Title:

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

Keywords

Methane, Natural Gas, Unconventional gas, Greenhouse gas, Emissions, VOCs

Text:

New extraction technologies are making natural gas from shale and tight sand gas reservoirs in the United States (US) more accessible. As a result, the US has become the largest producer of natural gas in the world[1]. This growth in natural gas production may result in increased leakage of methane, a potent greenhouse gas[2], offsetting the climate benefits of natural gas relative to other fossil fuels. Methane emissions from natural gas production are not well quantified because of the large variety of potential sources, the variability in production and operating practices, the uneven distribution of emitters[3,4], and a lack of verification of emission inventories with direct atmospheric measurements. Here we determined gas emissions to be 8.8±2.6% (1σ) of natural gas production in the Uintah County, Utah (UT) natural gas field from atmospheric measurements made during aircraft research flights in February 2012. This emissions estimate is 1.8 to 38 times inventory-based estimates from this region[5] and five times the US EPA nationwide average estimate of leakage from the production and processing of natural gas[6]. Although the emissions for Uintah reported here may not be representative of other natural gas fields[5], this study demonstrates the importance of verifying emissions from natural gas production to enable an accurate assessment of its overall climate impact.

As concern grows over the climate impact of increasing greenhouse gas (GHG) emissions and expenditures for imported fuels, the US is looking to exploit natural gas as an attractive domestic energy source. Natural gas is an efficient energy source because its combustion produces more energy per carbon dioxide (CO_2) molecule formed than coal

or oil (177% and 140% respectively)[7]. Despite this efficiency, leakage of gas to the atmosphere from the point of extraction to the point of consumption reduces its climate (and economic) benefits. For example, previous research 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 GHG footprint than that from a coal-fired plant[3].

A critical gap in determining the climate impact of the recent increase in US natural gas production is an accurate and reliable estimate of fugitive gas emissions. In particular, the methodology used to account for methane (CH_4) emissions during production is in question. This is demonstrated by the recent change in natural gas-related CH_4 emissions reported by the US Environmental Protection Agency (EPA), which caused the estimated national average production-sector leak rate to jump from 0.17% to 1.44% of production[8,6]. This substantial increase was driven largely by a change in EPA's inventory methodology for calculating emissions from liquid unloading, unconventional completions, and work-overs of natural gas wells. The revised estimate has been questioned by the oil and gas industry, which contends that methane emissions from hydraulic fracturing of unconventional wells are half of the revised EPA estimate[9].

The ongoing debate between the EPA and the gas industry highlights an important point: most CH_4 emissions from oil and gas operations are estimated from the "bottom-up", a method 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[10] 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[5], and the uncertainty in the activity data and the emission factors is not well represented in the bottom-up estimates. From this perspective, there is an urgent need to assess the emission factors and extrapolation approaches used in bottom-up inventories with independent measurements and assessments of CH_4 emissions[11]. To date, a few studies have evaluated inventory estimates of oil and gas production basin emissions[12,13] with direct measurements of changes in the CH_4 mole fraction (moles CH_4 per mole of dry air). Although these studies have demonstrated the utility of direct atmospheric measurements, their emissions estimates rely on some factors that were not directly measured, such as transport of emissions or gas composition profiles.

During February 2012 we calculate the CH_4 flux from the Uintah County oil and gas field using a mass balance approach with direct measurements of CO_2, CH_4, water vapor (H_2O) and transport. The trace gas mole fractions were measured from a single-engine turboprop aircraft and planetary boundary layer (PBL) depth, wind speed, and wind direction were measured by High Resolution Doppler Lidar (HRDL) (Supplementary Methods 2.1 - 2.3). The mass balance approach offers a transparent and verifiable method, with quantifiable uncertainties, for estimating the total emission of a trace gas released from a defined point[14] or area source[15-17] (Methods Summary and Supplementary Methods 2.4). The Uintah County oil and gas field is well-suited to this approach for deriving CH_4 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 x 60 km^2, Fig. 1)[18]; an aircraft travelling at 60 m s^{-1} is able to make several transects and profiles over this field during a single flight. Twelve flights averaging four hours

each were made over the basin during the month-long campaign. On February 3, 2012, a well-defined boundary layer and steady winds led to ideal meteorological conditions for the mass balance calculation. On February 7, 2012, low and variable winds allowed for the confirmation of fluxes measured on February 3 but with much higher uncertainties.

Wind speeds on February 3, 2012 peaked in the early morning (9:00 GMT, 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 three hours before the downwind transect was flown (22:25 GMT, 15:25 LT). The PBL height (1700 m above ground level [magl]) was determined from three aircraft vertical profiles (Supplementary Fig. 1) and HRDL measurements. The rest of the flight measurements were made within the PBL between 100 and 1000 magl (Fig. 1).

The flight transect downwind of the gas field, along its southern and western edges and between 400 and 600 magl, showed elevated CH_4 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 (Fig. 2). Winds (averaged throughout the PBL) from HRDL measurements were used to construct a back trajectory of the air mass sampled in this plume (Fig. 1, red arrow). The trajectory indicates that the source of enhanced CH_4 is 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 a ~3 hr period prior to being sampled. We integrated the methane enhancement above the background value (derived from measurements made upwind of the location of oil and gas wells) along the flight path to calculate the flux from the oil and gas basin (Fig. 2 and Methods Summary). We are able to derive an appropriate uncertainty in each term in the mass

balance equation, producing a total uncertainty of 28% (1σ) on the total CH_4 flux derived on this day: $56\pm15 \times 10^3$ kg hr^{-1} (Supplementary Table 1 and Supplementary Methods 2.4).

On February 7, low winds (0.5 – 1.5 m s^{-1}) from the south and a shallower PBL led to large CH_4 enhancements above background (245 ppb on average) in the plume downwind of the same gas field (Supplementary Fig. 3a). The CH_4 flux calculated for this plume using the direct mass balance approach is $30\pm19 \times 10^3$ kg hr^{-1} with a high overall uncertainty of 62%, primarily from the variability in wind speed (Supplementary Table 2 and Supplementary Methods 2.5). We also implement a tracer ratio approach using CO_2 emitted by a nearby power plant as a tracer for air mixing and dilution. This approach uses data from two flight transects across a plume of CO_2 downwind of the Bonanza Creek Power Plant (Supplementary Fig. 3b), whose CO_2 emissions are monitored by a Continuous Emissions Monitoring System (CEMS) and reported hourly[19]. We use the ratio of the two plume integrals (Supplementary Fig. 4), along with the known power plant emissions, and estimate the CH_4 flux from the field to be $53\pm36 \times 10^3$ kg hr^{-1} and $66\pm50 \times 10^3$ kg hr^{-1} for the two transects (Supplementary Table 3 and Supplementary Methods 2.5).

The low uncertainty in the emission derived on the February 3, 2012, flight is the result of steady winds, consistent boundary layer height, and low measurement uncertainties. The derived emissions estimate from this day is our best estimate of emissions from the basin. Although the uncertainties for the flight on February 7, 2012 are high, this flight provides a second independent assessment of the CH_4 emissions and indicates that the result from February 3 was not anomalous (Table 1). While inconsistent meteorological conditions on the ten other flight days prevent direct mass balance analysis of CH_4

emissions, we note that CH_4 enhancements were large on all days and that the enhancements observed on February 3 and 7 were representative of those measured in the basin throughout the month of February (Supplementary Fig. 5).

Although no hydrocarbon measurements were made on the February 3, 2012 flight, analyses of 67 discrete air samples collected over Uintah County aboard the aircraft throughout the month of February 2012 show excellent correlations of propane and butane with CH_4 ($R^2 > 0.85$, Fig. 3). Correlations of CH_4 with carbon monoxide (CO), a tracer for vehicle exhaust, are weak ($R^2 = 0.28$, increasing to 0.52 when a single outlier with high CO is removed from the analysis). The strong correlations of CH_4 with propane and butane point to these CH_4 enhancements being primarily the result of emissions from oil and gas operations[13].

A flux of $2x10^3$ kg CH_4 hr^{-1} (< 4% of our best estimate of 56 kg CH_4 hr^{-1}) was subtracted from the total we derived to account for emissions from cattle and natural seepage, as estimated from inventories[20-22] (Supplementary Methods 2.6), to give a total CH_4 emission from natural gas that is 8.8±2.6% (1σ) of the average hourly February gas production in Uintah County, UT[23] (Table 1). Based on production data and publically available activity data, there is little evidence that emissions on either February 3 or 7 might be abnormal relative to other days in January, February or March 2012 (Supplementary Discussion 4.1, Supplementary Fig. 6 and 7)

Given the large greenhouse warming potential of CH_4, an 8.8% leak rate of natural gas during production negates any climate benefit of natural gas from this basin for electricity generation compared to coal and oil[3,4]. An inventory analysis by the US Government

Accountability Office (GAO) suggests, however, that the emissions from Uintah may be significantly higher than in other Western US basins. Using the Western Regional Air Partnership (WRAP) phase III[24] 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 higher (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[5].

While the WRAP III-based analysis concluded that 5% of production was lost to venting and flaring in the Uintah Basin, operators in this basin reported annual production losses of only 0.24% to the US Department of Interior Oil and Gas Operations Report (OGOR)[5]. Our independent measurement-based estimate of 8.8±2.6% is nearly twice the WRAP and almost 38 times the OGOR reported volumes (possibly more, as those include both flaring and venting). This discrepancy highlights the value of our study, which provides the first atmospheric measurement-based estimate of CH_4 emissions from a producing gas and oil field to date that does not rely on atmospheric transport models or bottom-up inventory information. 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 over other fossil fuels.

Methods Summary

Mass balance approach

In the mass balance approach for flux estimation, the mole fraction enhancement, relative to the upwind mole fraction, is integrated across the width of a well-defined plume in the PBL downwind of the source[16]. When the wind is 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 CH_4 flux is derived by:

$$flux_{CH_4} = V \int_{-b}^{b} \Delta X_{CH_4} \left(\int_{z_{ground}}^{z_{PBL}} n_{air} dz \right) cos\theta dx. \quad (1)$$

In equation (1), $flux_{CH4}$ represents the molar flux (moles s^{-1}) of CH_4 from the basin. V is the mean 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 prevalent wind direction. The CH_4 enhancement over the background mole fraction, ΔX_{CH4}, 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}, here assumed constant).

References:

1 EIA. *International Energy Statistics, Natural Gas*, <http://www.eia.gov/cfapps/ipdbproject/IEDIndex3.cfm?tid=3&pid=3&aid=1> (2012).

2 IPCC. 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 University Press and New York, NY, Cambridge, United Kingdom, 2007).

3 Alvarez, R. A., Pacala, S. W., Winebrake, J. J., Chameides, W. L. & Hamburg, S. P. Greater focus needed on methane leakage from natural gas infrastructure. *Proc. Natl. Acad. Sci. U. S. A.* **109**, 6435-6440, doi:10.1073/pnas.1202407109 (2012).

4 Howarth, R. W., Santoro, R. & Ingraffea, A. Methane and the greenhouse-gas footprint of natural gas from shale formations. *Climatic Change* **106**, 679-690, doi:10.1007/s10584-011-0061-5 (2011).

5 Rusco, F. FEDERAL OIL AND GAS LEASES: Opportunities Exist to Capture Vented and Flared Natural Gas, Which Would Increase Royalty Payments and Reduce Greenhouse Gases. Report No. GAO-11-34, (US Government Accountability Office (GAO), 2010).

6 US Environmental Protection Agency. INVENTORY OF U.S. GREENHOUSE GAS EMISSIONS AND SINKS: 1990 – 2009. (EPA, Washington, DC, 2011).

7 US Department of Energy Energy Information Administration. Natural Gas 1998 Issues and Trends. Report No. DOE/EIA-0560(98), (DOE/EIA, 1999).

8 US Environmental Protection Agency. INVENTORY OF U.S. GREENHOUSE GAS EMISSIONS AND SINKS: 1990 – 2008. (EPA, Washington, DC, 2010).

9 Shires, T. & Lev-On, M. Characterizing Pivotal Sources of Methane Emissions from Unconventional Natural Gas Production: Summary and Analysis of API and ANGA Survey Responses. 48 (American Petroleum Institute and America's Natural Gas Alliance, 2012).

10 Harrison, M. R., Shires, T. M., Wessels, J. K. & Cowgill, R. M. Methane Emissions from the Natural Gas Industry, Volume 1: Executive Summary. Report No. DCN: 96-263-081-17, (Radian International for the Gas Research Institute (GRI) and US EPA, 1996).

11 Frost, G. J. *et al.* New Directions: Toward a community emissions approach. *Atmos. Environ.* **51**, 333-334, doi:10.1016/j.atmosenv.2012.01.055 (2012).

12 Katzenstein, A. S., Doezema, L. A., Simpson, I. J., Blake, D. R. & Rowland, F. S. Extensive regional atmospheric hydrocarbon pollution in the southwestern United States. *Proc Natl Acad Sci U S A* **100**, 11975-11979, doi:10.1073/pnas.1635258100 (2003).

13 Petron, G. *et al.* Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. *J. Geophys. Res.-Atmos.* **117**, doi:10.1029/2011jd016360 (2012).

14 Ryerson, T. B. *et al.* Observations of ozone formation in power plant plumes and implications for ozone control strategies. *Science* **292**, 719-723, doi:10.1126/science.1058113 (2001).

15 Turnbull, J. C. *et al.* 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**, 705-721, doi:10.5194/acp-11-705-2011 (2011).
16 White, W. H. *et al.* FORMATION AND TRANSPORT OF SECONDARY AIR-POLLUTANTS - OZONE AND AEROSOLS IN ST-LOUIS URBAN PLUME. *Science* **194**, 187-189, doi:10.1126/science.959846 (1976).
17 Mays, K. L. *et al.* Aircraft-Based Measurements of the Carbon Footprint of Indianapolis. *Environ. Sci. Technol.* **43**, 7816-7823, doi:10.1021/es901326b (2009).
18 State of Utah Department of Natural Resources Division of Oil Gas and Mining. *Well Information Query*, <http://oilgas.ogm.utah.gov/Data_Center/LiveData_Search/well_information.htm> (2012).
19 US Environmental Protection Agency. *Air Markets Program Data, Bonanza Creek Power Plant Emissions*, <http://ampd.epa.gov/ampd/> (2012).
20 US Department of Agriculture. 2007 Census of Agriculture, Utah, State and County Data, Volume 1. Report No. AC-07-A-44, (National Agricultural Statistics Service, 2009).
21 Klusman, R. W. Rate measurements and detection of gas microseepage to the atmosphere from an enhanced oil recovery/sequestration project, Rangely, Colorado, USA. *Appl. Geochem.* **18**, 1825-1838, doi:10.1016/s0883-2927(03)00108-2 (2003).
22 Griffith, D. W. T., Bryant, G. R., Hsu, D. & Reisinger, A. R. Methane emissions from free-ranging cattle: Comparison of tracer and integrated horizontal flux techniques. *J. Environ. Qual.* **37**, 582-591, doi:10.2134/jeq2006.0426 (2008).
23 State of Utah Department of Natural Resources Division of Oil Gas and Mining. Summary of Production Report. (https://fs.ogm.utah.gov/pub/Oil&Gas/Publications/Reports/Prod/County/Cty_Feb_2012.pdf, 2012).
24 Bar-Ilan, A. *et al.* A Comprehensive Emissions Inventory of Upstream Oil and Gas Activities in the Rocky Mountain States. (ENVIRON International Corporation, Western Energy Alliance, and Western Governors' Association Western Regional Air Partnership (WRAP), 2006).
25 Bar-Ilan, A. *et al.* Development of 2012 Oil and Gas Emissions Projections for the Uinta Basin. (ENVIRON International Corporation, 2009).
26 Bar-Ilan, A. Personal Communication, (2012).

Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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) and 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.

Author Contributions:

C.S., G.P. and M.T. designed the study; C.S., T.N., S.W., J.K., S.C. and G.P. performed aircraft field work; P.L. and B.R.M. measured flask samples; M.H., R.B., A.B. and R.Z. performed and analyzed meteorological measurements; A.K., C.S., G.P. and M.T. analyzed data; A.K. and C.S. wrote the manuscript; G.P., P.T., S.M., G.J.F., R.S., E.D., and M.T. gave technical support and conceptual advice. A.K. and C.S. have contributed equally to this manuscript.

Author Information:

The authors declare that there are no competing interests as defined by Nature Publishing Group, or other interests that might be perceived to influence the results and/or discussion reported in this article. Reprints and permissions information is available at www.nature.com/reprints. Correspondence and requests for materials should be addressed to Dr. Colm Sweeney (colm.sweeney@noaa.gov).

Table:

Table 1. Summary of methane flux calculated from the aircraft flights on February 3 and February 7.

Flight day	**Method**	**CH_4 flux (10^3 kg hr^{-1})**	**% of average February production**
February 3	Mass balance	56±15*	8.8±2.6*
February 7	Mass balance	30±19	4.6±2.9
February 7 (Transect 1)	Ratio mass balance	53±36	8.3±5.7
February 7 (Transect 2)	Ratio mass balance	66±50	10.5±8.0
WRAP Inventory	Projected 2012 emissions	36**	5**

* Best estimate of CH_4 flux

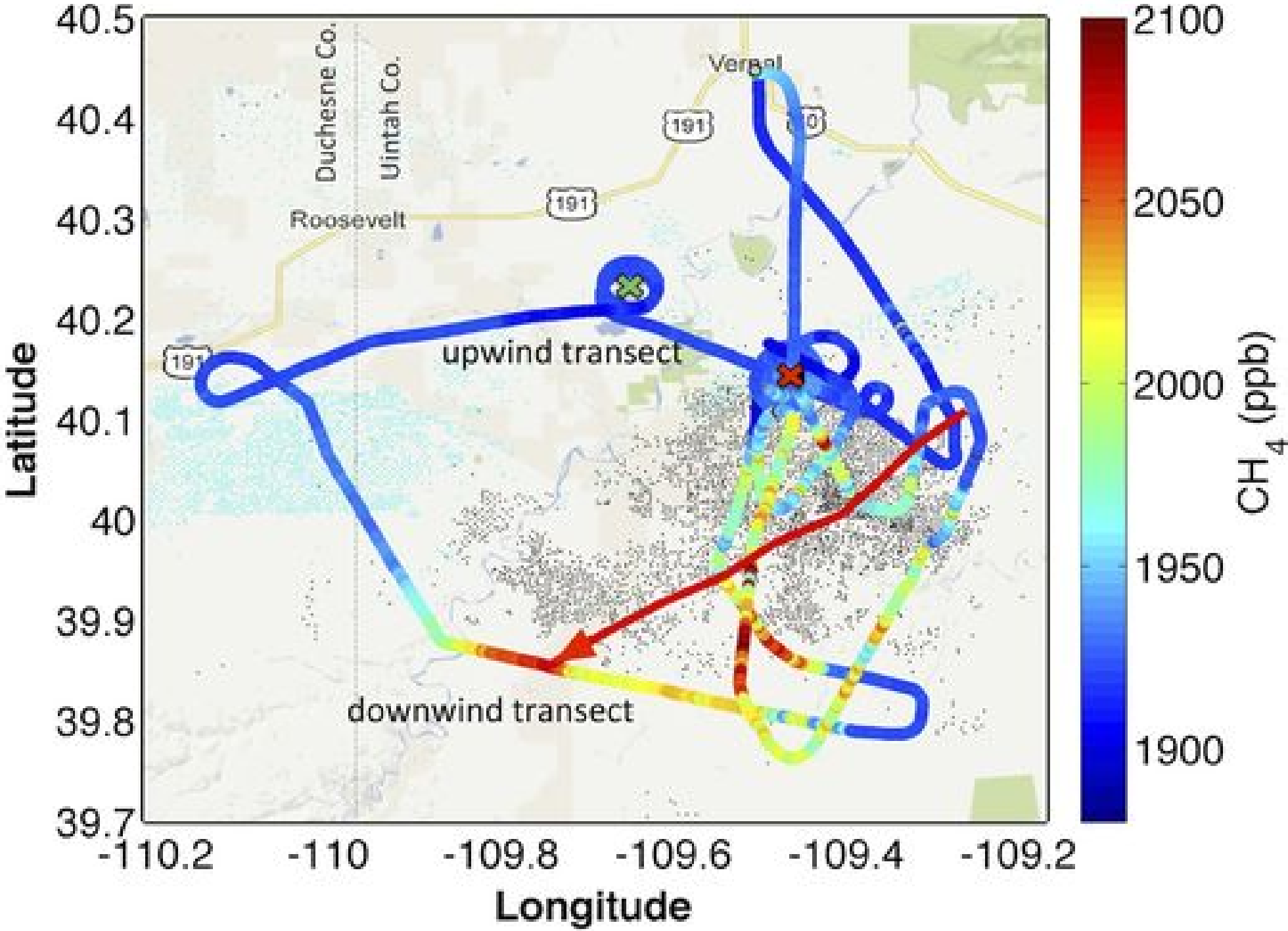
**Calculated using WRAP III-projected[25] VOC emissions for 2012 and the WRAP III vented conventional gas composition profile[26].

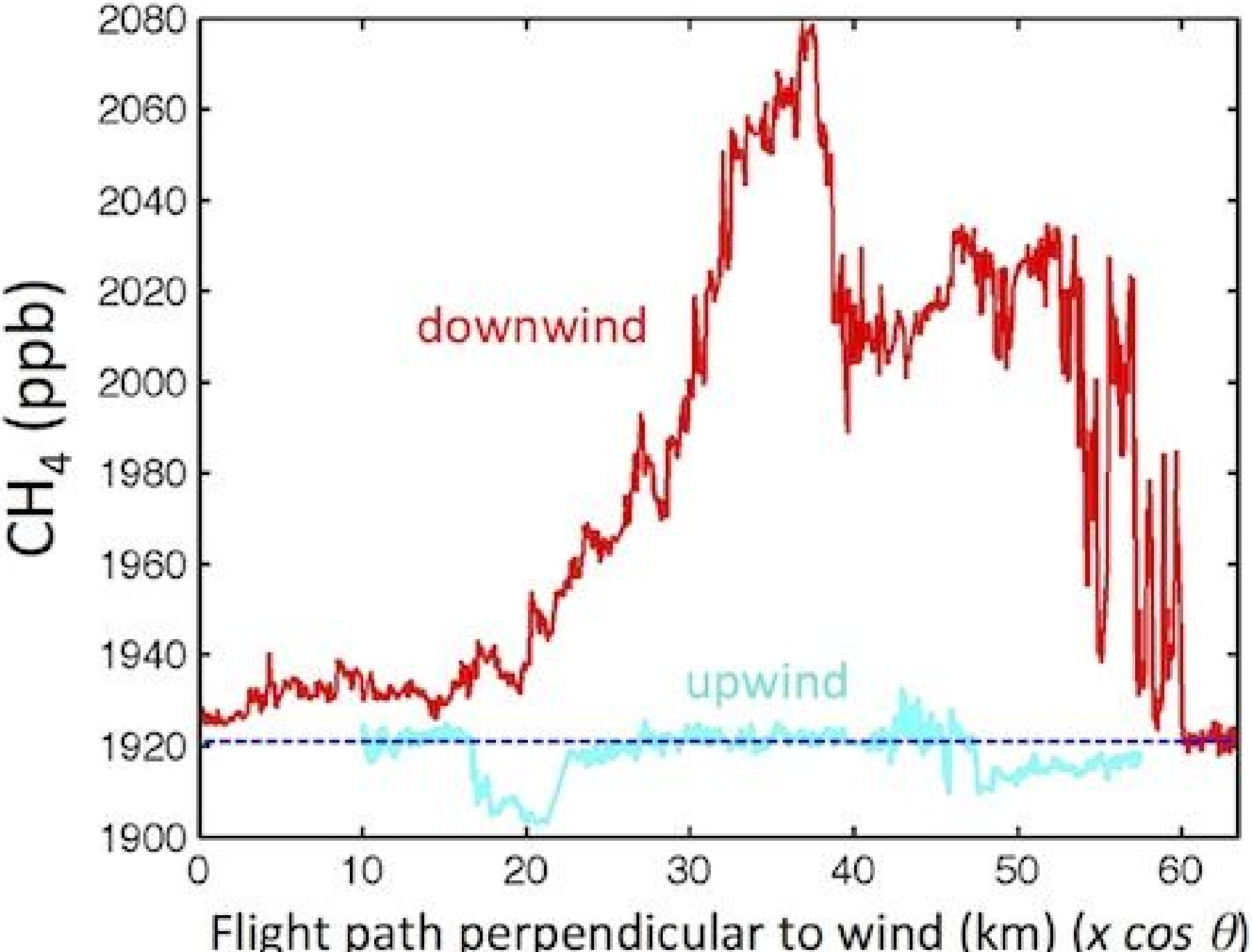
Figure Legends:

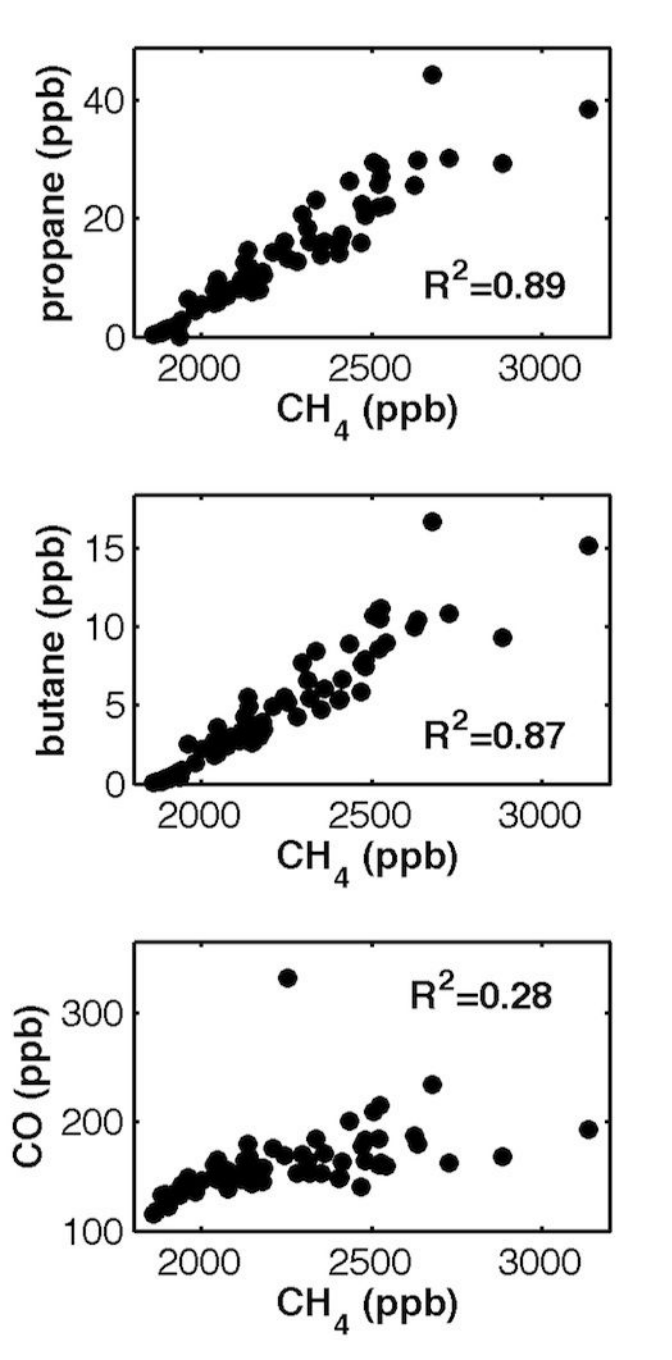
Figure 1. CH_4 plume measurements, February 3, 2012. Aircraft flight track overlaid on natural gas (black dots) and oil (blue dots) well locations along with color-coded CH_4 mole fraction. Bold red arrow shows the 3-hr trajectory of the downwind air mass. The locations of two vertical profiles over Horse Pool (red X) and one northwest of Horse Pool (green X) are also indicated.

Figure 2. CH_4 mole fraction measured in the downwind plume (red line) as a function of distance perpendicular to the wind direction. The CH_4 mole fraction in the upwind transect is in light blue, and its average (1921 ppb) is represented by the dark blue dashed line.

Figure 3. Mole fractions of trace gases measured in discrete air samples collected over the Uintah Basin in February 2012, shown as functions of CH_4 mole fraction. Correlation coefficients (R^2) are shown in each figure.


40.5
40.4
40.3
40.2
40.1
40
39.9
39.8
39.7
Latitude
-110.2
-110
-109.8
-109.6
-109.4
-109.2
Longitude
Duchesne Co.
Uintah Co.
Roosevelt
Vernal
191
191
191
upwind transect
downwind transect
2100
2050
2000
1950
1900
CH4 (ppb)


2080
2060
2040
2020
2000
1980
1960
1940
1920
1900
CH_4 (ppb)
downwind
upwind
0
10
20
30
40
50
60
Flight path perpendicular to wind (km) ($x \cos \theta$)


propane (ppb)
40
20
0
2000
2500
3000
CH4 (ppb)
R2=0.89
butane (ppb)
15
10
5
0
2000
2500
3000
CH4 (ppb)
R2=0.87
CO (ppb)
300
200
100
2000
2500
3000
CH4 (ppb)
R2=0.28