Environmental Science & Technology

Article

pubs.acs.org/est

Airborne Quantification of Methane Emissions over the Four Corners Region

Mackenzie L. Smith,[†] Alexander Gvakharia,[†] Eric A. Kort,[*,†] Colm Sweeney,[‡,§] Stephen A. Conley,[∥,⊥] Ian Faloona,[⊥] Tim Newberger,[‡,§] Russell Schnell,[§] Stefan Schwietzke,[‡,§] and Sonja Wolter[‡,§]

[†]Climate and Space Sciences and Engineering, University of Michigan, Ann Arbor, Michigan 48109, United States

[‡]Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado 80309, United States

[§]NOAA Earth System Research Laboratory, Boulder, Colorado 80305, United States

[∥]Scientific Aviation, Boulder, Colorado 80301, United States

[⊥]Department of Land, Air, & Water Resources, University of California Davis, Davis, California 95616, United States

Supporting Information

ABSTRACT: Methane (CH_4) is a potent greenhouse gas and the primary component of natural gas. The San Juan Basin (SJB) is one of the largest coal-bed methane producing regions in North America and, including gas production from conventional and shale sources, contributed ~2% of U.S. natural gas production in 2015. In this work, we quantify the CH_4 flux from the SJB using continuous atmospheric sampling from aircraft collected during the TOPDOWN2015 field campaign in April 2015. Using five independent days of measurements and the aircraft-based mass balance method, we calculate an average CH_4 flux of 0.54 ± 0.20 Tg yr^{-1} (1σ), in close agreement with the previous space-based estimate made for 2003−2009. These results agree within error with the U.S. EPA gridded inventory for 2012. These flights combined with the previous satellite study suggest CH_4 emissions have not changed. While there have been significant declines in natural gas production between measurements, recent increases in oil production in the SJB may explain why emission of CH_4 has not declined. Airborne quantification of outcrops where seepage occurs are consistent with ground-based studies that indicate these geological sources are a small fraction of the basin total (0.02−0.12 Tg yr^{-1}) and cannot explain basinwide consistent emissions from 2003 to 2015.

1. INTRODUCTION

The San Juan Basin (SJB) in New Mexico and Colorado is a major natural gas production basin and the largest coal-bed methane producing region in North America as of 2015.[1−3] An elevated CH_4 anomaly over the SJB was detected from space from 2003 to 2009. These observations were validated with ground-based measurements and, in combination with an atmospheric transport model, used to determine a CH_4 flux from the SJB of 0.59 Tg yr^{-1} [0.50−0.67 2σ].[4] At the time, this estimate exceeded inventory estimates for emissions from the region [EPA Greenhouse Gas Reporting Program (GHGRP) estimate for 2012 was 0.33 Tg yr^{-1}; we note this inventory estimate for 2012 has since changed, which is described below]. Since the 2003−2009 time period, gas production in the San Juan Basin declined to reach a level in 2015 that is 34% below average production from 2003 to 2009, while oil production has increased rapidly (~260%).[5] Subsequent analyses with satellite data have estimated fluxes from the region consistent with the original study.[6,7] The TOPDOWN2015 campaign in the SJB region was designed to follow up on the satellite finding, determine total CH_4 flux from the basin, assess the role of geologic seepage, and better understand the distribution of contributing sources. Given the large change in production of oil and gas in this region, assessing how total emissions may have changed also provides key insights into response to changing production volumes and practices.

Airborne studies quantifying the total basinwide CH_4 flux from the SJB have not yet been published; however, the role of point sources contributing to the total flux has been investigated (understanding the distribution of contributing sources). During the TOPDOWN2015 campaign, Frankenberg et al.[8] used aircraft-mounted infrared spectrometers to identify and quantify fluxes from individual point sources of CH_4 in the region. They found that the largest 10% of sources contributed disproportionately (49−66%) to the sum of observed CH_4 point source fluxes of 0.23−0.38 Tg yr^{-1}. This estimate includes only the identified large point sources sampled by the remote sensing aircraft but

Received: December 2, 2016
Revised: April 14, 2017
Accepted: April 18, 2017
Published: April 18, 2017

DOI: 10.1021/acs.est.6b06107
Environ. Sci. Technol. 2017, 51, 5832−5837

aids in understanding the origins of the total flux as well as apportionment of CH_4 to anthropogenic versus natural sources. The majority of sources observed by Frankenberg et al. were anthropogenic, largely related to gas and oil production sectors, as well as a coal mine venting shaft and geologic seepage.

In this work, we use airborne observations taken during the TOPDOWN2015 campaign to determine a quantified estimate of total CH_4 flux from the San Juan basin using the mass-balance methodology similarly to previous applications to other regions of oil and gas production.[9−14] We also determine the CH_4 flux from several large point sources using in situ airborne measurements,[15] including regions of geologic seepage. Finally, we compare our quantified emissions with estimates from inventories and discuss the implications of emissions observed in 2015 compared with previous findings for 2003−2009.

2. METHODS

2.1. Instrumentation and Aircraft. Airborne observations over the SJB of southern Colorado and northern New Mexico were collected onboard Twin Otter (NOAA) and Mooney (Scientific Aviation) aircraft during the TOPDOWN2015 field campaign in April 2015. Both aircraft were equipped with a cavity ring-down spectrometer (Picarro G2401-m; ∼0.5−1 Hz sampling frequency) that quantified atmospheric CH_4 and H_2O mole fractions. Inflight calibrations were used to determine the reported CH_4 dry mole fraction on the WMO scale (X2004 scale maintained by NOAA GMD). Precision and accuracy of the measurement was 0.5 ppb. Further details regarding previous operation and validation of Picarro spectrometers on both airborne platforms can be found in Karion et al. (2013),[16] Karion et al. (2015),[9] and Peischl et al. (2016).[17] Wind speed and direction, ambient temperature and pressure, relative humidity, and GPS location were measured on both aircraft as described in Karion et al. (2015).[9] Additionally, a wind profiler (915-MHz boundary layer wind profiler)[18] was deployed in the southwest region of the study area (Farmington, NM) that recorded wind speed and direction with altitude continuously during the study period.

Flights were designed to determine the total CH_4 flux from the SJB, as well as the flux from individual sources. The studied area is bounded approximately by 36.0° to 37.5° latitude and −108.5° to −107° longitude, which encompasses the majority of gas-producing wells in the region. This region also coincides with the location of the elevated CH_4 anomaly observed by Kort et al.[4] derived from space-based SCIAMACHY retrievals from 2003 to 2009.

2.2. Basinwide Flux. The CH_4 enhancement downwind of the SJB study area was measured on 5 days when the wind speed and direction were stable for 5−7 h prior to the downwind flight transect (wind speed variance was on the order of 1−2 m s^{-1}, direction variance ranged from 20° to 60°). Wind speeds and directions were considered stable if wind back trajectories constructed using data from the wind profiler located in the southwest of the study area (Farmington, NM) indicated that the air intercepted by a downwind flight transect originated outside of the study area (section S1 and Figure S1, no data for April 7, 2015). Wind vectors for each hour were constructed using hourly vertical profiles of wind speed and direction measured by the NOAA wind profiler in Farmington, NM (Figure S2) in high-resolution mode (mode 1), which were averaged vertically from 250 magl to a PBL height that varied with time of day. The trajectories were run back in time from the time and location of a downwind transect (using the midpoint of the transect). These combined constraints ensured that emissions contributing to the enhancement were from the current day rather than accumulation in the basin, either overnight or trapped by swirling winds and topography (cf. section S1). Multiple flights were conducted from April 7 to 30; measurements from five flights conducted between ∼15:30 to 18:00 local time were determined to be suitable for mass balance analysis. This assessment and confirmation is of particular importance in the SJB where the topography of the region often leads to air pooling in the SJB overnight. Downwind flight transects were conducted within the well-mixed boundary layer (determined using aircraft vertical profile measurements; see below) at altitudes of ∼630 to 1650 magl.

The molar flux is calculated using the mass balance method, which has been applied previously to calculation of regional fluxes,[9,13,14,16,17] according to eq 1:

$$\text{flux}_{CH_4} = \nu \cos\theta \int_{-b}^{b} X_{CH_4}\, dx \int_{z_{ground}}^{z_1} n_{air}\, dz \quad (1)$$

where $-b$ to b is the width of the enhancement plume, X_{CH_4} the molar CH_4 enhancement above background mixing ratios, n_{air} the molar density of air, z_{ground} to z_1 the height of the planetary boundary layer (PBL), and $\nu \cos(\theta)$ the component of horizontal wind perpendicular to the flight path.

Methane background mole fractions are defined by taking the mean of the CH_4 mixing ratios at the edges of the enhancement plumes (Figure S3). The well-mixed boundary layer height (z_1) is calculated from vertical gradients in CH_4 and H_2O, which decrease sharply with increasing altitude as they transition from well-mixed layer to free tropospheric mixing ratios (further details provided in section S2 and Figure S4).[17] We note that the mixing layer heights determined from the vertical profiles agree with the PBLs determined from the wind profiler (data not shown). Wind speeds and directions used for the flux calculation are derived from differential GPS measurements onboard the aircraft[15] and averaged during the time a downwind transect was conducted. The average values determined from the aircraft-based wind measurements agree with the wind speeds and directions determined using the ground-based wind profiler data within error (Table S1). Uncertainties are calculated and propagated as is standard in the approach and described in Kort et al.[14] and Peischl et al.[17]

2.3. Individual Source Flux. The CH_4 flux from 18 point sources, including gas processing plants (3), compressor stations (6), a reinjection facility, a power plant, a coal mine vent shaft, and a geological seep in the SJB, were investigated by circling individual sources with the Mooney aircraft, using the method of Conley et al.[15] Point sources sampled by aircraft were chosen based on known large point sources in the EPA GHGRP, as well as randomly sampling sources when a large CH_4 signal was observed in flight. After a plume from a point source was detected, measurements were taken at multiple altitudes spaced approximately 50 m apart, starting as close to ground level as possible (typically a few hundred meters above ground level) and ending at an altitude where the plume was no longer observed.

This method enables quantification of emissions from defined point sources provided the source is persistently emitting during the time of aircraft circling, which typically takes ∼20 min. Quantification of fluxes is possible with this approach, but the significant time needed to sample an individual site limits the number of point sources that can be assessed in this manner.

DOI: 10.1021/acs.est.6b06107
Environ. Sci. Technol. 2017, 51, 5832−5837

3. METHANE FLUX FROM THE SAN JUAN BASIN

Observations collected on 5 days during the larger TOP-DOWN2015 campaign in April 2015 are used to determine an average CH_4 flux from the San Juan Basin during the study period of 62 ± 27 kg hr^{-1} (1σ). Flight paths colored by CH_4 mixing ratio for the 5 days used in our analysis are shown in Figure 1. Extrapolating our 5 day average to a yearly flux, we find 0.54 ± 0.20 Tg yr^{-1} CH_4 are emitted. The range of observed fluxes (0.31−0.84 Tg yr^{-1}; Table 1) underscores the importance of multiday realizations of emissions to determine a central estimate. One contributing reason for the range of observations is that each flight sampled slightly different source regions (e.g., some geological outcrops do extend beyond the boundaries, which may bias some estimates low). The observed flux from the SJB is of the same order of magnitude as other studied fossil fuel production basins, with CH_4 fluxes from the Bakken, Uintah, Barnett, Marcellus, Denver-Julesberg, Haynesville, and Eagle Ford basins ranging from 0.12 to 0.70 Tg yr^{-1} across all basins.[9−11,16,17] Note that these estimates are based on measurements made in afternoon hours spanning at most a month, whereas the satellite-derived estimates are made from retrievals collected in all seasons over multiple years. We also calculated daily CH_4 fluxes using the wind speeds and directions measured by the wind profiler (Table S1) for four of the five flights (profiler data not available for April 7, 2015) to assess the influence of different wind estimates (i.e., aircraft winds averaged over a downwind transect compared to winds determined from profiler data). The 4 day mean CH_4 flux of 0.51 Tg yr^{-1} based on the profiler winds agrees within error with the mean CH_4 flux calculated using the aircraft-based wind data (0.54 ± 0.20).

4. BOTTOM-UP EMISSIONS ESTIMATES

We can investigate how this emission value compares with bottom-up inventories. The US EPA GHG gridded methane inventory for anthropogenic sources is recently available for the year 2012.[19] Summing all sources for the San Juan basin totals 0.40 Tg yr^{-1}, where >80% of the emission is attributed to natural gas production and processing, with oil and coal representing most of the remaining emissions. Only ∼3−4% of the total is from enteric fermentations and landfills. A very small seasonality exists in the inventory for this region, but if we consider only April, extrapolated to a yearly flux, 0.40 Tg yr^{-1} remains the estimate. This estimate includes all source categories in the basin with the exception of geologic seepage. In the Colorado portion of the basin, ongoing measurements of geologic seepage have been recorded, where large interannual variance is observed, with annual fluxes not showing a trend in recent years but fluctuating from 0.04 Tg yr^{-1} in 2007 to 0.02 Tg yr^{-1} in 2011 and 0.12 Tg yr^{-1} in 2015.[20] If we combine these bottom-up estimates, considering 0.40 Tg yr^{-1} from the EPA inventory, a range of 0.02−0.12 for geologic seeps (representing the upper and lower bound from ground-based observations), we find an estimated bottom-up value ranging from 0.42 to 0.52 Tg yr^{-1}, though we note there are no robust uncertainties associated with these bottom-up estimates. Our top-down total basin estimated flux, an annual extrapolation based on 5 flight days, is within uncertainty of the bottom up estimates, and the bottom-up estimate is dominated by gas production and processing emissions.

5. POINT SOURCES

To further investigate the magnitude of emissions from individual point sources contributing to total CH_4 emissions in the basin, the Mooney aircraft circled oil, gas, coal, and geologic seeps at different altitudes while measuring atmospheric CH_4 mixing ratios as described in Conley et al.[15] The largest point source observed by the Mooney was a coal mine vent shaft (also

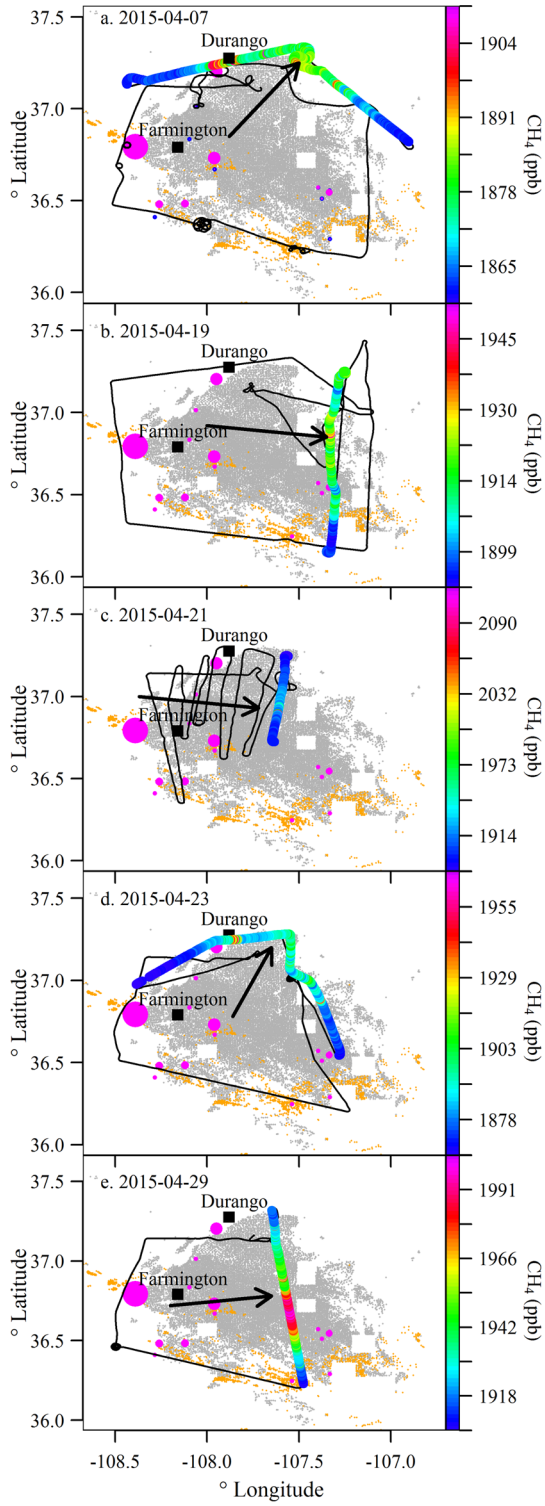

Figure 1. Mass balance flight transects. Color scale shows observed CH_4 mixing ratio along a downwind flight transect. Arrows show approximate wind direction. Also shown are gas wells (gray); oil wells (orange); point sources circled by the Mooney aircraft, scaled by relative measured CH_4 flux (magenta); and the locations of Durango, CO and Farmington, NM (black squares).

DOI: 10.1021/acs.est.6b06107
Environ. Sci. Technol. 2017, 51, 5832−5837

Table 1. Summary of Inputs to eq 1 with 1σ Uncertainties and CH_4 Fluxes Calculated from Observations Onboard the Mooney Aircraft (Top Row) and the NOAA Twin Otter (Bottom Rows)[a]

date	local hr (−6 UTC hr)	no. of transects	Θ (deg)	v (m s^{-1})	z_1 (magl)	$flux_{CH_4}$ (Tg yr^{-1})
Mooney						
4/07/2015	15.5	1	42 ± 10	10 ± 2	2138 ± 71	0.45 ± 0.15
Otter						
4/19/2015	16.2	1	93 ± 24	8.1 ± 2.6	2250 ± 124	0.57 ± 0.25
4/21/2015	16.2−17.2	4	95 ± 22	6.8 ± 1.9	2263 ± 106	0.31 ± 0.13
4/23/2015	15.8	1	45 ± 20	7.0 ± 1.8	2450 ± 257	0.55 ± 0.19
4/29/2015	17.0	1	83 ± 25	5.8 ± 1.6	2150 ± 347	0.84 ± 0.30
				Campaign Mean:		**0.54 ± 0.20**[b]

[a]Flux values are daily means. Wind speeds and directions are determined using the mean values derived from aircraft data during each transect. [b]Error on daily flux estimates are determined by summing the error of the components of eq 1 in quadrature; error on campaign flux mean is the 1σ of the mean.

observed by Frankenberg et al.,[8] Table 2), which comprised 2.4% of total basin emissions (5 day average of 1446 kg hr^{-1} CH_4).

Table 2. Examples of Some of the Largest CH_4 Point Sources Quantified by Aircraft Measurements (Top Rows) and Remote Infrared Sensing (Bottom Rows), as Well as the Sum of All Sources Observed but Not Listed[a]

reported point source	$flux_{CH_4}$ (Tg yr^{-1})	% total basin $flux_{CH_4}$
this work (Mooney)		
one geological seep	0.0062	1.2
coal mine vent shaft	0.013	2.4
Σ observed sources (n = 18)	**0.047**	**8.7**
Frankenberg et al.[8] (2016)		
coal mine vent shaft	0.014	2.6
Σ observed sources ($n \geq 200$)	**0.23−0.38**	**43−72**

[a]Estimates of the coal mine vent shaft from this work and Frankenberg et al.[8] are expected to agree because data from the Mooney aircraft was used to validate the method of Frankenberg et al.

Additionally, the Mooney aircraft measured CH_4 geological seepage from a coal outcrop to investigate its magnitude; a large outcrop southwest of Durango was investigated on 2 separate days, and a flux of 709 ± 325 kg hr^{-1} was determined, which is ∼1% of the total basinwide emission (Table 2). Thus, even the largest sources make up only a few percent of overall emissions, in contrast to the even more outsized role of large point sources reported in the Barnett Shale region.[21] Although the sources measured by Frankenberg et al. follow a heavy-tail distribution,[8] small emissions distributed across the field contribute substantively to the overall CH_4 flux.

6. TIME SERIES OF EMISSIONS AND GAS PRODUCTION

Kort et al. estimated no trend in CH_4 flux in the SJB during 2003−2009 (0.59 Tg yr^{-1} [0.50−0.67; 2σ]).[4] The present study estimates an average CH_4 flux in 2015 of 0.54 Tg yr^{-1} [0.34−0.74; 1σ], which is consistent with the average for 2003−2009, even though gas production decreased in the SJB between the years 2003 to 2015. According to San Juan county gas production data, production in the region for 2015 decreased from the 2003−2009 average by 34% (time series of production shown in Figure 2). Different measurements and models are used in the studies of Kort et al. and our current study on emissions from the SJB, but if we consider the reported uncertainties as robust, using a t test for significance at a 95% confidence level, we find that we

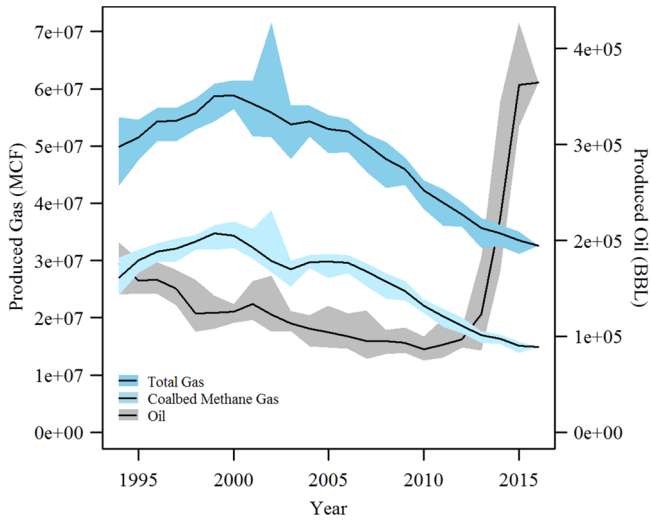

Figure 2. Gas and oil production in San Juan county from 1994 to 2016. Shading shows range of monthly reported values, and line shows the annual average. Production data from the New Mexico Energy, Minerals and Natural Resources Department, Oil Conservation Department.[5]

are able to statistically quantify if a decrease in emissions of 26% or more occurred between the earlier study of Kort et al. and 2015 (section S3). Thus, we would be able to detect a decrease in basinwide emissions if they were tightly correlated with gas production (which decreased by 34%) and there was no other major source of emissions. A common assumption is that the leak rate of natural gas scales with gas production,[11] though this has been shown to not be true on all scales, such as when considering individual gas production pads.[22] Excluding other potential sources of CH_4 emissions our results indicate that in the SJB gas production is a poor predictor for total oil and gas CH_4 emissions over time (multiyear scales) as gas can be, and is, lost in ways not related to total gas production.

Considering other potential sources that might explain why emissions do not appear to have changed over the past decade, one hypothesis is that emissions from geological seepage have increased over time. However, the seepage determined from the Mooney aircraft measurement of one outcrop area where we were able to observe elevated methane signals indicated this source contributed ∼1% to total CH_4 emission. Other outcrop areas we monitored (in CO and NM) did not present as discernible signals in the aircraft data. Given the magnitude from aircraft and ground-based sampling, and the lack of any large

DOI: 10.1021/acs.est.6b06107
Environ. Sci. Technol. 2017, 51, 5832−5837

increasing trend in ground-based sampling, geologic seepage cannot explain the persistent emissions in the basin over time.

A second hypothesis to explain the persistent emissions despite the decrease in natural gas production is the sharp increase from 2013 to 2015 (~260%) in oil production. Fossil fuel production regions with higher oil and liquids production are often associated with higher fugitive emissions. Lyon et al.[23] observed that oil-producing well-pads were more likely to be associated with observable CH_4 emissions than predominately gas-producing well-pads. It is possible then that in the San Juan basin as natural gas production decreased and methane emissions associated with those activities decreased, this fugitive loss was counterbalanced by increased oil production and fugitive emissions from these processes. To confirm or deny this hypothesis, more in depth analysis is needed, including fine-scale information on emissions associated with each process over time, but it does provide a possible explanation for the persistent emissions in the region in the face of declining natural gas production.

7. IMPLICATIONS

Combining the airborne results from 2015 with previous work reporting average emissions from 2003 to 2009,[4] we find that the flux of CH_4 from the San Juan Basin has not changed in a statistically detectable manner for over a decade, despite a 34% decrease in gas production from the 2003−2009 average to 2015. Recent work with space-based observations has also suggested persistent and stable emissions for the Four Corners region over this time period.[7] As geologic outcrops and other anthropogenic sources (ruminants and landfills) are not large enough sources nor do they exhibit the necessary trend to explain this phenomenon, the implication is that basinwide emissions of CH_4 do not scale with total basin gas production and that fugitive losses are not dominated by pathways that correlate with gas production volume. Further work should continue to investigate how changes (or lack thereof) in basinwide flux estimates over time are related to underlying factors, including type of fuel produced.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b06107.

Text describing selection of downwind flight transects for mass balance analysis and figures showing wind back-trajectories for four mass balance days; vertical profiles of wind speed, average wind speeds and directions, and CH_4 flux estimates using winds measured by the wind profiler; example of CH_4 enhancement plume; calculation of atmospheric mixing depths and vertical profiles of H_2O used to determine mixing depth height; t test for significant differences between satellite and airborne CH_4 flux estimates (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: eakort@umich.edu. Phone: (734) 763-8414. Fax: (734) 764-0437. Address: 2553 Space Research Building, 2455 Hayward St., Ann Arbor, MI 48109-2143.

ORCID

Alexander Gvakharia: 0000-0003-1260-4744

Stephen A. Conley: 0000-0001-6753-8962

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by the NOAA AC4 program under Grant NA14OAR0110139 and NASA Grant NNX14AI87G. Portions of this work were supported by funding from the Bureau of Land Management, Grant L15PG00058; the Climate Program Office, National Oceanic and Atmospheric Administration (NOAA); the Directors Office, Earth System Research Laboratory, NOAA; and a NOAA grant to the Cooperative Institute for Marine Ecosystems & Climate (#201503225). Access to facilities and reservation lands by the Southern Ute Indian Tribe, Ignacio, CO and wind profiler installations and data, Physical Science Division, NOAA, Boulder, CO are also acknowledged. We thank Anna Karion and the NOAA Aircraft Operations Center and Scientific Aviation staff and flight crew for their efforts in helping collect these data. We thank the broader community interested in methane emissions in Four Corners for their support and input.

■ REFERENCES

(1) *Coalbed Methane Production*. https://www.eia.gov/dnav/ng/ng_prod_coalbed_s1_a.htm, U.S. Energy Information Administration, Washington, DC, 2015.

(2) *County Production and Injection by Month*. https://wwwapps.emnrd.state.nm.us/ocd/ocdpermitting//Reporting/Production/CountyProductionInjectionSummary.aspx. New Mexico Energy Minerals and Natural Resources Department, Santa Fe, NM, 2015.

(3) *Monthly Coalbed Methane Produced by County*. http://cogcc.state.co.us/COGCCReports/production.aspx?id=MonthlyCoalGasProdByCounty. Colorado Oil & Gas Conservation Commission, Denver, CO, 2015.

(4) Kort, E. A.; Frankenberg, C.; Costigan, K. R.; Lindenmaier, R.; Dubey, M. K.; Wunch, D. Four corners: The largest US methane anomaly viewed from space. *Geophys. Res. Lett.* **2014**, *41* (19), 6898−6903.

(5) *County Production and Injection by Month*. https://wwwapps.emnrd.state.nm.us/ocd/ocdpermitting//Reporting/Production/CountyProductionInjectionSummary.aspx. New Mexico Energy Minerals and Natural Resources Department, Santa Fe, NM, 1994−2015.

(6) Turner, A. J.; Jacob, D. J.; Benmergui, J.; Wofsy, S. C.; Maasakkers, J. D.; Butz, A.; Hasekamp, O.; Biraud, S. C. A large increase in U.S. methane emissions over the past decade inferred from satellite data and surface observations. *Geophys. Res. Lett.* **2016**, *43* (5), 2218−2224.

(7) Buchwitz, M.; Schneising, O.; Reuter, M.; Heymann, J.; Krautwurst, S.; Bovensmann, H.; Burrows, J. P.; Boesch, H.; Parker, R. J.; Detmers, R. G.; et al. Satellite-derived methane hotspot emission estimates using a fast data-driven method. *Atmos. Chem. Phys. Discuss.* **2016**, in review; DOI: 10.5194/acp-2016-755.

(8) Frankenberg, C.; Thorpe, A. K.; Thompson, D. R.; Hulley, G.; Kort, E. A.; Vance, N.; Borchardt, J.; Krings, T.; Gerilowski, K.; Sweeney, C.; et al. Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners region. *Proc. Natl. Acad. Sci. U. S. A.* **2016**, *113* (35), 9734−9739.

(9) Karion, A.; Sweeney, C.; Kort, E. A.; Shepson, P. B.; Brewer, A.; Cambaliza, M.; Conley, S. A.; Davis, K.; Deng, A.; Hardesty, M.; et al. Aircraft-based estimate of total methane emissions from the Barnett Shale region. *Environ. Sci. Technol.* **2015**, *49* (13), 8124−8131.

(10) Pétron, G.; Karion, A.; Sweeney, C.; Miller, B. R.; Montzka, S. A.; Frost, G. J.; Trainer, M.; Tans, P.; Andrews, A.; Kofler, J.; et al. A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin. *J. Geophys. Res.: Atmos.* **2014**, *119* (11), 6836−6852.

(11) Peischl, J.; Ryerson, T. B.; Aikin, K. C.; de Gouw, J. A.; Gilman, J. B.; Holloway, J. S.; Lerner, B. M.; Nadkarni, R.; Neuman, J. A.; Nowak, J. B.; et al. Quantifying atmospheric methane emissions from the

Haynesville, Fayetteville, and northeastern Marcellus shale gas production regions. *J. Geophys. Res.: Atmos.* **2015**, *120* (5), 2119−2139.
(12) Schwarz, J. P.; Holloway, J. S.; Katich, J. M.; McKeen, S.; Kort, E. A.; Smith, M. L.; Ryerson, T. B.; Sweeney, C.; Peischl, J. Black carbon emissions from the Bakken oil and gas development region. *Environ. Sci. Technol. Lett.* **2015**, *2* (10), 281−285.
(13) Smith, M. L.; Kort, E. A.; Karion, A.; Sweeney, C.; Herndon, S. C.; Yacovitch, T. I. Airborne ethane observations in the Barnett Shale: Quantification of ethane flux and attribution of methane emissions. *Environ. Sci. Technol.* **2015**, *49* (13), 8158−8166.
(14) Kort, E. A.; Smith, M. L.; Murray, L. T.; Gvakharia, A.; Brandt, A. R.; Peischl, J.; Ryerson, T. B.; Sweeney, C.; Travis, K. Fugitive emissions from the Bakken shale illustrate role of shale production in global ethane shift. *Geophys. Res. Lett.* **2016**, *43* (9), 4617−4623.
(15) Conley, S.; Faloona, I.; Mehrotra, S.; Suard, M.; Lenschow, D. H.; Sweeney, C.; Herndon, S.; Schwietzke, S.; Petron, G.; Pifer, J.; Kort, E. A.; Schnell, R. Application of Gauss' Theorem to quantify localized surface emissions from airborne measurements of wind and trace gases. *Atmos. Meas. Tech.* **2017**, in review; DOI: 10.5194/amt-2017-55.
(16) Karion, A.; Sweeney, C.; Petron, G.; Frost, G.; Hardesty, R. M.; Kofler, J.; Miller, B. R.; Newberger, T.; Wolter, S.; Banta, R.; et al. Methane emissions estimate from airborne measurements over a western United States natural gas field. *Geophys. Res. Lett.* **2013**, *40* (16), 4393−4397.
(17) Peischl, J.; Karion, A.; Sweeney, C.; Kort, E. A.; Smith, M. L.; Brandt, A. R.; Yeskoo, T.; Aikin, K. C.; Conley, S. A.; Gvakharia, A.; et al. Quantifying atmospheric methane emissions from oil and natural gas production in the Bakken shale region of North Dakota. *J. Geophys. Res.: Atmos.* **2016**, *121* (10), 6101−6111.
(18) Carter, D. A.; Gage, K. S.; Ecklund, W. L.; Angevine, W. M.; Johnston, P. E.; Riddle, A. C.; Wilson, J.; Williams, C. R. Developments in UHF lower tropospheric wind profiling at NOAA's Aeronomy Laboratory. *Radio Sci.* **1995**, *30* (4), 977−1001.
(19) Maasakkers, J. D.; Jacob, D. J.; Sulprizio, M. P.; Turner, A. J.; Weitz, M.; Wirth, T.; Hight, C.; DeFigueiredo, M.; Desai, M.; Schmeltz, R.; et al. Gridded National Inventory of U.S. Methane Emissions. *Environ. Sci. Technol.* **2016**, *50* (23), 13123−13133.
(20) *2015 Fruitland Outcrop Monitoring Report, La Plata County, Colorado, September 2015*. http://cogcc.state.co.us/documents/library/AreaReports/SanJuanBasin/3m_project/2015%20FRUITLAND%20OUTCROP%20MONITORING%20REPORT_La_Plata.pdf, LT Environmental, Inc., Durango, CO, 2015.
(21) Zavala-Araiza, D.; Lyon, D. R.; Alvarez, R. A.; Davis, K. J.; Harriss, R.; Herndon, S. C.; Karion, A.; Kort, E. A.; Lamb, B. K.; Lan, X.; et al. Reconciling divergent estimates of oil and gas methane emissions. *Proc. Natl. Acad. Sci. U. S. A.* **2015**, *112* (51), 15597−15602.
(22) Brantley, H. L.; Thoma, E. D.; Squier, W. C.; Guven, B. B.; Lyon, D. Assessment of methane emissions from oil and gas production pads using mobile measurements. *Environ. Sci. Technol.* **2014**, *48* (24), 14508−14515.
(23) Lyon, D. R.; Alvarez, R. A.; Zavala-Araiza, D.; Brandt, A. R.; Jackson, R. B.; Hamburg, S. P. Aerial surveys of elevated hydrocarbon emissions from oil and gas production sites. *Environ. Sci. Technol.* **2016**, *50* (9), 4877−4886.

DOI: 10.1021/acs.est.6b06107
Environ. Sci. Technol. 2017, 51, 5832−5837