

Methane emissions estimate from airborne measurements over a western United States natural gas field

Anna Karion,^{1,2} Colm Sweeney,^{1,2} Gabrielle Pétron,^{1,2} Gregory Frost,^{1,2} R. Michael Hardesty,^{1,2} Jonathan Kofler,^{1,2} Ben R. Miller,^{1,2} Tim Newberger,^{1,2} Sonja Wolter,^{1,2} Robert Banta,² Alan Brewer,² Ed Dlugokencky,² Patricia Lang,² Stephen A. Montzka,² Russell Schnell,² Pieter Tans,² Michael Trainer,² Robert Zamora,² and Stephen Conley³

Received 12 June 2013; revised 18 July 2013; accepted 23 July 2013; published 27 August 2013.

[1] Methane (CH₄) emissions from natural gas production are not well quantified and have the potential to offset the climate benefits of natural gas over other fossil fuels. We use atmospheric measurements in a mass balance approach to estimate CH₄ emissions of $55 \pm 15 \times 10^3 \text{ kg h}^{-1}$ from a natural gas and oil production field in Uintah County, Utah, on 1 day: 3 February 2012. This emission rate corresponds to 6.2%–11.7% (1 σ) of average hourly natural gas production in Uintah County in the month of February. This study demonstrates the mass balance technique as a valuable tool for estimating emissions from oil and gas production regions and illustrates the need for further atmospheric measurements to determine the representativeness of our single-day estimate and to better assess inventories of CH₄ emissions. **Citation:** Karion, A., et al. (2013), Methane emissions estimate from airborne measurements over a western United States natural gas field, *Geophys. Res. Lett.*, 40, 4393–4397, doi:10.1002/grl.50811.

1. Introduction

[2] As concern grows over the climate impact of increasing greenhouse gas emissions and the actual and associated political costs of imported fuels, the U.S. is looking to exploit natural gas as a domestic energy source. Natural gas is an efficient energy source because its combustion produces more energy per carbon dioxide (CO₂) molecule formed than coal or oil (177% and 140%, respectively) [*U.S. Department of Energy Energy Information Administration*, 1999]. Despite this efficiency, leakage of natural gas to the atmosphere from the point of extraction to the point of consumption reduces its climate benefits because the major component of natural gas is CH₄, a greenhouse gas that is 25 times more potent than CO₂ over a 100 year time horizon [*Intergovernmental Panel on Climate Change*, 2007]. Although assessing the exact climate impact of natural gas has many complexities, a recent

study has suggested that if more than 3.2% of natural gas leaks to the atmosphere on its way from the point of extraction to a gas-fired power plant, the electricity produced will have a larger immediate climate impact than that from a coal-fired plant [*Alvarez et al.*, 2012].

[3] A critical gap in determining the climate impact of the recent increase in U.S. natural gas production is the lack of accurate and reliable estimates of associated emissions. In particular, the methodology used to account for fugitive CH₄ emissions during production is in question. This is demonstrated by large year-to-year revisions in natural gas-related CH₄ emissions reported for 2008 by the U.S. Environmental Protection Agency (EPA), which caused the estimated national average production-sector leak rate for this year to increase from approximately 0.16% of production in the 2010 report to 1.42% in the 2011 and 2012 reports [*U.S. Environmental Protection Agency*, 2010, 2011, 2012]. This rate was revised back down to 0.88% in the 2013 report [*EPA*, 2013]. These changes were driven largely by changes in EPA's assumptions for calculating emissions from liquid unloading (removing the accumulation of fluids in gas wells), unconventional completions with hydraulic fracturing, and refracturing of natural gas wells. In particular, the main driver for the 2013 reduction in production emissions was a report prepared by the oil and gas industry, which contended that CH₄ emissions from liquid unloading were more than an order of magnitude lower than EPA's 2011 report estimate and that emissions from refracturing wells in tight sands or shale formations were less than half of EPA's 2011 report estimate [*Shires and Lev-On*, 2012]. The substantial changes in the CH₄ inventory between 2010 and 2013 have led the EPA's Office of Inspector General to release a report calling for the improvement of the agency's air emissions data for the natural gas production sector [*U.S. Environmental Protection Agency Office of Inspector General*, 2013].

[4] Such large revisions and differences in inventory-based emission estimates highlight an important point: most CH₄ emissions from oil and gas operations are estimated from the “bottom up,” in which emission factors for multiple processes are multiplied by an inventory of activity data. Most of the 80 different EPA emission factors associated with oil and gas operations are based on a study done in the 1990s [*Harrison et al.*, 1996] and assume consistency throughout the industry in a variety of different regions. In reality, the distribution of emissions may be highly variable from region to region [*Rusco*, 2010], and the recent revisions have suggested uncertainties in activity data and emission factors. Thus, there is a need to assess the emission factors

Additional supporting information may be found in the online version of this article.

¹Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, Colorado, USA.

²NOAA ESRL, Boulder, Colorado, USA.

³University of California, Davis, California, USA.

Corresponding author: C. Sweeney, Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO 80309, USA. (Colm.Sweeney@noaa.gov)

©2013. American Geophysical Union. All Rights Reserved.
0094-8276/13/10.1002/grl.50811

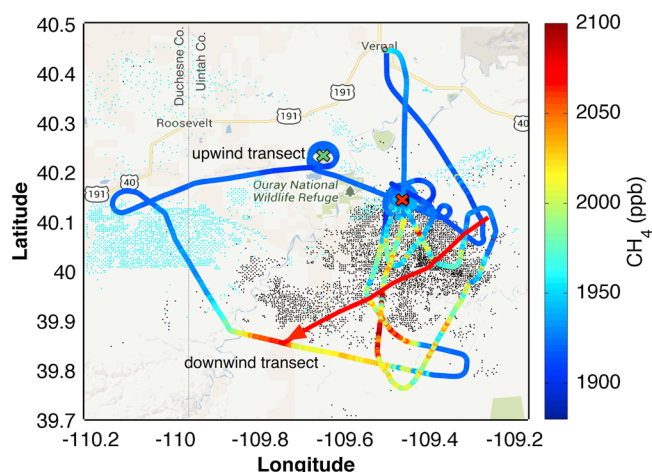


Figure 1. CH₄ measurements on 3 February 2012. Aircraft flight track overlaid on natural gas (black dots) and oil (blue dots) well locations along with color-coded CH₄ mole fraction. Bold red arrow shows the 3 h trajectory of the downwind air mass. The locations of two vertical profiles over Horse Pool (red X) and one northwest of Horse Pool (green X) are also indicated.

and extrapolation approaches used in bottom-up inventories with independent measurements and assessments of CH₄ emissions.

[5] Previous studies that have evaluated inventory estimates of oil and natural gas emissions [Katzenstein *et al.*, 2003; Pétron *et al.*, 2012] in a production basin with direct CH₄ measurements have concluded that CH₄ emissions from oil and gas production were likely underestimated by the available inventories. Because these studies took place in different U.S. regions (Oklahoma, Texas, and Kansas in Katzenstein *et al.* [2003] and Colorado in Pétron *et al.* [2012]) and over different time periods, it is difficult to assess to what extent this underestimate is found in all natural gas-producing regions or whether a trend is apparent. Here we present results from an oil and gas region not yet studied with atmosphere-based methods (the Uintah Basin) to the list of those that may have their CH₄ emissions underestimated by bottom-up inventories. The advantage of this study over previous ones is that the CH₄ emissions estimate does not require critical assumptions about either emission ratios using other trace gases or boundary layer flushing time.

2. Methods

2.1. Mass Balance Approach

[6] The mass balance approach is a measurement-based method for estimating the total emission of a trace gas released from a defined point [Ryerson *et al.*, 2001] or area source [Mays *et al.*, 2009; Turnbull *et al.*, 2011; White *et al.*, 1976], which allows for the direct assessment of uncertainties. The mass balance approach, as applied in this study, requires the assumption of steady horizontal winds, a well-developed convective planetary boundary layer (PBL), and measurements sufficiently downwind of the emission source; the uncertainties associated with these assumptions are identified and included in the uncertainty analysis (supplementary text section 4 in the supporting information). The Uintah County oil and gas field is well suited to this approach for deriving CH₄ fluxes using measurements from aircraft,

because the majority of the 4800 gas wells and nearly 1000 oil wells are concentrated in a relatively small area (40 × 60 km², Figure 1) (State of Utah Department of Natural Resources Division of Oil Gas and Mining, Well Information Query, 2012, http://oilgas.ogm.utah.gov/Data_Center/LiveData_Search/well_information.htm); an aircraft traveling at 60 m s⁻¹ is able to make several transects over the entire field and one to three vertical profiles during a 3–4 h flight.

[7] In the mass balance approach for flux estimation, the enhancement of the CH₄ mole fraction downwind of the source, relative to the upwind mole fraction, is integrated across the width of a horizontal plume in the planetary boundary layer (PBL) downwind of the source [White *et al.*, 1976]. When the mean horizontal wind speed and direction are steady during the transit of an air mass across an area, the resulting calculated flux is equal to the surface flux between upwind and downwind measurements. The CH₄ flux is derived to be

$$\text{flux}_{\text{CH}_4} = V \int_{-b}^b X_{\text{CH}_4} \left(\int_{z_{\text{ground}}}^{z_{\text{PBL}}} n_{\text{air}} dz \right) \cos \theta dx \quad (1)$$

[8] In equation (1), flux_{CH₄} represents the molar flux (moles s⁻¹) of CH₄ from the basin. V is the mean horizontal wind speed over the region, averaged over the altitude between the ground and the top of the PBL, and over the time an air mass transits the basin. The angle θ is the angle between the mean wind direction and the direction normal to the aircraft track downwind, so that $\cos \theta dx$ is the flight track increment perpendicular to the mean horizontal wind direction. The CH₄ enhancement over the background mole fraction, i.e., ΔX_{CH_4} , is integrated over the width of the plume ($-b$ to b) along the flight track and multiplied by the integral of the molar density of air (n_{air}) from the ground (z_{ground} , a function of path distance x) to the top of the PBL (z_{PBL}). In this calculation, ground-based heat flux measurements are used to characterize the mean time required to mix surface emissions from the ground to the top of the PBL (supplementary text section 4.3 in the supporting information).

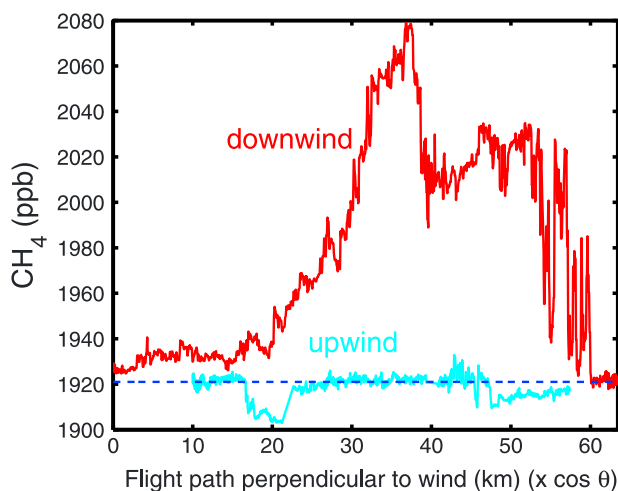


Figure 2. CH₄ mole fraction measured in the downwind plume (red line) as a function of distance perpendicular to the wind direction. The CH₄ mole fraction in the upwind transect is in light blue, and its average (1921 ppb) is represented by the dark blue dashed line. The lower upwind CH₄ measurements at ~20 km were made above the top of the PBL during a vertical profile.

2.2. 3 February 2012

[9] During 3 February 2012, moderate and steady horizontal winds and a well-defined PBL allowed us to use the mass balance approach to estimate the CH₄ emission flux from the Uintah County field. The CH₄ mole fraction was measured from an instrumented single-engine turboprop aircraft; and the PBL depth, wind speed, and wind direction were measured by high-resolution Doppler lidar (HRDL) (instrument details are in supplementary text sections 1–3 in the supporting information).

[10] Horizontal wind speeds on 3 February 2012 peaked during the night (2:00 local time (LT)) at 13 m s⁻¹ (averaged throughout the PBL), flushing out the basin before decreasing to a steady 5–6 m s⁻¹ from the northeast in the 3 h before the downwind transect was flown (at 15:30 LT). The PBL height (1700 ± 125 m meters above ground level (magl)) was determined from aircraft vertical profiles (Figure S1) and HRDL measurements. HRDL measurements showed the PBL height to be relatively constant throughout the time of the flight. Other than the vertical profiles, the rest of the flight measurements were made within the PBL between 100 and 1000 magl (Figure 1).

[11] The flight transect downwind of the natural gas field, along its southern and western edges and between 400 and 600 magl at 15:20–15:40 LT, showed elevated CH₄ mole fractions averaging 56 parts per billion (ppb) greater than the average upwind value of 1921 ± 5 ppb, with a peak enhancement of ~150 ppb. Horizontal winds from HRDL measurements averaged throughout the PBL were used to construct a back trajectory of the air mass sampled in this plume (Figure 1, red arrow). The trajectory indicates that the source of enhanced CH₄ was primarily the region containing the gas field in Uintah County and that the air mass traveled in a consistent southwesterly direction through the gas field in the ~3 h period prior to being sampled. Variability in the observed CH₄ mole fraction reflects the

extent to which a point source emission is horizontally and vertically mixed, with individual narrow plumes likely originating from point sources closer to the flight path than the sources of wider plumes. We integrated the CH₄ enhancement above the background value of 1921 ppb, which was derived from measurements made upwind of the location of oil and gas wells, along the downwind flight path to calculate the flux from the oil and gas basin (Figure 2 and equation (1)). The altitude-averaged wind speed and direction were also averaged over the approximate transit time of the air mass through the basin, from 12:40 to 15:40 LT, corresponding to nine individual HRDL profiles (HRDL provided wind measurements as 20 min averages).

[12] Based on the variability and uncertainty in each term of the mass balance equation, we derived a total uncertainty of ±27% (1σ) on the total CH₄ flux estimate on 3 February of 56 ± 15 × 10³ kg h⁻¹ (Table S1 and supplementary text section 4 in the supporting information). The relatively small uncertainty in the emission derived for this flight is the result of steady horizontal winds, consistent boundary layer height, and low measurement uncertainties.

2.3. Other Flight Days

[13] Twelve flights were made over the Uintah Basin in February 2012. Nonideal meteorological conditions (in particular, low, variable, and sometimes recirculating winds in the 0.5–1.5 m s⁻¹ range) on the 11 other flight days made direct mass balance analysis of CH₄ emissions impossible. For example, horizontal wind speed and direction measured at the ground site could not be assumed to be representative of winds throughout the basin on the days with low and variable winds, given the complex terrain-driven meteorology of the basin. CH₄ enhancements measured on the other flight days were large, however, with average mole fractions from 2030 to 2650 ppb inside the PBL (Figure S3). Flight tracks passing over the field on 7 and 18 February show increased CH₄ over the locations of the gas and oil wells, with several large and distinct enhancements, in addition to more uniform enhancements over the remainder of the field; there is no evidence that a single, large point source is responsible for all of the CH₄ emissions (Figure S3).

[14] Although no hydrocarbon measurements were made on the 3 February 2012 flight, analyses of 67 discrete whole air samples collected over Uintah County aboard the aircraft throughout the month of February 2012 show excellent correlations of propane (C₃H₈) and butane (C₄H₁₀) with CH₄ (R² > 0.85, Figures 3a and 3b). Correlations of CH₄ with carbon monoxide (CO), a tracer for vehicle exhaust, are weaker (R² = 0.28, increasing to 0.52 when a single outlier with high CO is removed from the analysis (Figure 3c)). The strong correlation of CH₄ with C₃H₈ and C₄H₁₀ suggests that these CH₄ enhancements were primarily the result of emissions from oil and gas operations [Pétron *et al.*, 2012].

3. Results

[15] Because of the low uncertainty and the fact that the basin was so well cleaned out by the high winds prior to our flight on 3 February, the derived emissions estimate from this day is the focus of this study. A flux of 1.4 ± 1.1 × 10³ kg CH₄ h⁻¹ (~2.5% of our 3 February estimate of 56 × 10³ kg CH₄ h⁻¹) was subtracted from the total flux to account for emissions from cattle and natural seepage, as estimated from

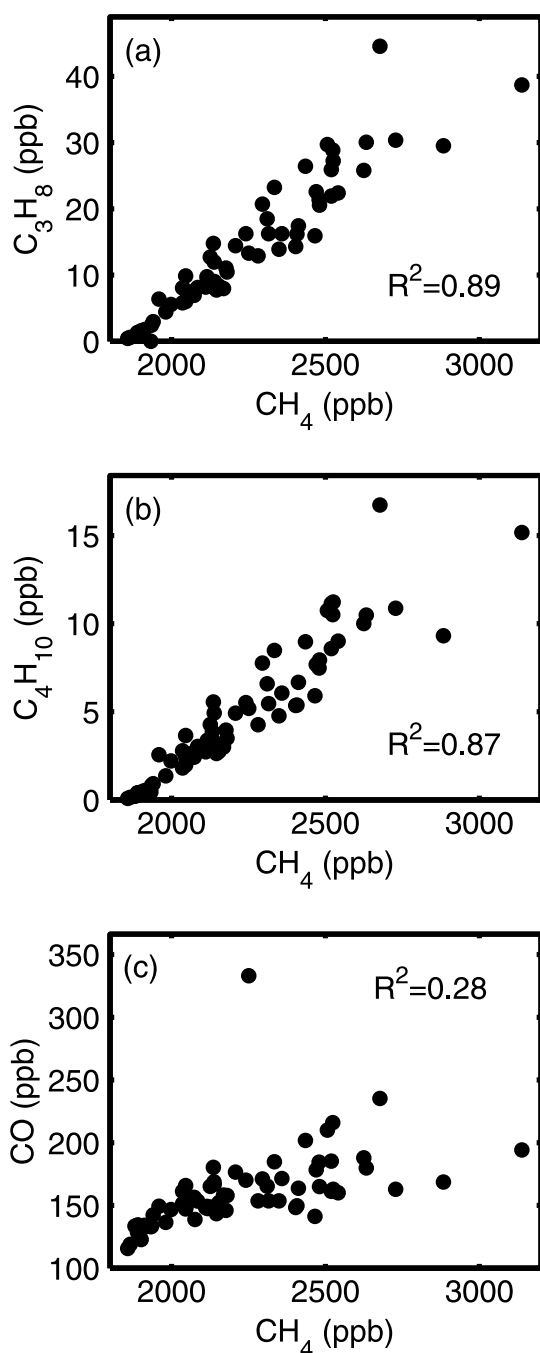


Figure 3. Mole fractions of (a) propane (C₃H₈), (b) butane (C₄H₁₀), and (c) carbon monoxide (CO) measured in discrete air samples collected over the Uintah Basin in February 2012, shown as functions of CH₄ mole fraction. Correlation coefficients (R^2) are shown in each panel.

inventories [Griffith *et al.*, 2008; Klusman, 2003; U.S. Department of Agriculture, 2009] (supplementary text section 5 in the supporting information), to give a total CH₄ emission of $54.6 \pm 15.5 \times 10^3$ kg CH₄ h⁻¹ from oil and natural gas sources on 3 February 2012. The oil and gas wells whose emissions were estimated from our flight transect are almost entirely contained in Uintah County (Figure 1), so we calculate the amount of raw natural gas that would correspond to our estimated CH₄ emission and compare it to the average hourly natural gas production from Uintah

County from both oil and gas wells (there is no coal bed CH₄ production in Uintah County). The total volume of natural gas produced from oil and gas wells in Uintah County in February 2012 was 7.1×10^8 m³ (from the Utah Department of Natural Resources Division of Oil, Gas and Mining at <https://fs.ogm.utah.gov/pub/Oil&Gas/Publications/Reports/Prod/County/>), or 1.0×10^6 m³ per hour on average. We convert our hourly CH₄ emissions estimate to natural gas units using a volume fraction of CH₄ in natural gas of 0.89 (composition profile for Uintah Basin raw natural gas from A. Bar-Ilan, personal communication, 2012) and the industry standard conditions (288.7 K and 101.3 kPa). Allowing for additional uncertainty on the production amount (estimated at 5% based on the average month-to-month variability in daily production) and on the composition of the emissions (estimated at 11% to encompass a realistic volume fraction of CH₄ from 0.79 to 0.99), the hourly emission rate we determined on 3 February 2012 corresponds to 6.2%–11.7% of the average hourly natural gas production from oil and gas wells in Uintah County during February 2012.

[16] Based on production data and publically available activity data, there is little evidence that emission magnitudes on 3 February were unusual relative to other days in January, February, or March 2012 (supplementary text section 6 in the supporting information and Figures S5 and S6). Furthermore, it should be noted that there are thousands of potential point sources (oil and gas wells, compressors, processing plants, etc.) in Uintah County and that there is no clear evidence in the data from our 12 flight days that a single point source is responsible for a large fraction of the emissions; we infer that it is unlikely that emissions differ drastically from one day to another. However, further work is needed to assess the variability of CH₄ emissions in this basin and to determine how representative our 1 day estimate is of Uintah's average natural gas leak rate.

4. Discussion

[17] Given the large global warming potential of CH₄, a natural gas leak rate of 6.2%–11.7% during production negates any short-term (<70 years) climate benefit of natural gas from this basin for electricity generation compared to coal and oil [Alvarez *et al.*, 2012; Howarth *et al.*, 2011]. This leakage also represents a potential economic loss and safety and air pollution hazard. An inventory analysis by the U.S. Government Accountability Office (GAO) suggests, however, that the fraction of natural gas emissions relative to production from the Uintah, a basin that produces approximately 1% of total U.S. natural gas, is atypical of many western U.S. basins. Using the Western Regional Air Partnership (WRAP) phase III [Bar-Ilan *et al.*, 2006] inventory and production numbers for 2006 from federal leases, the GAO estimates that the proportion of Uintah natural gas that is flared or vented is much greater (5% of production) than in surrounding regions, including the Denver-Julesburg (2.1%), Piceance (2.5%), N. San Juan (0.34%), and S. San Juan (1.13%) Basins [Rusco, 2010].

[18] The average leak rate we estimated from 3 February of $8.9 \pm 2.7\%$ is a factor of 1.8 greater than the GAO/WRAP bottom-up estimate (possibly more, as the GAO estimate of 5% included both flaring and venting; our measurements do not include CH₄ that is flared and converted to CO₂). Further measurements over several days and different

months and seasons would be necessary to evaluate the variability of emissions in Uintah County, because our result represents a snapshot of emissions from this region. Our result is consistent, however, with results from previous top-down studies of oil and gas production regions, which also found inventory estimates too low by similar factors [Katzenstein et al., 2003; Pétron et al., 2012]. More measurement-based evaluations of bottom-up inventories are needed to determine the consistency of results across different regions and determine trends in emissions that may result from increased production, new extraction techniques, or new regulations. Such independent verification of inventory-based estimates is essential for evaluating inventory methodologies, quantifying the effectiveness of future regulatory efforts, and accurately determining the climate impact of natural gas relative to other fossil fuels.

[19] **Acknowledgments.** This study would not have been possible without the support from participants of the 2012 Uintah Basin Winter Ozone and Air Quality Study, which was funded by Uintah Impact Mitigation Special Service District (UIMSSD), Western Energy Alliance, Bureau of Land Management (BLM), National Oceanic and Atmospheric Administration (NOAA), Environmental Protection Agency (EPA), National Science Foundation (NSF), and the State of Utah. We thank Ken Davis (Pennsylvania State University), Christopher Fairall (NOAA/PSD), Kelly Sours, Molly Crotwell, Jack Higgs, Don Neff, Doug Guenther, Carolina Siso, and Chris Carparelli (University of Colorado and NOAA/ESRL) for their assistance and contributions to this project.

[20] The Editor thanks Euan Nisbet and an anonymous reviewer for their assistance in evaluating this paper.

References

- Alvarez, R. A., S. W. Pacala, J. J. Winebrake, W. L. Chameides, and S. P. Hamburg (2012), Greater focus needed on methane leakage from natural gas infrastructure, *Proc. Natl. Acad. Sci. U. S. A.*, *109*(17), 6435–6440.
- Bar-Ilan, A., J. Grant, R. Parikh, R. Morris, K. Sgamma, T. Moore, and L. Gribovicz (2006), A comprehensive emissions inventory of upstream oil and gas activities in the Rocky Mountain States, ENVIRON International Corporation, Western Energy Alliance, Western Governors' Association Western Regional Air Partnership.
- Griffith, D. W. T., G. R. Bryant, D. Hsu, and A. R. Reisinger (2008), Methane emissions from free-ranging cattle: Comparison of tracer and integrated horizontal flux techniques, *J. Environ. Qual.*, *37*(2), 582–591.
- Harrison, M. R., T. M. Shires, J. K. Wessels, and R. M. Cowgill (1996), Methane emissions from the natural gas industry, Volume 1: Executive summary, Radian International for the Gas Research Institute, Washington, D. C and EPA National Risk Management Research Laboratory, Research Triangle Park, NC, U.S.
- Howarth, R. W., R. Santoro, and A. Ingraffea (2011), Methane and the greenhouse-gas footprint of natural gas from shale formations, *Clim. Change*, *106*(4), 679–690.
- Intergovernmental Panel on Climate Change (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge Univ. Press, New York.
- Katzenstein, A. S., L. A. Doezema, I. J. Simpson, D. R. Blake, and F. S. Rowland (2003), Extensive regional atmospheric hydrocarbon pollution in the southwestern United States, *Proc. Natl. Acad. Sci. U. S. A.*, *100*(21), 11,975–11,979.
- Klusman, R. W. (2003), Rate measurements and detection of gas microseepage to the atmosphere from an enhanced oil recovery/sequestration project, Rangely, Colorado, USA, *Appl. Geochem.*, *18*(12), 1825–1838.
- Mays, K. L., P. B. Shepson, B. H. Stirm, A. Karion, C. Sweeney, and K. R. Gurney (2009), Aircraft-based measurements of the carbon footprint of Indianapolis, *Environ. Sci. Technol.*, *43*(20), 7816–7823.
- Pétron, G., et al. (2012), Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study, *J. Geophys. Res.*, *117*, D04304, doi:10.1029/2011JD016360.
- Rusco, F. (2010), Federal oil and gas leases: Opportunities exist to capture vented and flared natural gas, which would increase royalty payments and reduce greenhouse gases, GAO, Washington, D. C.
- Ryerson, T. B., et al. (2001), Observations of ozone formation in power plant plumes and implications for ozone control strategies, *Science*, *292*(5517), 719–723.
- Shires, T., and M. Lev-On (2012), *Characterizing Pivotal Sources of Methane Emissions from Unconventional Natural Gas Production: Summary and Analysis of API and ANGA Survey Responses*, pp. 48, Am. Pet. Inst., Am. Natl. Gas Alliance, Washington, D. C.
- Turnbull, J. C., et al. (2011), Assessment of fossil fuel carbon dioxide and other anthropogenic trace gas emissions from airborne measurements over Sacramento, California in spring 2009, *Atmos. Chem. Phys.*, *11*(2), 705–721.
- U.S. Department of Agriculture (2009), 2007 Census of Agriculture, Utah, State and County Data, vol. 1, Natl. Agric. Stat. Serv, Washington, D. C.
- U.S. Department of Energy Energy Information Administration (1999), *Natural Gas 1998: Issues and Trends*, DOE/EIA.
- U.S. Environmental Protection Agency (EPA) (2010), *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2008*, EPA, Washington, D. C.
- U.S. Environmental Protection Agency (EPA) (2011), *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2009*, EPA, Washington, D. C.
- U.S. Environmental Protection Agency (EPA) (2012), *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2010*, EPA, Washington, D. C.
- U.S. Environmental Protection Agency (EPA) (2013), *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2011*, EPA, Washington, D. C.
- U.S. Environmental Protection Agency Office of Inspector General (2013), *EPA Needs to Improve Air Emissions Data for the Oil and Natural Gas Production Sector*, EPA OIG, Washington, D. C.
- White, W. H., J. A. Anderson, D. L. Blumenthal, R. B. Husar, N. V. Gillani, J. D. Husar, and W. E. Wilson (1976), Formation and transport of secondary air-pollutants: Ozone and aerosols in St. Louis urban plume, *Science*, *194*(4261), 187–189.