the study region is computed. These data are generally collected from EIA (see Data File S1 for detailed listing of sources).

Transmission sector emissions would ideally be estimated using the fraction of national gas transmission in a region. Because the volume of gas transmitted through a region is generally not available, an estimate of transport intensity is generated based on production and consumption shares in the region of interest. The weighting factor for gas transmitted is estimated as the simple average of the regional share of dry gas produced and the regional share of gas consumed in the region. EIA data are used where applicable (see Data File S1 for details).

Distribution sector emissions are scaled by the fraction of U.S. gas consumption in the region. Fractional consumption is computed on the basis of gas volumes delivered to

consumers. EIA data are used here, except in cases where consumption is not reported on a sub-state basis, in which case regional consumption data or population are used to scale the estimates of consumption.

1.4.2.4.2 Petroleum emissions

Petroleum emissions are scaled by the fraction of U.S. petroleum production for the study year that occurred in the study region. EIA data are generally used. Sub-state data from state regulators (for regional air studies) are used in some cases. See Data File S1 for details.

1.4.2.4.3 Coal mining emissions

Coal mining emissions are scaled by the fraction of coal production for the study year occurring within the study region. EIA data are used where applicable, as are sub-state data where needed (for regional atmospheric studies). See Data File S1 for details.

1.4.2.4.4 Livestock emissions

Manure management and enteric fermentation emissions are scaled by the fraction of U.S. cattle contained in the study region. Data are generally provided by USDA regional livestock statistics (see Data File S1 for details). Cattle provide a first approximation of enteric and manure emissions sources, because cattle represent ~95% of emissions from enteric fermentation [table A-195 in (*31*)], 55% of emissions from manure management [table A-205 in (*31*)], and 85% of total livestock emissions.

1.4.2.4.5 Landfill emissions

Landfill emissions are scaled using the fraction of U.S. population in the region. In California cases, it is known experimentally that California landfill emissions rates are lower than national averages due to stronger CH_4 capture regulations (*2*). For this reason, California average landfill emissions factors are applied to California studies (a scaling factor of ~0.5 relative to national emissions rates).

1.4.2.4.6 Other sources

All other sources are scaled by the fraction of U.S. population living in the region.

1.4.2.5 Step 5: Scale EPA GHGI emissions

Using the region- and sector-specific study scaling factors calculated as above, we scale emissions from the GHGI for the study year to the study region. Note that for each study, the result from the 2011/2013 EPA GHGI (*17*) is used for the year in which measurements were made, not for the most current year of the inventory. Because the 2011/2013 EPA GHGI contains estimates for the years 1990 to 2011, all studies except one contain data from the GHGI for the year in which measurements were made (*1*). For the case of Karion *et al*., which contains measurements from 2012, EPA GHGI estimates from 2011 were used. Central estimates from the EPA GHGI were used due to lack of ability to apply EPA uncertainty ranges to sub-national scales.

Details of scaling for each study are described below.

Miller et al. 2013

Miller *et al*. (*6*) create multiple estimates for CH_4 emissions. National scale emissions are estimated as 33.4 ± 1.5 Tg C as CH_4 per year. Regional emissions for the South-Central United States (TX, OK, KS) are estimated as 8.1 ± 0.96 Tg C as CH_4/year. Also, using the fitted sources for the South-Central U.S. region, they estimate possible emissions of C as CH_4 of 3.7 ± 2 Tg/year from the oil and gas industry.

The national-scale estimate is converted to Tg CH_4 and compared to the national-scale estimate for 2008 from the EPA 2011/2013 GHGI. The regional estimate for the South-Central United States is compared to a scaled EPA 2011/2013 GHG for the study region, using state-level data reported by EIA. Estimated excess emissions from the oil and gas industry are compared to regional inventoried emissions from the NG and petroleum sectors.

Kort et al. 2008

The estimate of total national emissions from Kort *et al*. (*5*) for 2003 is 32.4 ± 4.5 Tg C as CH_4/year from all sources. Kort *et al*. compare this estimate to EDGAR v3.2 estimates for U.S. emissions (30 ± 2.87 Tg C as CH_4).

We convert the Kort *et al*. estimate for 2003 emissions to Tg CH_4 and compare this estimate to the EPA 2011/2013 GHGI estimate for the year 2003. No scaling is applied, as Kort produces a national estimate.

Xiao et al. 2008

Xiao *et al*. (*8*) use C_2H_6 measurements and 3 cases of possible CH_4/C_2H_6 ratios from fossil fuel sources to arrive at three estimates for possible CH_4 emissions from energy industries (NG, petroleum, coal). Estimated CH_4 emissions from these industries are 13.3,

16.0, and 24.0 Tg CH_4. Xiao *et al*. compare these estimates to estimates for 2004 CH_4 emissions from these industries from both the 2007 and 2008 EPA GHGI.

We compare the estimated C_2H_6 emissions from Xiao *et al*. to estimates for C_2H_6 generated from the 2011/2013 GHGI CH_4 estimate from the energy industries. We scale CH_4 emissions from the energy industries (NG, petroleum, and coal) by the same CH_4/ C_2H_6 ratios used in Xiao to arrive at implied C_2H_6 emissions.

Wang et al. 2004

Xiao notes that Wang *et al*. (*13*) found emissions of 20 Tg CH_4/year from energy production in the United States for 1998. The source of this estimate was not found in the Wang *et al*. paper, and is cited as a personal communication in Xiao. It appears to be a regional result extracted from the global assessment of Wang (e.g., Wang's paper focused on global results, but regional results were also generated). Xiao compares this estimate from Wang to an estimate of 10 Tg CH_4/year from the energy industries.

We compare the Wang estimate of 20 Tg CH_4/year to the EPA 2011/2013 GHGI estimate for 1998 for the primary energy industries (NG, petroleum and coal).

Katzenstein et al. 2003

Katzenstein *et al*. (*9*) estimate emissions of 4–6 Tg CH_4 per year in the South Central United States (TX, OK, KS). Katzenstein *et al*. compare this emissions rate to a rate of 4–8 Tg CH_4 per year from oil and gas industries, using a 1997 EPA source as the basis for comparison.

We compare the Katzenstein *et al*. emissions estimate for the region to a scaled version of petroleum and NG emissions from the region. The EPA 2011/2013 GHGI estimate for 2001 emissions from NG and petroleum sources is scaled to the region using the fractional production of oil and gas in the region in 2001.

We note that attribution in Katzenstein is made to the hydrocarbon industries (NG and petroleum) on the basis of evidence from higher alkane prevalence. They do not appear to do any rigorous attribution of emissions to the possible sources in the region.

Pétron et al. 2012

Pétron *et al*. (*3*) estimate emissions from the Denver-Julesberg (DJ) basin of 129.6 Gg CH_4 (71.6 - 251.9). They compare this estimate to WRAP inventory estimates of 64.3 Gg CH_4 (46 - 86). Because the estimate of Pétron *et al*. is created by scaling higher hydrocarbons (C_3H_8) that have no significant biological sources, it is fundamentally an estimate of NG and petroleum emissions.

We create a basin-level scaled estimate of GHG emissions from oil and gas operations by scaling the EPA 2011/2013 GHGI emissions from oil and gas operations to local production. Because isolating wind direction is used to exclude emissions from the nearby Denver metropolitan area, we exclude distribution and transmission emissions from this large population center. We do include consumption within Weld County itself, as scaled by fraction of U.S. population within the study region.

Production in the DJ basin in 2008 equaled 17.6 x 10^6 bbl of petroleum and 204.4 BCF of NG. In figure 4 of a reply to Levi (*32*), Pétron *et al*. note that 40% of the NG production in the DJ basin comes from wells classified by GOR as "dry" gas or "leaning dry". We assume that this production would be classified as gas wells in the EPA methodology, while other gas would be classified as production of oil-associated gas.

Levi 2012

Levi (*25*) re-analyzes the Pétron data, using C_4H_{10} inventory fluxes and the C_4H_{10} to CH_4 ratios observed in the atmosphere to scale CH_4 concentrations to flux. He estimates a central flux of 51.8 Gg CH_4/year (42.5 –78.9). Levi then compares these to the WRAP inventory for the region.

We compare the Levi estimates to the scaled EPA GHGI for the year 2008 for the DJ basin, constructed identically to that for Pétron *et al*. above.

Wunch et al. 2009

Wunch *et al*. (*12*) estimate emissions of 0.6 ± 1 Tg CH_4/year for the South Coast Air Basin (SoCAB) for the 2007–2008 time frame (August 2007 to June 2008). They compare this estimate to a scaled version of the CARB inventory for urban CH_4 fluxes.

We construct a regionally-scaled EPA 2011/2013 GHGI for the years 2007–2008, following the methodology noted above with county-level SoCAB data.

Hsu et al. 2010

Hsu *et al*. (*10*) estimate CH_4 emissions in the portion of Los Angeles county contained within the SoCAB. They estimate emissions of 0.2 Tg CH_4 (±0.01 Tg CH_4). They compare these estimates to a scaled version of the CARB state GHG inventory for the region.

We construct a scaled estimate of the EPA 2011/2013 GHGI for the region in a similar fashion to that of the Wunch *et al*. study above. One note of interest: some emissions sources are scaled by the fraction of U.S. population in Los Angles county within the

study region. To approximate this study region, we take the population of Los Angeles county and subtract the populations of Lancaster and Palmdale, two key cities in the non-SoCAB portion of Los Angeles county.

Wennberg et al. 2012

Wennberg *et al*. (*7*) estimate emissions of all sources of 0.44 (0.4–0.48) Tg CH_4/year. They estimate the "maximum contribution" from the gas system of 0.38 (0.23–0.53) Tg CH_4/year.

We compare the results for all sources to a regionally-scaled version of the 2011/2013 EPA GHGI for the year 2008.

Peischl et al. 2013

Peischl *et al*. (*2*) estimate total emissions in the SoCAB of 0.411 (0.374–0.448) Tg CH_4/year. They further estimate sources associated with oil and gas activities of 0.224 (0.163 – 0.285) Tg CH_4/year.

We compare these results to regionally-scaled versions of the 2011/2013 EPA GHGI for the year 2010. We construct two scaling cases. First we construct an "all sources" case that includes all plausible sources from the GHGI in the SoCAB. We also construct a NG and petroleum sources case, which scales NG and petroleum sources from the 2011/2013 EPA GHGI to the region (production, processing, transmission, and distribution).

Karion et al. 2013

Karion *et al*. (*1*) estimate oil and gas operations emissions of 54.6 (39.1–70.1) t CH_4/h, or 0.478 (0.343–0.614) Tg CH_4/year in Uintah county. Estimates in Karion *et al*. were made based on measurements taken in 2012.

We compare this to a scaled version of results from 2011 for the EPA 2011/2013 GHGI that includes NG, petroleum, and other sources. Cattle-associated sources (enteric fermentation and manure management) are removed from our comparable GHGI estimate because Karion *et al*. remove cattle emissions from their estimate. For completeness, we do include population scaled emissions sources but these are trivial in the region of interest (no significant population centers in the Uintah basin).

1.4.2.6 Step 6: Compute normalized ratios

Using the scaled EPA GHGI estimate, we compute the ratio between the author estimates (central, low, high) and the scaled EPA GHGI.

These calculations are recorded in the Data File S1 in the "Calc – Figure 1 - Inset" worksheet.

1.4.3 The second text chart: Exploration of possible sources of excess CH_4

The second text chart illustrates possible sources of excess CH_4 for the NG sector relative to the national EPA GHGI of 2011. The second chart also includes sources that could confound estimated NG emissions, such as petroleum system emissions or natural seeps. Note: The second chart is intended as an order of magnitude "thought experiment" to explore potential magnitude of possible source contributions to excess CH_4 emissions, given currently available evidence. The resulting uncertainty ranges on source contributions are large, as might be expected from the current state of knowledge of leakage sources.

These calculations are recorded in Data File S1 in the "Calc - Figure 2" worksheet.

1.4.3.1 Baseline excess CH_4

The first text chart (inset) suggests that, based on current evidence, the ratio of measured and inventoried CH_4 emissions is approximately ~1.5 at multi-state and larger scales (*5, 6, 8*). We utilize this excess ratio to scale emissions from the EPA GHGI by a factor of 1.5 (±0.25) so as to develop an estimate of the range of possible national excess CH_4. This results in baseline excess CH_4 emissions rate of 14 (±7) Tg CH_4. This range is plotted as the blue bar in the second chart in the main text.

Importantly, this excess includes any sources of CH_4 that are emitted in excess of EPA GHGI estimates. For example, there are indications that CH_4 emissions from livestock are poorly understood (*33*). A recent atmospheric study suggested that livestock emissions could be underestimated by 5.7 Tg C per year (*6*). *It should therefore not be expected that NG sources would make up all of estimated excess CH_4 in the second text chart.*

1.4.3.2 "Upstream" emissions: Natural gas production and processing leakage in excess of EPA GHGI estimates

Leakage from upstream operations (production and processing) could be in excess of rates estimated in EPA inventories. Several recent studies estimate high emissions from production and processing operations (*1*, *3, 45*).

The Pétron *et al*. study *3*) found a central estimate of 4% leakage from the Denver-Julesberg basin. The Karion *et al*. study (*1*, *45*) found a central estimate of 8.9% leakage

from the Uintah basin. We consider the Karion *et al*. study results as the emissions rate for "high emitting" NG production and processing operations. We assume for purposes of comparison to the EPA GHGI that both of these studies measure all emissions from production, gathering, tanks, and gas processing, in addition to 25% of emissions from transmission (due to initial compression work to bring gas to the transmission pressure).

No data are available on the fraction of gas production that occurs at "high emitting" gas production and processing operations. In 2012, the Uintah basin produced approximately 1.4% (320 BCF) of gross U.S. gas production (see Data File S1 for sources). The second text chart plots three illustrative prevalence cases:

- Low: 1% of gas production and processing occurs from "high emitting" gas fields
- Medium: 10% of gas production and processing occurs from "high emitting" gas fields
- High: 25% of gas production and processing occurs from "high emitting" gas fields

These three cases result in excess emissions above EPA GHGI emissions of 0.54 to 9.43 Tg CH_4/year. EPA GHGI emissions in this sector were equal to ~1.5 Tg CH_4 in the study year (see Data File S1). These Low, Medium and High cases for excess upstream emissions are plotted as points *a*, *b*, *c* in the "NG production and processing" bar in the second chart in the main text.

Given the high leakage rates estimated in these studies (*1, 3*), it is unlikely that a significant fraction of North American gas fields have leakage rates at this "high emitter" rate (as total excess CH_4 in the second text chart would be more than satisfied).

1.4.3.3 "Downstream" sources: Natural gas distribution and end-use leakage in excess of EPA estimates

Leakage from downstream sources (distribution and end use of gas) also could be in excess of amounts estimated in EPA inventories. The Wennberg *et al*. study (*7*) found a central estimate of 3.5% leakage from the distribution system and "after the meter" end uses of gas. The Peischl *et al*. study (*2*) found a central estimate of 2% leakage of gas consumed in the region. The Peischl study notes some confounding with geologic seeps (e.g., La Brea tar pits in Los Angeles). Because we consider geologic seeps elsewhere in the second text chart, we do not include these sources here. We consider the Wennberg *et al*. emissions rates as our "high emitting" distribution and consumption system rate.

No data are available on the fraction of gas consumption that occurs at the "high emitting" gas distribution and consumption systems. The SoCAB was responsible for ~4.3% (992 BCF) of total U.S. end-use gas consumption in 2008 (see Data File S1). The second text chart plots three exploratory prevalence cases:

- Low: 1% of gas consumption occurs from "high emitting" distribution and consumption systems
- Medium: 10% of gas consumption occurs from "high emitting" distribution and consumption systems
- High: 25% of gas consumption occurs from "high emitting" distribution and consumption systems

These three cases result in excess emissions of 0.1 to 3.1 Tg CH_4/year above EPA GHGI emissions. These Low, Medium and High cases for excess downstream emissions are plotted as points *a*, *b*, *c* in the "NG distribution and use" bar in the second chart in the main text.

Given the high leakage rates estimated in these studies, we consider it unlikely that a majority of consuming regions have effective leakage rates at the "high emitter" rate. For this reason, we do not explore cases with higher prevalence of these leakage rates.

1.4.3.4 Excess petroleum production emissions

Petroleum production emissions of CH_4 are classified as petroleum sector emissions in the EPA GHGI, even though a significant fraction of U.S. NG is produced from "oil" wells.

Peischl *et al*. (*2*) examine emissions from production operations in the SoCAB. SoCAB is primarily an oil-producing region: 98% of the NG produced in the region is oil-associated gas (*42*). Therefore, emissions from production of gas in the region provide insight on CH_4 emissions from petroleum production.

Peischl *et al*. estimate a leakage rate of 17% of the gas produced in the basin (range of 14% to 20%) (*2*). Peischl *et al*. also note that a California Air Resources Board (CARB) survey estimated leakage rates in the SoCAB basin of 12% of the gas produced (*2*). To create illustrative scenarios, we assume leakage of 17% of the associated gas produced for "high emitting" oil fields with associated gas production.

No data are available on the prevalence of "high emitting" oil fields. It is known that SoCAB was responsible for 0.007 TCF of gas production, which is ~0.1% of total U.S. associated gas production of 5.91 TCF in 2011. The second text chart plots three exploratory prevalence cases:

- Low: 1% of associated gas production occurs from "high emitting" oil fields
- Medium: 10% of associated gas production occurs from "high emitting" oil fields
- High: 25% of associated gas production occurs from "high emitting" oil fields

These cases are meant to "benchmark" emissions rates at tangible levels, and no data exist to assign a probability to one or more of these cases. These three cases result in excess emissions above EPA GHGI estimated petroleum production emissions of 0.1 to

3.7 Tg CH_4/year. Given the very high leakage rates estimated in the Peischl study, we consider it unlikely that a significant fraction of oil fields emit at this "high emitter" rate.

These Low, Medium and High cases for excess petroleum production emissions are plotted as points *a*, *b*, *c* in the "Petroleum production" bar in the second chart in the main text.

1.4.3.5 Hydraulic fracturing for NG

Hydraulic fracturing of NG has been examined as a potentially large source of CH_4 emissions (*41, 43, 44*). Recent EPA GHGI modifications have also focused on shale gas emissions (*17, 31*). Emissions during the well completion flowback period are a chief concern.

EPA estimates that net emissions in 2011 from hydraulic fracturing of NG wells (shale gas, tight gas, or any other hydraulically fractured well) were 0.796 Tg of CH_4. Because they estimate that 8077 wells were hydraulically fractured in 2011, this amounts to a per-well net emissions rate of 98.7 Mg CH_4 per well. Note: this value is different from the per-region emissions factors per well (as reported in EPA 2011/2013 GHGI Annex 3) because these net emissions subtract any reductions associated with green completions technologies or other voluntary reductions programs reported to EPA.

O'Sullivan and Paltsev compute two cases for flowback emissions in the 5 major shale gas plays in 2010 (*41*). He estimates a drilling-weighted "current practice" emissions rate of 54.6 Mg CH_4/well (including CO_2 as CH_4 eq.). He also estimates an "all venting" case, where no green completions technologies or flaring is applied, of 228.5 Mg CH_4/well. Because O'Sullivan only focuses on the 5 major shale plays, he estimates a smaller number of wells completed than EPA (3948 wells in 2010).

Howarth *et al*. (*43, 44*) estimate emissions during flowback, but do not compute total emissions for completed wells in a year, nor do they state the prevalence of wells completed using a particular method (e.g., all flowback vented, as they estimate). This makes the Howarth *et al*. results non-comparable to EPA GHGI EFs.

We estimate two cases for hydraulic fracturing of shale gas wells: shale gas wells in the 5 major shale gas plays, and all hydraulically fractured NG wells. In 2012 New Source Performance Standards (NSPS) for this source category began to be phased in. Therefore, it is unlikely, going forward from 2012 that these sources will be major contributors.

1.4.3.5.1 All hydraulically fractured NG wells

To estimate emissions from all hydraulically fractured gas wells (i.e., ones from the 5 largest plays included above plus all other shale as well as tight gas fractured wells), we perform sensitivity analysis on the EPA net emissions rate per hydraulically fractured well. EPA average net CH_4 emissions per hydraulically fractured well were 98.7 Mg

CH_4/well. Our sensitivity cases multiply this emissions rate by an illustrative factor of 0.5 and 2 for low and high cases, respectively. Note that these emissions rates of 49.3 and 197.4 Mg CH_4/well are similar in scale to the "current practice" and "all venting" cases for shale gas plays noted in O'Sullivan (*41*), and are deemed to be reasonable bounds on uncertainty with respect to flowback venting emissions.

These low and high bounds are applied to all wells hydraulically fractured in the EPA 2011 GHGI (8077 wells) resulting in excess emissions with respect to the EPA GHGI of –0.4 Tg CH_4 and 0.8 Tg CH_4 in the low and high cases, respectively. These results are plotted as results *d* and *e* in the second chart in the main text.

1.4.3.5.2 Shale gas wells in 5 major shale gas plays

The HDPI database suggests that a total of 4012 wells in the 5 major shale gas plays were completed in 2011 (see above for method of data gathering from HDPI database). To estimate emissions from these wells, we assume that O'Sullivan's (*41*) estimates of flowback emissions intensity per well from 2010 are applicable to 2011.

The low case applies O'Sullivan's estimate for the "current practice" case, while the high case applies O'Sullivan's estimate for the "all vented" case. These per-well intensities (weighted by completions) equal 56.0 and 234.0 Mg CH_4/well.

Applying these completions emissions intensities to the number of wells assumed drilled in the 5 major shale gas plays in 2011 (4012 wells), while assigning EPA default intensity to other hydraulically fractured wells, we arrive at excess emissions of –0.17 and 0.51 Tg CH_4/year in the low and high cases, respectively. These results are plotted as points *f* and *g* in the second chart in the main text. Note that a negative "excess" relative to the EPA GHGI can exist as the lower bound if the alternative emissions estimate is lower than the EPA estimate.

1.4.3.6 Sources that are not included in EPA GHGI

The second chart in the main text includes three sources that are not included in EPA CH_4 inventories, but which could be mistaken for NG emissions by chemical or isotopic composition. These sources include and hydraulic fracturing of oil wells (e.g., shale oil wells), abandoned oil and gas wells, and geologic seeps.

1.4.3.6.1 Hydraulic fracturing of oil wells

EPA does not include hydraulic fracturing of oil wells in the GHGI. Activity data for hydraulic fracturing of oil wells are taken from the two largest shale oil plays, the Bakken and the Eagle Ford plays. Also included are completions in the Permian basin of TX. The Barnett play is not included in this portion of the analysis (reflecting tight-oil emissions) because it is largely a gas-bearing play.

Well data were captured from the Drilling Info HPDI database on August 22nd, 2013 (*40*). Filters were applied to identify wells for analysis. In both the Eagle Ford and Bakken plays, all new production wells are horizontal. In the Permian, new production wells are both horizontal and vertical owing to the fact that in many cases Permian wells target "tight sand formations," where a vertical trajectory is optimal. To generate an upper bound estimate of emissions, it is assumed that all new oil wells in the three analyzed plays are completed using hydraulic fracture stimulation.

Filters applied by play include:

All plays:

- Well production status: Active
- Completion year: 2010 or 2011
- First production year: 2011

Eagle Ford:

- Basin: TX & LA Gulf Coast Basin
- Reservoir: Eagle Ford, Eagle Ford Shale, Eagle Ford-1, Eagle Ford-2, Eagleford, Eagleford Shale
- Well production type: Oil
- Drill type: Horizontal

Bakken Well Details

- Basin: Williston
- Reservoir: Bakken
- Well production type: Oil (MT wells), and Oil & Gas (ND wells)
- Drill type: Horizontal

Permian Well Details

- Basin: Permian
- Reservoir: Trend Area (Spraberry), Wolfcamp and assoc., Bonespring and assoc., Yeso and assoc., Glorieta and assoc.
- Well production type: Oil
- Drill type: All

A total of 2969 wells were completed in these plays in 2011. Potential flowback emissions from these tight oil wells are estimated using the method of O'Sullivan (*41*). Peak gas production (typically first month of production) is converted to a daily initial production (IP) rate. Production during flowback is assumed to increase linearly with time for 9 days prior to IP. "Potential emissions" estimates assume all flowback emissions are vented. Because of productivity differences between wells and varying gas-oil-ratios, each play has a different per-well potential flowback emissions rate. Per-well emissions rates were found to be 31.1, 90.9 and 31.2 Mg CH_4/well, respectively, in the Bakken, Eagle Ford, and Permian basins.

The resulting emissions estimate for this source is computed as the number of wells completed multiplied by the per-well potential emissions. Total potential emissions are ~0.12 Tg CH_4/year for 2011. This emissions rate is plotted as point *h* in the second chart in the main text. Any reduced emission completion technologies applied would result in lower emissions than this. No activity data are available for use of RECs for oil well fracturing, so none are assumed in this assessment.

1.4.3.6.2 Abandoned oil and gas wells

Abandoned oil and gas wells represent conduits to hydrocarbon bearing formations. If these conduits were not properly sealed upon abandonment, then leakage from formations could be occurring. From 1859 to 2009, 3.74 $x10^6$ oil and gas wells were drilled in the United States (*36–38*). As of 2009, some 825,000 oil and gas wells were operating in the United States (*39*). This implies that ~3 $x10^6$ non-operating (abandoned and plugged) oil and gas wells exist in the United States.

No empirical data was found to characterize leakage rates from abandoned oil and gas wells. For this reason, we perform an order of magnitude exploration. We vary average per-well gas leakage rates of 0.1 MCF/year to 100 MCF/year and gas compositions equal to EPA default production composition (*31*). This results in U.S. emissions rates between ~0 and 4.7 Tg CH_4/year. While emissions at the high end of this range seem implausible, this uncertainty cannot be reduced further without empirical studies. The total emissions rates associated with 1, 10, and 100 MCF/well/year are plotted as points *i*, *j*, *k* in the second chart in the main text.

1.4.3.6.3 Natural geologic seeps

Geologic seeps occur in all hydrocarbon-bearing sedimentary basins (*34, 35*). To estimate a potential magnitude of CH_4 emissions from geologic seeps, we make the following assumptions:

- We include emissions from mud volcanoes, other macro-seeps, micro-seeps, and geothermal vents. Because we are interested in terrestrial sources, we do not include CH_4 emissions from oceanic seeps. Best estimates of global emissions from these sources ranges from 21.5–44.3 Tg CH_4/year (*35*).
- We scale global emissions by the U.S. fraction of Earth's surface area. The United States represents 6.6% of Earth's terrestrial surface area (9.8 x 10^6 km^2). We scale terrestrial seeps by this ratio, arriving at 1.4 to 2.9 Tg CH_4/year.

This range of 1.4 to 2.9 Tg CH_4/year is plotted as points *l* and *m* in the second chart.

For comparison, Miller *et al*. (*6*) estimate that geologic seeps could represent an overall uncertainty of 5% on their estimated CH_4 emissions, or 2.1 Tg CH_4/year. Regional variability in seeps is likely large, and is poorly understood. It is possible that the United States has a larger fraction of Earth's geologic seeps, because of its extensive sedimentary basins.

1.4.4 Calculating leakage percentages associated with possible NG leakage

Given the estimated U.S. national excess emissions from the second chart (main text) of 7–21 Tg CH_4/year, we can put bounds on the possible leakage rates from the NG system. First, current EPA GHGI estimates for leakage in 2011 are 1.8% of end use consumption or 1.4% of gross gas withdrawals (see SI spreadsheet). This amounts to 6.9 Tg CH_4 emitted in 2011. If all excess CH_4 is due to the NG industry, and the excess NG leakage has a similar molar CH_4 fraction as the current leakage profile, then excess emissions of 7–21 Tg CH_4/year correspond to total emissions of 2 to 4 times GHGI estimates for the NG industry (e.g., 14–28 Tg CH_4/year).

Removing sources that are known not to be in the GHGI, but measured in atmospheric observations (wild ruminants, and termites) the unexplained excess decreases to 6.8 to 20.8 Tg CH_4/year, or yields an excess percentage leakage of 1.8% to 5.4% of end use gas. Coupled with the current estimate of 1.8% leakage of end use gas consumed, this generates a high-end estimate of 7.1% gas leakage (on an end use basis). This worst-case scenario is unlikely: it would require *all* excess CH_4 to come from the NG industry, and require total excess at the high end of the observed range from national-scale studies.

Modeling has shown climate benefits from coal to NG switching for power generation over all time periods (i.e., starting immediately) if the well-to-power-plant leakage rate is below 3.2%, while benefits are seen over a 100 year period if leakage is below 7.6% (*28*) Therefore, available evidence suggests climate benefits from NG substitution for coal in the power sector over a 100-year assessment period. Alvarez *et al*. (*28*) found benefits from NG use in transport at leakage rates below 1.7% to 3.8% for 100 year assessment periods (gasoline and diesel substitution, respectively). Therefore, some scenarios appear to support use of NG in passenger vehicle gasoline displacement, but benefits from diesel substitution in heavy-duty trucking are less likely.

Uncertainties about the role of CH_4 in climate forcing (*46*) imply a need for reassessment of these decision points as knowledge improves. This includes updating of the framework developed in Alvarez *et al*. (*28*) based on recently increased global warming potential of CH_4 reported in the early publication version of the Intergovernmental Panel on Climate Change 5th Assessment Report.

The calculations reported in this section are recorded in the Data File S1, “Calc - Fuel switching” worksheet.

2 Supplementary text

This supplementary text describes in more detail some points of discussion from the main text. The order of discussion follows that of the main text: top-down studies; bottom-up studies and inventories; and policy and decision-making implications.

2.1 Top-down studies of CH_4 emissions

Top-down studies of CH_4 fluxes use atmospheric observations of CH_4 concentrations combined with modeling to infer emissions of CH_4. The strength of the top-down method is the direct observation of the end product of concern (atmospheric CH_4), whereas the challenges associated with the top-down method lie in accurately representing atmospheric transport and attributing CH_4 to specific sources.

The central challenge in using atmospheric observations to infer emissions is to quantify the link between the observable (dry air mole fraction of CH_4) and the quantity of interest (spatial distribution of surface mass emission of CH_4 per unit time). Atmospheric transport is the end result of complex, nonlinear, multi-scale dynamical processes, presenting a challenge for accurate simulation and observation at sufficient spatial scales and density. Studies approach this problem in different ways.

2.1.1 Transport box models (mass balance models)

Some studies (*1, 9, 47, 48*), have used variations of a simple box model or mass balance model to estimate CH_4 fluxes. These models do not rely on simulated wind fields, and instead make measurements of CH_4 upwind and downwind along concentration gradients and utilize assumptions about transport. A few examples are explained below to provide further insight into this approach.

Katzenstein *et al*. (*9*) collected data from two ground sampling campaigns from September 2001 to May 2002. Samples were analyzed for CH_4 and higher hydrocarbons. A simple box model based upon assuming a ventilation time and boundary layer height was used to estimate CH_4 emissions of 4–6 Tg/year. They note that the U.S. NG industry was estimated to have emissions of 6 ± 2 Tg of CH_4 per year at this time. Weighting the sample area emissions by either the fraction of the nation's NG produced in Kansas, Oklahoma and Texas (37% of NG production), or by those states plus the Gulf of Mexico offshore production (62%), one can calculate that the measured CH_4 emissions are 1.6–4.7 times the EPA GHGI estimate (KA, OK, TX) or 0.9–2.8 times the EPA GHGI estimate (KA, OK, TX, GOM).

Mays *et al*. (*48*) performed measurements around the city of Indianapolis by flying the perimeter of the city and comparing upwind and downwind CH_4 concentrations. They found elevated CH_4 concentrations relative to CO_2 concentrations, at orders of magnitude

larger than the ratios would be if incomplete combustion were the cause. This implies that CH_4 is derived "mostly from noncombustion sources" such as landfills, NG leakage and wastewater treatment. No more specific source attribution was possible with this model.

Karion *et al*. (*1*) recently used a mass-balance approach to estimating emissions from a NG field in North East Utah (Uintah basin). Their approach involved flying upwind and downwind transects near the gas field, with wind direction and velocity measurements used to integrate emissions across the horizontal plume downwind of the source. The resulting emissions determined from this flight were 6.2–11.7% of the gas production in the basin on the day of measurements.

The simple box model approach is appealing in that assumptions are straightforward, but large uncertainty ranges are a consequence of these straightforward assumptions, so uncertainty ranges of 50% or greater are the norm.

2.1.2 Tracer-tracer approaches

Tracer-tracer approaches also do not rely on simulated wind fields. Instead, a gas with more-precisely quantified emissions (gas A) than the gas of interest (gas B) is exploited to estimate emissions of gas B. If sources for A and B are co-located, or sufficient mixing has occurred between the point of emission and the point of measurement, the observed slope of the concentration of the two gases can be convolved with inventory estimates of emissions of gas A to produce an estimate for gas B. This approach assumes that: 1) emissions of gas A are well known, 2) the gases of interest are sufficiently well mixed, and 3) that atmospheric processes affect both gases equally.

Multiple studies have been performed to date in the Los Angeles basin using these approaches (*7, 9, 10, 12*). These tracer-tracer approaches have similarities with tracer methods applied at smaller scales with purposefully released tracers like SF_6 (*49*). Numerous studies of this type were performed in the 1990s, including a number of studies to support the EPA/GRI study (*50*).

Wennberg *et al*. use the California Air Resources Board (CARB) carbon monoxide (CO) inventory to scale CH_4 concentrations measured via aircraft in 2008 and 2010. Samples from the Mt. Wilson observatory and total air column concentrations from Wunch *et al*. (*12*) were also used. Wennberg *et al*. estimate CH_4 fluxes of 0.38 ± 0.1 Tg/year to 0.47 ± 0.1 Tg/year [table 2 in (*7*)]. These estimates can be compared to South Coast Air Quality Management District sector-based inventories of 0.212 Tg/year [table 3 in (*7*). Uncertainty is introduced into these methods by the uncertainty in the tracer gas inventory (CO emissions rates ±10%, included in above uncertainty ranges) [p. 9286 in (*7*)].

Wennberg *et al*. also examined C_2H_6 concentrations along with ratios of C_2H_6 to CH_4 in gas sold over the study time period. The bottom-up CH_4 inventory accounts for 35–73% of excess CH_4 emissions, but only 15% of the excess C_2H_6 . A NG source of 0.23 Tg with

a C_2H_6 :CH_4 ratio of 2.6% (similar to pipeline gas composition of 2.09%) simultaneously closes both of these budgets. Wennberg *et al*. note that the similar time trend in pipeline C_2H_6:CH_4 ratio and observed atmospheric C_2H_6:CH_4 ratio are further suggestive of pipeline gas being responsible for the majority of the observed atmospheric signal.

An earlier study by Wunch *et al*. (*12*) used Fourier transform spectroscopic (FTS) analysis to determine concentrations of CO_2, CO, CH_4 and N_2O. Correlations between concentrations of these species are used, along with CO_2 emissions from EDGAR and CARB inventories for the region, to estimate the fluxes of the other gases. They estimate 0.6 ± 1 Tg CH_4 per year. A similar approach using the CARB CO inventory to scale the flux arrives at a CH_4 flux of 0.4 ± 0.1 Tg/year, in agreement with Wennberg *et al*. above. Wunch *et al*. then scale the CARB CH_4 inventory to the South Coast Air Basin (SoCAB) by population (after removing emissions from agriculture and forestry, which are unimportant in the Los Angeles Basin). The scaled CARB CH_4 inventory result is 0.26 Tg/year, significantly below either inferred FTS result that uses CO_2 or CO as the tracer.

Finally, a study by Hsu *et al*. (*10*) used data from the Mt. Wilson observatory and the CARB CO inventory to estimate CH_4 fluxes in the region. They arrive at a result of 4.2 ± 0.12 MMT CO_2 eq./year, which can be compared to a spatially disaggregated CARB inventory for the region of 3.0 MMT CO_2 eq/year. Uncertainty arises in this study due to the limited spatial representation of the sampling and because the CARB bottom-up CH_4 inventory (used for comparison) is extrapolated from a 2004 inventory. As such, the study finds good agreement between the top-down estimate (derived from the observed CH_4/CO slope & CO inventory) and the bottom-up CH_4 inventory, even though under and/or un-inventoried sources may be present.

Numerous Los Angeles studies use CO as the tracer gas. As a consequence of addressing air quality problems, CARB has refined the CO inventory annually and the uncertainty in this inventory is considered to be small (best guess ~10%), compared with the CH_4 inventory. The usage of a tracer such as CO helps circumvent the need to directly represent atmospheric transport. It does however convolve the resulting top-down answer with a bottom-up inventory. For example, in the Wunch *et al*. study, CH_4 emissions differ by as much as 50% if a different CO_2 inventory is used (*12*).

In another study region, Pétron *et al*. (*3*) use alkane signatures to estimate emissions from the NG industry in the Denver-Julesberg basin of Northeastern Colorado. Emissions of 129.6 Gg CH_4/year (71.6 - 251.9 Gg) were estimated for the region, in comparison to 64.3 (46 - 86) Gg CH_4 estimated in a regional inventory. This study employs a tracer-tracer method, leveraging inventory estimates of higher HC (C_3H_8) emissions along with gas emissions composition profiles to estimate a CH_4 source from observed CH_4 and C_3H_8. Observations were generated using a tall tower and ground-based sampling. There has been controversy associated with this study (*25, 32*), which is discussed below. Note that the Pétron attribution to oil and gas emissions is further supported by studies which attribute VOCs in the region of the Boulder Atmospheric Observatory tower largely to oil and gas activities (e.g., 72% of C_2H_6 and 90% of C_3H_8 in the region) (*51*).

Tracer release studies developed in the 1990s to estimate emissions across a facility use a similar approach with similar assumptions (*49, 52, 53*). In this case, the tracer is purposely released, and is a gas species that is (a) easily detected, and (b) present at essentially no background concentration. In these studies SF_6 was chosen as the tracer gas. The tracer gas is released in the same vicinity as the suspected gas leaks, and the concentrations of the two gases are measured downstream in the plume after sufficient atmospheric mixing has occurred between the two sources.

2.1.3 Transport inversion modeling

Using full simulations of atmospheric transport (wind fields), observed concentrations can be linked to fluxes. The sensitivity of an observation to a given location's flux can be calculated, and then an inverse modeling approach can be employed—either to scale a prior emissions field (Bayesian inversion) or to derive fluxes from observations and predictor variables (Geostatistical inversion).

A number of studies have been performed on CH_4 in North America using atmospheric inversion (*5, 6, 8, 9*). Kort *et al*. (*5*) used data from a campaign of 38 flights undertaken in 2003. Approximately 300 flask samples were acquired and concentrations were measured for a number of species, including CH_4. The resulting concentration data (e.g., 300 data points) were combined with a time inverted transport model to simulate the transport of air parcels. This modeling technique explicitly links the observed concentrations with surface fluxes. Spatial emissions inventories (EDGAR) are used to initialize sources in the model, and a Bayesian inversion is performed. The model and measurements provided strong constraints on continental-scale emissions but did not have the resolution to distinguish different sources of CH_4 or to examine regional scale inventories. Good agreement was found with the EDGAR v3.2 2000 inventory (*5, 6*), but emission rates were higher than in the EPA GHGI and the updated EDGAR v4.1 inventory (*6*).

More recently, Miller *et al*. (*6*) used a two year record of observations (~12000 observations from aircraft and tall towers) spanning most of the United States with a geostatistical approach to estimate CH_4 fluxes with less reliance on prior inventory spatial distributions. A version of the inverse model that used the EDGAR spatial emissions dataset found the emissions to be too low and the spatial representation to be incorrect. With the high-resolution geostatistical approach Miller *et al*. find significant underestimation of emissions of CH_4 from the south-central United States. Also, Miller *et al*. use alkane (C_3H_8) data from aircraft measurement to help partition excess CH_4 in the region between fossil fuel extraction, livestock and other sources.

These two studies (*5, 6*) used different datasets [aircraft transecting North America for (*5*), tall-towers and vertical profiling aircraft for (*6*)], for different years (2003 and 2008), with different spatial coverage, and used different inversion techniques [Bayesian for (*5*), Geostatistical for (*6*)]. Despite these differences, both top-down methods found similar emission rates (35.11 & 32.4 Tg C/year), substantially larger than EDGAR v4.2 or U.S. EPA GHGI emissions estimates.

Xiao *et al*. (*8*) used a global chemical transport model to model the global emissions inventory for C_2H_6. C_2H_6 concentrations from dozens of sampling campaigns from 1989 to 2004 were input to the model. Using results from the U.S. region (U.S. C_2H_6 emissions estimated at 2.4 Tg/year), as well as U.S. GHGI results, Xiao *et al*. examine whether CH_4 emissions inventories are likely to be accurate. Given an approximate inventory emissions rate of 10 Tg CH_4/year from the EPA GHGI, they find that the related C_2H_6 emissions should be 1.2 to 2.5 times lower than observed [table 4 in (*8*)]. Thus, C_2H_6 measurements, which cannot be attributed to biogenic sources, imply larger CH_4 emissions than calculated in the EPA GHGI.

The largest source of uncertainty in transport modeling is typically representation of transport. In particular, bias errors in features such as boundary layer height can directly map into flux fields. This can result in a high fidelity (narrow error-bar) answer with potentially larger bias errors.

2.1.4 Approaches for source attribution

Attributing the CH_4 observed in measurements to NG production is challenging. A variety of approaches have been proposed to overcome these attribution challenges: isotopic measurements; alkane fingerprints; and proximity and flux direction.

Townsend-Small *et al*. (*11*) sampled air near multiple CH_4 sources (e.g., power plants, oil fields, landfills). $\delta^{13}C$ and δD (deuterium) values for CH_4 were collected for these exhaust streams [table 1 and figure 2 in (*11*)]. Extrapolating from measured data to pure CH_4 isotopic ratios suggests ratios of $\delta^{13}C \approx -41.5$ and $\delta D \approx -203$ to -228. The authors suggest that these ratios are in the range of those observed in oil fields, refineries and power plants. The number of samples in the study is small, and correlations are weak [e.g., Figure 4 in (*11*)]. The largest limitation to isotopic attribution is instrumentation. With small enhancements of the ambient CH_4 background, there are only very small perturbations to the isotopic signature, and present instrumentation is challenged to detect this. This study was unable to distinguish between natural geologic CH_4 or CH_4 from refineries, NG, or oil refining.

A debate over the Pétron *et al*. study (*3, 25, 32*) centers on assumptions made in convolving inventory estimates of emissions, and the composition of such emissions, with atmospheric observations to produce estimated emissions of CH_4. Two conclusions can be drawn from this discussion: first, alkane signatures robustly indicate the presence of CH_4 from NG; and second, convolving top-down methods with bottom-up methods entrains bottom-up uncertainty into the top-down estimate. Without robustly superior inventory information (such as for CO emissions in California) such a convolution is not definitive.

Another method to address the problem of attribution is to use alkane signatures to constrain the sources of emissions. In simple terms, biological systems produce CH_4 in anaerobic metabolic pathways, but no comparable large-scale production pathways exist

for C_2H_6 and C_3H_8. Xiao *et al*. use global C_2H_6 measurements in this way (*8*). Wennberg *et al*. (*7*) use C_2H_6 to constrain CH_4 emissions in the LA basin, and find that this strongly suggests that the missing CH_4 source is fossil in origin. Katzenstein *et al*. use a complete HC signature with higher HCs of various types, which point to a fossil source for CH_4 over the south-central United States (*9*). At large scales, Miller *et al*. argue that continental-level CH_4 to C_3H_8 ratios imply a fossil source for the excess CH_4 (*6*).

Lastly, CH_4 can be attributed to sources in some study designs by simple proximity (spatial and/or temporal). That is, CH_4 observed near a given facility can be assumed to be emitted from that facility. Such attribution was used to eliminate dairy farm emissions from ARCTAS flight data in the Wennberg *et al*. study (*7*). Similar results have been found in NOAA driving campaigns (*3*) and in calibrating isotopic signatures to sources of various types (*11*). This approach was also used in recent work in the Uintah basin to distinguish oil-associated CH_4 from natural-gas-derived CH_4 (*45*).

2.1.5 Uncertainty in top-down measurements

2.1.5.1 CH_4 measurement uncertainty

Most CH_4 observations made in studies analyzed here are made either via whole air flask samples measured in the lab by gas chromatography (*5*), direct absorption spectroscopy (such as used in aircraft), or spectroscopy of an air column [such as in (*12*)]. Flask samples and direct absorption spectroscopy typically have accuracies of 2 ppb or better (1-sigma), and upward-looking FTS observations of 3.5 ppb (1-sigma). These measurement uncertainties do not have substantial impact on the overall uncertainty of flux estimations made employing these observations.

2.1.5.2 Overall assessment of source flux computation uncertainty

Quantitative simulation of atmospheric transport represents the main source of uncertainty in top-down methods. Recent studies have endeavored to minimize this problem by combining surface and tower measurements with aircraft profiles and total column observations. The column concentration data provide a direct measure of the total mass of CH_4 in the atmosphere, and spatial gradients of column amounts define the total mass excess in source regions. On sufficiently large scales (e.g. TX/OK/KS), the flux derived from an inverse model depends on the measurement of this excess and the synoptic scale winds, which are in general well constrained by meteorological data. For example, Miller *et al*. (*6*) quote uncertainties of about 12% for the South-Central region of the United States. Uncertainties in individual flux estimates for smaller regions or less dense samples are larger, >50% in some cases.

2.1.5.3 Uncertainty in alignment with inventories

There are challenges in the alignment of atmospheric studies with inventory definitions. For example, oil and gas wells are often located in the same air basin. If oil wells contribute to ambient CH_4 concentrations, but all emissions are attributed to NG activities, then NG emissions could be significantly overestimated (emissions of CH_4 from oil-associated gas production are classified as petroleum sector emissions in the GHGI).

A major hindrance in this area is a lack of understanding of the compositions of produced and sold gases. For example, uncertainties in attribution of CH_4 to NG in Wennberg (*7*) were dominated by uncertainty in α, the ratio of C_2H_6 to CH_4 in the sold NG [p. 9287 in (*7*)]. Some studies attempt to use raw and processed gas compositions to better discern the stage of gas emissions (*2*). Unfortunately, such methods of attribution depend strongly on the accuracy of gas composition data. Gas composition can vary quite significantly from field to field, as observed in Colorado (*3*), from California oil fields [figure 3.10 in (*54*)], and in the Barnett shale (*55*).

2.1.5.4 Other sources of variability or error

Natural gas emissions are both spatially and temporally variable. Spatial variability can exist at the scale of wells, fields, or "plays" (productive geologic formations). Temporal variability includes emission variations that result from variations in the annual rate of well workovers and well completions and variations in vent times and the number of vents per vented wells.

The impact of temporal variability on top-down studies is not well explored. This is likely to be a larger problem at small scale (e.g., basin or city measurements) than at large scale (due to mixing across days at continental scale that will smooth stochastic variations). Two types of confounding temporal variation could occur. First, there could be correlation between sampling time and emissions times. A significant fraction of recent EPA GHGI emissions came from liquids unloading events. Allen *et al*. (*26*) also found significant potential emissions from liquids unloading. Because some of these liquids unloading events are manual or semi-manual in nature, they will tend to be performed more commonly in daylight hours. Also, unloading of condensate from tanks into trucks in liquids-rich plays would likely be similar in their time profiles.

Aircraft measurements are typically performed in the afternoon, because a well-developed boundary layer and well-mixed conditions found in afternoons are easier to simulate. Therefore, afternoon measurements might see a larger signal from daytime liquids unloading (or other maintenance) activities. However, this is unlikely for observations that span the whole atmospheric column, for which the turnover time is measured in days. Compressor emissions or drilling rig emissions will tend to be more constant in flux, and would not contribute to this possible distortion (e.g., drilling rigs operate 24 hours per day).

Second, there are emissions occurrences that are one-time or rare, such as venting emissions during well flowback. Since these occur once or a few times in a well's

lifetime, missing or including this source within a short time of a single aircraft flight could lead to an under or overestimate of emissions. We believe that this is likely to be a small effect because drilling rigs tend to operate on a continuous basis, so in a large basin, there will likely be a relatively steady well completion and flowback rate (e.g., one well completed per day on average). Also, large scale studies such as Kort (*7*) and Miller (*6*) have used data from many aircraft flights over extended spatial scales and long time intervals.

Analysis of the variability associated with each of these sources could be addressed by constructing a hierarchical model developed using Bayesian statistics [see, for example (*56*)]. This approach may be especially useful when data are available from several levels of observational units (e.g., wells, fields, or formations).

2.1.5.5 Coverage and representativeness of atmospheric studies

An important parameter for atmospheric studies included in the first text chart is the coverage of the studies, including both spatial scale and density of measurements. Due to limited sampling funds and efforts, atmospheric studies must rely on a limited number of samples. Thus, regional- and state-level studies tend to have more dense measurements over a smaller area, while national-scale measurements tend to rely on more spatially and temporally sparse measurements taken over a larger area. Table S5 lists all atmospheric studies included in the first text chart, along with their fractional coverage of U.S. gas non-associated gas production and gas consumption. In addition, the number of samples or sampling campaign length and frequency are given for included studies. Large-scale studies cover wide spatial extent (e.g., all United States), with variable fidelity in a given region depending on the local sampling density. For example, see Miller *et al*. [figure S3 in (*6*)] for spatial representation of sample coverage.

Table S5. Atmospheric studies: spatial coverage and sample size

Study	Region	Frac. of U.S. non-assoc. gas prod. in study region	Frac. of U.S. gas cons. in study region	Number of samples
Miller *et al*.	US	100%	100%	~12,000
	South-central US	45.5%	18.5%	~900 tower obs. Flight obs. not given
Kort *et al*.	US	100%	100%	~300
Katzenstein *et al*.	South-central US	37.7%	21.8%	~300
Wang *et al*.	US	100%	100%	Not given
Xiao *et al*.	US	100%	100%	Not given
Pétron *et al*.	Denver-Julesberg basin	0.41%	0.08%	~400–500 tower ~100 ground.
Wunch *et al*.	SoCAB	0.002%	4.63%	131 days - Continuous at 0.5 Hz
Hsu *et al*.	Los Angeles County within SoCAB	0.002%	2.97%	36 days - Canisters at 1/hr - Continuous at 1 Hz
Wennberg *et al*.	SoCAB	0.002%	4.63%	See Hsu and Wunch Additional: 4 flights
Peischl *et al*.	SoCAB	0.0005%	4.32%	16 flights - Continuous at 1Hz - Flasks, freq. unknown
Karion *et al*.	Uintah basin	1.42%	0.01%	1 flight - Continuous at 0.5 Hz.

2.2 Bottom-up methods: Inventories and emissions factors

The U.S. Environmental Protection Agency (EPA) creates a yearly greenhouse gas inventory (GHGI) of CH_4 emissions from NG systems (*17*). The methodology used has been relatively consistent since the 1990s, with the methods outlined in summary form (*57*) and in a detailed 15 volume report (*14, 15, 50, 58–69*). The EPA GHGI follows methodologies outlined in Intergovernmental Panel on Climate Change (IPCC) guidelines (*70*).

The general approach of the EPA GHGI is to generate emissions factors (EFs) for typical sources in each industry segment or stratum. In some cases, EFs are used to generate facility-level emission rates. In all cases, EFs are multiplied by a population (e.g., number of compressors) or activity factor (e.g., SCF transmitted) to develop an emissions rate for a given segment or stratum.

2.2.1 EPA/GRI study

2.2.1.1 Study timeline and goals

For the last ~20 years the EPA GHGI has largely relied upon a detailed study undertaken from March 1991 - June 1996 by EPA and the Gas Research Institute (GRI), with work performed by a number of sub-contractors. The methods are outlined in summary form (*58*) and in a detailed 15 volume report (*14, 15, 50, 58–69*). Additional reports were published by GRI, some of which are not currently available for purchase.

The original EPA/GRI study modeled national emissions of CH_4 for 1992 at 314 + 105 Bscf (+ 33% at 95% confidence level). This leakage rate was equivalent to 1.4 + 0.5% of gross NG production. Since a key goal of the study was to estimate leakage to + 0.5% of gross production, the study authors considered the estimate a success.

A major implication of the EPA/GRI study was that NG contributes less to global warming than coal or oil – and that fuel switching is supported as a GHG mitigation method (*57*).

2.2.1.2 Structure of the EPA/GRI study

The EPA/GRI study (*50*) included all segments of the NG industry, including: production, gas processing, transportation, storage, and distribution (*57*). It did not include leakage beyond the consumer or industry meter. Each industry segment was analyzed to determine important equipment types (e.g., separators, glycol dehydrators). Expertise was applied to determine the largest potential sources, and these sources were targeted for more thorough sampling campaigns (*15*).

On-site measurements are used to project national emissions from a source by generating EFs. The relevant equation for scaling EFs is as follows:

$E = EF \times AF$

Where E is emissions from a given source (e.g., MCF/year), EF is the emissions factor (e.g., MCF/activity) and AF is the activity factor (activities/year). This is a simplification: a variety of structures are used for the activity factor, depending on the industry segment.

Equipment source emissions measurements underlying the EFs were made using a variety of techniques. EPA Method 21 was used in the EPA/GRI device-level studies (*50*). In Method 21, a correlation technique is used: local CH_4 concentration is measured, and this concentration is related to an emission rate by a correlation equation. Correlations are generated with direct flux measurement, tracer gas methods, or leak statistics methods. A major concern with screening techniques is the uncertainty associated with screening correlations (*15, 21, 60*). Generally, the instrument used for local concentration measurements is an organic vapor analyzer that uses flame ionization detection (FID), with concentration sensitivity ranges between 10,000 to 100,000 ppmv.

Alternatively, a more labor-intensive technique used was the total enclosure technique (bagging), which can be used to measure the flux over time into an enclosed volume, generating a leakage rate. In bagging, the leaking area is isolated and uncontaminated air is blown through an area containing the component.

EFs were determined from average emissions rates for basic components such as valves, flanges, seals and other connectors (*76–80*). Nearly 200,000 components were sampled at 33 facilities throughout the country.

The AF is the total number of sources (e.g., number of compressors) or the total amount of infrastructure (e.g., miles of transmission pipeline), in the target population. In some cases, equipment AFs are not well known. To extrapolate equipment activity factors representing regional or national averages, “extrapolation parameters” (EP) are used (*57*).

2.2.1.3 Categories of emissions in EPA/GRI study

Three main categories of emissions sources were outlined the EPA/GRI (*50*) study:

1. Vented and combustion sources (purposeful, consistent sources);
2. Blowdown and purge emissions (purposeful emissions, but sporadic and unplanned due to process upsets); and
3. Leaks (so-called “fugitive”, or unplanned emissions).

Vented emissions result from devices that vent some fraction of the gas they process. Examples include pneumatic devices, and dehydrator CH_4 slippage emissions. Blowdown and purge emissions are a subset of vented emissions. ‘Blowdown’ refers to venting of gas contained inside a pressure vessel, pipeline, or other piece of equipment. Purge is the process of clearing air from a piece of equipment by displacement with NG. Through

purging, some NG is emitted along with the air to the atmosphere. Equipment leaks result from malfunctioning or poor operation of equipment and are not part of designed operational profiles. Leaks are estimated using an EF and AF approach by counts of basic components in a facility (e.g., valves, seals).

2.2.1.4 Challenges with bias and uncertainty in EPA/GRI study and resulting inventory uncertainty

Current inventory methods rely on a set of key assumptions: a) samples used to generate device-level EFs are drawn from the same population whose emissions are being estimated; b) samples are of sufficient size to characterize the population, given heterogeneities in technologies and managerial practice; c) leakage volumes are not dominated by abnormal leakage rates or outlier events; d) activity counts are known with reasonable certainty; and e) all major sources of emissions are included in inventories. Current methods do not satisfy these assumptions, with varying degrees of uncertainty existing for each assumption.

2.2.1.4.1 Challenges with obtaining representative samples

With regard to the first challenge, the EPA/GRI study (*50*) authors devoted significant effort into obtaining an unbiased sample when generating EFs. The EPA/GRI methodological papers note that the sampling approach used was not truly random (*15, 50, 57, 60*). The sampling method used could be biased if:

1. The list of operators used to choose sites was not representative of the population of operators, or if facilities or technologies run by operators were not representative of the population of facilities (e.g., gas type, equipment vintage, equipment type, etc.).
2. The facilities which allowed on-site sampling were not representative of the typical facility;
3. The facilities sampled were altered before testing.

First, the list of operators might not be representative of all operators. A related problem would be if facilities included on sampling list were not representative of the total population of facilities. This problem could be significantly worse in the current day, given a shift in producing regions and producing technologies since the early 1990s (i.e., a list that was originally representative becomes less so over time). Bias in technology vintage may affect current use of EFs. Older equipment has been shown to have higher leak rates. For example, Clearstone *et al*. (*15*) found that equipment over 30 years of age had leakage rates over 5 times larger (kg of CH_4 per MMSCF of gas plant throughput) than equipment under 30 years of age [figure 21 in (*15*)]. This could lead to bias in current day use of EFs from EPA/GRI (*51*), even if no such vintage bias existed at the time of the study. In the opposite direction, there is significant new infrastructure in some regions that were not major gas producing regions until the recent shale gas expansion (e.g., Pennsylvania).

Second, it is possible that facilities which allowed on-site sampling were not representative of the population. All else being equal, it is plausible that operators with effective leak detection and repair programs were more likely to allow EPA-affiliated scientists on site to measure leaks. In the study of pipeline leaks, 30 companies were invited to participate, of which nine agreed to participate, and six eventually provided data [p. 5 in (*58*)]. Kirchgessner *et al*. note that "the companies contacted were not required to participate" (*58*). Also, contacted companies were asked to select a representative site for sampling (*15*). It is also plausible that companies may have selected sites with less problematic operations.

Third, it is possible that operators, expecting a visit from EPA scientists, might perform a check of their facility for leaks, at least to reduce any obvious large leaks that might pose safety concerns. Because (as shown below), a few large emissions sources generally contribute a significant share of emissions, even cursory "tune up" maintenance performed in anticipation of future sampling could cause significant bias in emissions estimates from the sampled facilities.

Ultimately, the direction and magnitude of sampling bias in the EPA/GRI study cannot be known. The above factors suggest that it is possible that the EPA/GRI samples were taken from a select population with an emissions distribution having a smaller mean rate and less variability (e.g., less likelihood of extreme leakage rates) than the total population. Importantly, this study design would be considered unacceptable in other fields, e.g. observational biomedical research, because of the potential for generating bias.

2.2.1.4.2 Challenges with sample size requirements

A fundamental challenge exists in sampling for emissions rates: the NG industry is large and diverse, and sampling is expensive. Given these challenges, the EPA/GRI study made significant sampling efforts (e.g., >200,000 sources sampled) that have not been surpassed to date. It was noted in the EPA/GRI study that sample sizes were limited by funding availability.

EFs are ideally defined as the average measured emissions from a large number of randomly selected sources in a source category. Stratification can be applied usefully to mitigate variability due to smaller-than-ideal sample sizes. Stratification reduces the number of samples required to reach a given level of uncertainty. This method was applied extensively in the original EPA/GRI sampling design (*20*). An example of stratification is given by compressor size: one can segment compressor drivers by the horsepower of the driver, and measure devices in each stratum. However, if reliable AFs are not collected by stratum then stratification does not improve accuracy (e.g., if yearly inventory compilation efforts will not have access to accurate estimates of numbers of compressors operating in each stratum). Stratification may provide additional insights into variability between and within source categories, which is of interest in determining emissions distributions (see below).

2.2.1.4.3 Challenges with highly heterogeneous emissions rates

The distribution of emissions between sources can affect the accuracy of sampling-derived EFs (*20*), independent of other biases in sampling. If total emissions are dominated by a small number of rare high emitters (e.g., "heavy-tailed" distributions), the construction of EFs using simple arithmetic means of the sampled population is problematic. Limited sampling from such distributions to generate a mean emissions rate has a higher probability of underestimating means than overestimating them. This challenge was recognized in the EPA/GRI study (*20*), but it was thought unimportant.

Evidence suggests that a small percentage of sources can be responsible for the majority of emissions at a given facility (*15, 28, 72–75*). For example, the Clearstone report noted that only 2.2% of sampled components were leaking above the detection limit (1,629 out of 74,438 sampled components registered local concentrations above 10,000 ppm). They also report that the largest 10 leaks (i.e., the top 0.013% of surveyed components) were responsible for 58% of the gas leakage. Also, Clearstone noted that the "top 10 leaks [at a facility] typically contribute more than 80% of emissions from leaks", for cases where many thousands of devices were sampled (*74*), indicating that these outlier-dominated distributions are characteristic.

Table S6 describes the details of various studies with findings of "super-emitting sources"

Table S6. Evidence of heterogeneity of emissions magnitudes across studies.

Study name	Industry stage	Measurement technique	Degree of heterogeneity noted	Pages with relevant statistics, tables, or quotes.
Allen *et al*. 2013	Production	Direct measurement of unloading events	"Four of nine events contribute more than 95% of total emissions"	Article p. 3.
Alvarez 2012	Production	Analysis of reported emissions	"10% of well sites accounted for 70% of emissions"	Article p. 3. Also see SI dataset in Microsoft Excel format.
Chambers 2006	Processing	Down-wind differential absorption LIDAR	"At plant B a single intermittent leak from a pressure relief valve was located that increased site emissions from 104 kg/hr to 450 kg/h."	p. 6
Clearstone 2002	Processing	Direct measurement using Hi-Flow sampler	>100,000 devices sampled across 4 facilities. between 35.7% and 64.6% of leakage from each facility was found leaking from top 10 leaks in each facility.	Executive summary, Table 4 (p. 24).
Cormack, 2007	Transmission compressors	Direct measurement with Hi-Flow	Top single leak accounted 40% of leakage. Top 20% of leaking components accounted for 80% of leakage.	Figure p. 15
Harrison *et al*. 2011	Compressor stations	IR camera, Hi-Flow sampler	Reported data in Appendix B show outliers. For example, ~2,800 valves and flanges were screened with IR camera and 29 leaks were found. The single largest of these leaks (>1000 mscf/year) is >100,000 times larger than valve and flange EF (0.05 or 0.09 mscf/year). Similar results seen elsewhere. See, e.g., blowdown line leaks from centrifugal compressors (table B2) where largest leak represents 70% of the total leakage.	See tabular data in Appendix B.
NGML, Clearstone, IES 2006	Processing, well sites, gathering compressor stations	Direct measurement using Hi-Flow sampler and optical methods	> 74,000 components sampled. Approx. 1600 were found to be leaking (~2%). From executive summary: "Repairs to 10 largest emitting cost-effective-to-repair components at each site…would reduce natural gas losses by approximately…58%"	Executive summary (p. iii). For details, see Appendix 1 (separate PDF) which ranks leaks by emissions rate for ~1600 leaking sources.
Picard, 2005	All stages	Sampling via various methods	"Top 10 leaks typically contribute more than 80% of emissions from leaks."	p. 3
Shorter, 1997	All stages	Remote sampling via tracer methods	Repeated evidence of skewed emissions distributions: See tables 1–7. Evidence includes: top emitters of size 100x to 10,000x larger than small emitters (table 9); standard deviations in excess of mean emissions rate in many cases, indicating heavy-tailed distribution (table 7).	Tables 1-9
Trefiak 2006	Compressor stations and gas plants	Optical measurement and Hi-Flow sampler	23% of the 144 fugitive emissions sources were responsible for 77% of leakage.	Fig. 2.1

Unfortunately, only a small number of datasets report emissions measurements in sufficient detail to allow analysis of distributions of emissions across sources. Figure S2 plots emission distributions known by the authors at the time of review (*15, 18, 28*). In Figure S2(b), the Harrison population distributions are plotted as multiples of the mean due to the different magnitudes of emissions.

Figure S3 shows the impacts of these distributions. In Figure S3(a) we plot from the NGML/Clearstone report (*15*) the counts of emissions sources by emissions strength. In Figure S3(b) we show the contribution to total emissions volume. As can be seen, the tail of the emissions source distribution (sources of magnitude >10 MSCF/d) are a small fraction of the total sources, but a large fraction of the total emissions rate.

2.2.1.4.4 Challenges with uncertainty or unavailability of activity data

Next, challenges exist if population data and AFs are inaccurate or biased. A recent study found that active well counts differ by up to 30% in EIA and EPA datasets [p. 37 in (*71*)], with EPA datasets being low in well numbers. Also, numbers of wells completed for 2010 were found to differ significantly between EPA (4871 wells, of which 86% were hydraulically fractured) and industry (IHS) datasets (18542 wells, of which 61% hydraulically fractured) [table 3 in (*71*)].

The use of HPDI (beginning in the 2011/2013 GHGI) as a primary data source should greatly improve the quality of activity data in the GHGI, particularly regarding wells that are hydraulically fractured. HPDI (as well as IHS and other commercial datasets), use multiple data sources - including company data - to build their well populations. Quality control for these datasets is likely to be better than state-level public datasets.

2.2.1.4.5 Challenges with missing sources from inventories

Finally, not all potential CH_4 sources from NG use are included in the EPA GHGI as NG sources. This could cause mis-attribution of emissions. For example, emissions from the production well phase of petroleum and associated gas are not included in NG inventories, but are instead classified as petroleum emissions. Also, emissions downstream of consumer or industrial meters are not included in the NG inventory, but are instead included as stationary source or transport emissions [p. 1 in (*15*)]. These emissions were estimated to be potentially of significant in scale in recent work (*7*). Also, some sources noted above (e.g., abandoned wells or other derelict infrastructure) are not included in the inventory, an oversight of unknown importance.

There is no single unique, “correct” way to draw boundaries between industry segments. However, confusion can be created if researchers report estimated emissions rates that do not align with EPA boundary definitions when comparisons with the GHGI are then made (by the authors or others).

2.2.2 Current EPA GHGI methodologies and results

The current EPA GHGI methodology was released in 2013, covering up to the 2011 calendar year. We therefore refer to this inventory as the EPA 2011/2013 inventory. Estimation methods used in this inventory are not significantly changed from the methods developed in the EPA/GRI (*50*) study, although many small changes have been introduced in the intervening time period (e.g., improved or updated EFs or added source categories).

The current methodology uses a three-step process. Step 1 estimates "potential" CH_4 emissions. Step 2 adjusts emissions for reductions resulting from regulatory requirements and voluntary actions (e.g., those reported through the Natural Gas STAR program). Step 3 calculates the "net" emissions by subtracting emissions reductions from the potential emissions estimate.

The potential CH_4 emissions estimate (Step 1) is established by combining activity data with potential emission factors for the various processes, stages and components that make up the NG system. This is largely the method developed in the EPA/GRI study. The activity data are taken from a variety of Federal, State and commercial sources. The results of the 2011/2013 EPA GHGI, estimating for calendar year 2011 CH_4 emissions from the NG industry, are shown in Figure S1.

The 2011/2013 EPA GHGI incorporates changes to both the EFs and AF data sources. These changes were made to reflect contemporary field practice, and more accurately characterize well populations. EFs for liquids unloading, condensate storage tanks and centrifugal compressors were updated from the 2012 GHGI.

The 2011/2013 GHGI adopted new net emission factors for liquids unloading from wells with and without plunger lifts. These EFs use data from an API/ANGA survey (*71*) of practices from ~90,000 wells. The 2011/2013 GHGI also modifies the potential EF used for well completions and workovers with hydraulic fracturing (from 9,175 Mscf per completion/workover to 9,000 Mscf).

The 2011/2013 GHGI used new activity data. The GHGI now uses the commercial data aggregation database HPDI (*40*). This provides EPA with more comprehensive and up-to-date data on U.S. well populations. An important change resulting from the use of HPDI data is that the number of wells hydraulically fractured has been revised upwards (from 50,434 non-associated wells in 2012 to 178,647 in 2013). The 2011/2013 GHGI also assumes a 1% yearly refracturing rate for these wells. The previous assumption was a 10% annual refracturing rate. The increase in well count in the 2011/2013 GHGI resulted in increased emissions from hydraulic fracturing (completions and refractures).

The method for calculating emission reductions due to regulation and voluntary actions in the 2011/2013 GHGI remains unchanged. In the 2011/2013 GHGI, however, EPA assumes that 15% of U.S. hydraulically fractured well completions are subject to specific

state regulation requiring controls. This is a downward revision resulting from the HPDI database providing a more comprehensive picture of where hydraulically fractured wells were located.

2.2.3 Bottom-up studies performed since EPA/GRI 1996

Several bottom-up studies have been performed since the EPA/GRI study of 1996. These studies are relatively few in number, due to the expense and challenge of making bottom-up measurements and a lack of attention on CH_4 emissions from NG systems during this time period. We note that a number of private studies likely occurred during this time period that we were unable to access.

2.2.3.1 Chambers et al.

Chambers *et al*. produced two reports studying emissions from NG processing plants in Canada (*14, 72*). These reports are both focused on studies performed at a small set of sweet and sour gas processing plants. The study used numerous technologies to determine leakage rates. First, leaks were visualized using a Hawk gas leak imaging camera.

Second, leakage volumes were quantified using a downwind transect with differential adsorption lidar (DIAL) technology. Lidar (sometimes described as "Light Detection and Ranging") technology is a backscatter-based technology, wherein laser light of a given wavelength is reflected back to the sensor by the substance of interest, and the time to return to the sensor is recorded to estimate distance to the substance. By utilizing two light wavelengths, one strongly absorbed by the species of interest, DIAL allows the calculation of gas concentration as a function of distance from the DIAL source. DIAL therefore produces a 2-dimensional "slice" or transect of gas concentrations across a plane downwind from the facility. When coupled with simultaneous wind measurements at the same site, the mass flux of emissions can be estimated across the plane.

Chambers *et al*. found that emissions from gas plants were generally higher than the predicted quantities generated using the CAPP (Canadian Association of Petroleum Producers) detailed emissions estimation methods.

2.2.3.2 Harrison et al.

Harrison *et al*. (*18*) produced a draft report for improving default emissions factors for compressors and other equipment under an EPA contract. The key focus of the study was on reciprocating and centrifugal compressors, including transmission, boosting/gathering and gas processing compressors. A smaller sampling effort including valves and flanges was also performed.

The Harrison *et al*. team first identified leaking components using a FLIR infrared camera. The volume of gas emissions from these leaking components was then quantified

using a Hi-Flow sampler. The Hi-Flow sampler draws a large volume of air from the vicinity of the leak, ensuring that the total volume of the leak is drawn into the device. The concentration of gas measured in the flow through the device, coupled with the flow rate, provides an estimate of gas emissions rate (±10% accuracy).

2.2.3.3 Clearstone

The Clearstone study of 2002 (*15, 81*) was prepared for the Gas Technology Institute and the EPA. It focused on emissions at gas processing plants, examining four processing plants in a detailed device-level sampling effort. Over 100,000 individual sources were sampled as part of the study.

The Clearstone methodology surveyed all individual sources using bubble tests, portable hydrocarbon gas detectors, or ultrasonic leak detectors. The bubble test was the most commonly used test, due to rapid, low-cost application. Devices that were determined to be leaking (~2.6% of tested components) were then tested with either the Hi-Flow sampler (*82*) (if the leak volume was not larger than device capacity) or through bagging and direct measurement if the leak volume was larger than Hi-Flow capacity.

The results from the Clearstone study found that emissions factors were generally higher than those defined under EPA/GRI (50) and that a very large number of the leaks subject to direct measurement would be cost-effective to repair.

2.2.3.4 NGML et al.

In 2006, the National Gas Machinery Laboratory partnered with Clearstone Engineering Ltd. and Innovative Environmental Solutions to release a Phase II report (*21*), building off of the original Clearstone 2002 report. The Phase II project studied five more gas plants, in addition to examining well sites and gathering compressor stations.

In this study, nearly 75,000 individual components were examined. Results were similar to the first phase study: ~2.2% of the components were found to be leaking (i.e., had a screening concentration measured above 10,000 ppm). The methodology for the Phase II study was similar to Phase I, except for the addition of an IR camera method to perform rapid screening of leaking components.

Similarly to the Phase I study, the Phase II study found that EPA/GRI EFs, on balance, underestimated emissions from gas plants. They also reinforced the finding from Phase I that the vast majority of leaks are able to be repaired at no net cost to operators.

2.2.3.5 GTI

In 2009, the Gas Technology Institute performed a study on distribution system emissions (*16*). The GTI study focused on meters and metering/regulation stations.

Residential, commercial, and industrial sites were studied. Sample sizes were modest to large, with 2400 residential meters sampled, 393 commercial meters, and 46 industrial meters. Qualitative assessment of leaks was performed using bubble tests, portable pas detectors and the FLIR GasFindIR camera. After leaking components were found, quantitative mass flux estimates were generated with a Hi-Flow sampler.

GTI found that residential meter emissions factors were lower than previous EPA/GRI Tier 3 method, while commercial and residential meters had far higher emissions than previous EFs.

2.2.3.6 Allen et al.

Allen *et al*. (*26*) performed measurements of production-phase operations including: completions flowback during hydraulic fracturing operations, chemical pumps, pneumatic controllers, liquids unloading (with and without plunger lifts) well workovers, and other equipment leaks. Notably, Allen *et al*. devised novel containment strategies to directly measure emissions from hydraulic fracturing flowback (both with and without reduced emissions completions technologies in place), allowing much improved understanding of both potential and net (after mitigation) emissions from completions operations. For smaller sources Allen *et al*. used initial scans with infrared cameras, followed by Hi-Flow sampling to quantify flux.

In aggregate, Allen *et al*. found generally good agreement between EPA production-phase inventoried emissions and their estimates. However, their source-specific results differed significantly from EPA: their emissions during hydraulic fracturing were much lower than EPA estimates, while emissions from pumps, controllers, and other equipment leaks were higher. Importantly, Allen *et al*. note that liquids unloading present a potentially large and highly variable source of emissions, which require additional study.

2.3 Alignment between bottom-up and top-down studies: Attribution and system boundaries

A key challenge in aligning the results of top-down and bottom-up studies is that the boundaries of analysis can be quite different: what is observed in the atmosphere does not correspond or cohere to boundaries that are placed on an analysis performed for an inventory or life cycle assessment (LCA).

To illustrate the problem, consider the following case: assume an ideal atmospheric experiment that can correctly measure the CH_4 emissions from oil and gas operations and distinguish these emissions from not only biogenic sources, but also other anthropogenic CH_4 sources such as coal mines. Aligning the results from such a study to EPA estimates of NG emissions would still be challenged for the following reasons:

1. EPA definitions of the NG system do not include emissions of CH_4 from oil production operations. That is, if a well is classified as an “oil well”, its

emissions of CH_4 are counted as petroleum sector emissions, even though the associated gas from that well will enter (possibly after some local processing) the NG sector as per EPA definitions.

2. EPA definitions of emissions from the NG system do not debit any of the emissions from NG operations according to the fraction of co-produced liquids. That is, if a well produces NG condensate that is blended into the liquid fuel supply, no share of the CH_4 emitted from the "gas" well or processing equipment is allocated to the petroleum sector, even though a (potentially significant) fraction of the energy produced by the well would end up as products commonly conceived of as "petroleum" products.

This problem of allocation of emissions to "co-products" is a general one in environmental assessment, and has been a topic of interest and research in the life cycle assessment literature for many years. There is not a generally agreed-upon "correct" method for performing such allocation.

2.4 Dedication

We dedicate this article to George P. Mitchell. He was an innovator in recovering vented and stranded natural gas, and as a petroleum engineer and geologist, Mitchell was credited with leading development of hydraulic fracturing techniques.

George Mitchell was a dedicated and early supporter of the use of science to advance sustainability. He realized that there were finite energy resources and raw materials for our world. This understanding stimulated his sponsorship of the 1970s Club of Rome study, *The Limits to Growth*. He subsequently founded the Houston Advanced Research Center (HARC), which focuses on critical regional issues such as clean energy, air, and water. More recently, the Mitchells underwrote the National Academies' landmark report Our Common Journey: A Transition Toward Sustainability.

2.5 Advisory and Financial Disclosures

Financial: Significant financial interest (equity holdings or stock options) in any corporate entity dealing with the material or the subject matter of this contribution. Please disclose the entity and the nature and amount of the holding:

A.R. Brandt
No individual stocks in oil and gas companies. All financial interests in the oil and gas sector are contained in retirement accounts in broad (non-sector specific) index funds.

E.A. Kort
No individual stocks in oil & gas companies. All financial interests in oil and gas sector are via broad (non sector specific) index funds.

F. O'Sullivan
I do not hold any individual oil, gas or other energy sector securities. I passively hold mutual fund investments in index funds including energy-sector index funds.

D. Arent
Inherited mineral rights, not "significant". All other financial interests in blind/broad mutual funds.

D. Eardley
I have no stocks or bonds in the energy sector, except as part of broadly index-based retirement accounts (Fidelity, TIAA/CREF).

Paid Consulting: Within the last 3 years, receipt of consulting fees, honoraria, speaking fees, or expert testimony fees from entities that have a financial interest in the results and materials of this study. Please enumerate. *I have a paid consulting relationship, described as:*

A.R. Brandt
I have received pay as a consultant in the last 3 years for the following organizations: International Council on Clean Transportation (ICCT), European Commission: Directorate General – Climate, UK Department for International Development (DFID).

F. O'Sullivan
In the last 3 years, have participated in sponsored research supported by the following groups: Accenture, BP, Cyprus Institute, Enel, MIT Energy Initiative Seed Fund Program, NREL/Joint Institute for Strategic Energy Analysis, The Sloan Foundation, Weatherford International. In the last 3 years, I have received modest speaking and review honoraria from the following organizations: BP, B-Tec Foundation, Cambridge Energy Research Associates/IHS, Enagas, Environmental Defense Fund, EPRI, IEA

S.M. Jordaan
I was employed at the Electric Power Research Institute as a Project Manager from April 2012-April 2013. At EPRI, I was involved in the publication of a report on shale gas production released in 2013, which was funded by 10 companies in the electric sector. I consulted British Petroleum (BP) on technology innovation and water use of unconventional fuels for $2000, a one time fee for a presentation at a workshop in July 2013. The Energy Technology Innovation Policy Research group at Harvard (where I was a fellow from 2011-2012) receives funding from (in part) from BP. I have been engaged with NRDC most recently (December 2013) in a workshop on air and water impacts of unconventional gas. Travel and lodging were provided, but no speaking fees or honoraria were involved.

N.J. Brown
I consulted for API with a group of scholars to write a report on Policy relevant background ozone. I was paid a modest stipend to serve on an EPA advisory committee and a search committee.

Management/Advisory affiliations: Within the last 3 years, status as an officer, a member of the Board, or a member of an Advisory Committee of any entity engaged in activity related to the subject matter of this contribution. Please disclose the nature of these relationships and the financial arrangements. *I have a management/advisory relationship, described as:*

A.R. Brandt
I am currently serving on the Science Advisory Board for a study led by the California Council on Science and Technology that is examining environmental impacts of unconventional oil production technologies.

D. Arent
I served on National Petroleum Council NARD Study Policy Subcommittee I serve on committee to advise USGCRP at National Academy

N.J. Brown
I serve on a technical advisory committee for the DOE for Unconventional Resources. I receive no financial support except for travel expenses. I also serve on advisory committees for the EPA, and I receive a modest stipend for this along with travel expenses.

D. Eardley
Member, NAS/NRC panel on Inertial Confinement Fusion Energy Targets. Member, Joint LANL/LLNL Mission Committee. Member, JASON Study Group, U.S. Department of Defense and Department of Energy Member, Sandia National Lab External Review Panel for High Energy Density Science. I receive or have received fees for the above.

Advisory and Funding
Other Funding Sources: Additional funding sources not noted in the manuscript are:

A.R. Brandt
In the last 3 years, I have participated in sponsored research paid for by the following groups: California Air Resources Board; Argonne National Laboratory; Carnegie Endowment for Global Peace; Global Climate and Energy Project; Institute for Integrated Economic Research.

In the last 3 years, my students (PhD and MS) have received fellowship funding from the following sources: Stanford School of Earth Sciences Graduate Fellowship; Stanford Graduate Fellowship (University-wide fellowship); National Science Foundation (NSF); Chevron Inc. (through Stanford School of Earth Sciences); Illich-Sadowsky Stanford Interdisciplinary Graduate Fellowship (University-wide fellowship); Stanford University Undergraduate Summer Research Fellowship.

G. Pétron
GP's contribution to this manuscript was supported by NOAA and the NSF AirWaterGas Sustainability Research Network.

D. Eardley
I have received honoraria for participation in workshops at the Center for Urban Science and Progress, New York University.

3 Supplemental Figures

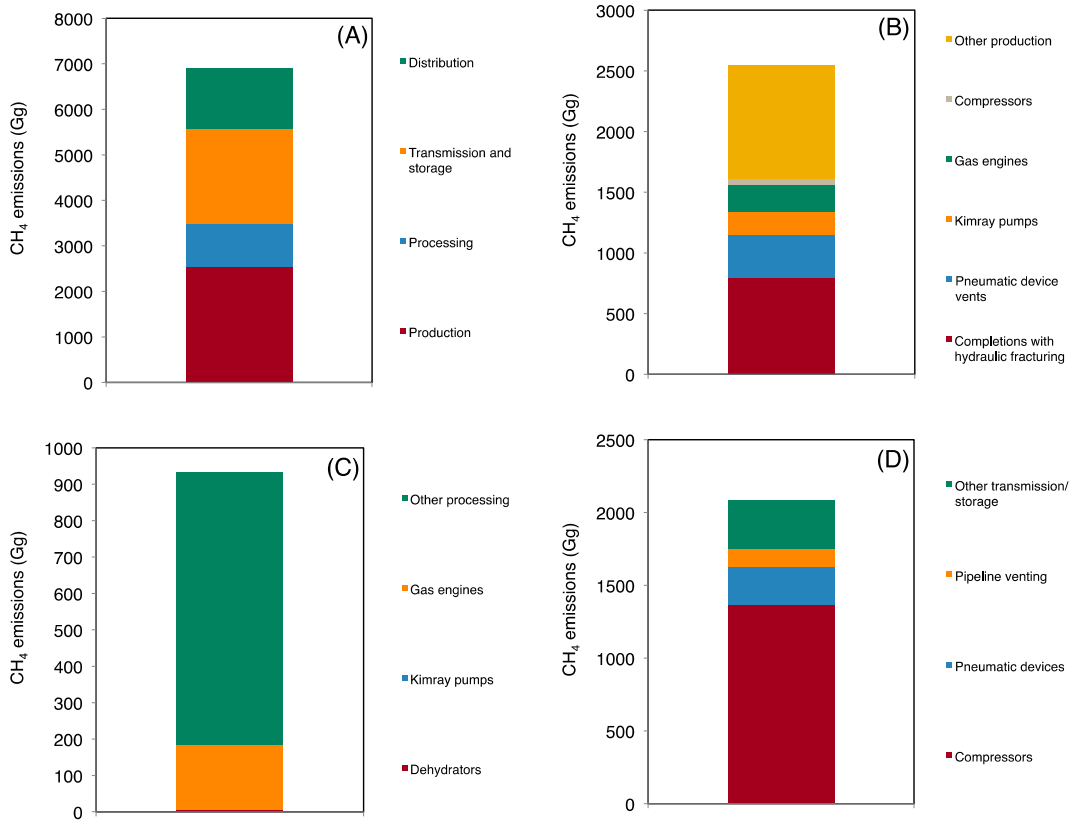

Fig. S1. CH_4 emissions from EPA 2011 inventory for the NG sector. (**A**) Total breakdown between four process stages. Sector specific breakdown for Production (**B**), Processing (**C**) and Transmission and Storage (**D**). Source: [table A-138 (*32*)].

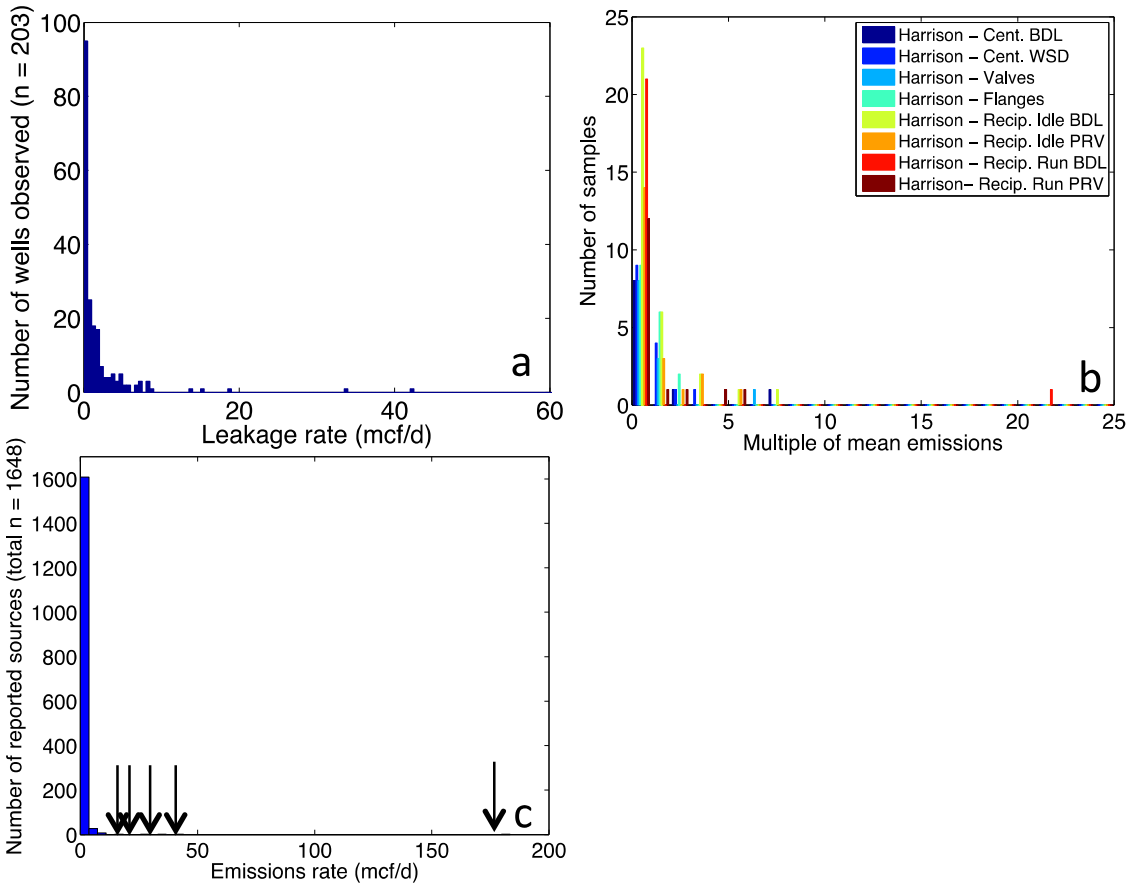

Fig. S2. (**a**) Observed distribution of leakage rates across 203 wells reported by Alvarez *et al.* (*28*). Original data from a study in Fort Worth region (*83*). (**b**) Observations of leakage rates by type of equipment and source from the Harrison *et al.* study (*13*). In the Harrison plot, emissions for each group of equipment are expressed as multiples of the mean emissions for that group. (**c**) Distribution of emissions across ~1600 extracted emissions rates from the Clearstone *et al.* study [Appendix I in (*16*)] Marked with black arrows are single sources that are unable to be seen due to the axis scale: high emitters at 10–50 mcf/d and one very high emitter above 150 mcf/d.

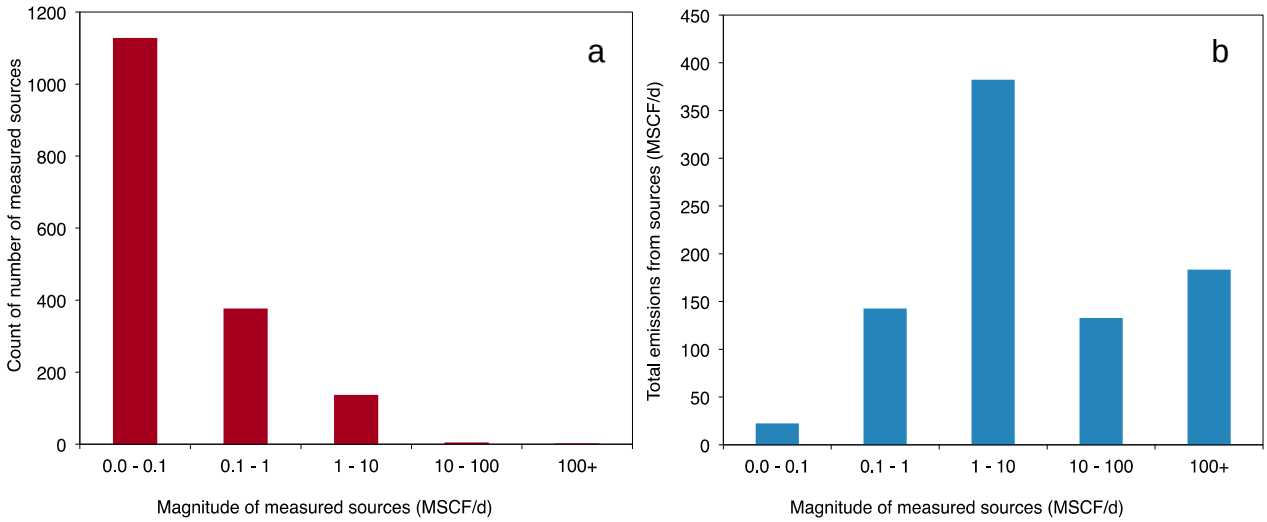

Fig. S3. Comparison of (**a**) the numbers of counts of sources by emissions strength, and (**b**) the volume of emissions by emissions strength for the NGML/Clearstone dataset (*16*). A small number of emissions sources in the tail of the emissions distribution (10+ MSCF/d) account for a large fraction of the total emissions.

4 References

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