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Key Points:
- Four Corners exhibits largest CH_4 anomaly seen from space
- Emissions of >0.5 Tg CH_4/yr have persisted since 2003
- Space- and ground-based CH_4 identify missing emissions from fossil fuel extraction

Supporting Information:
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Four corners: The largest US methane anomaly viewed from space

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Abstract Methane (CH_4) is a potent greenhouse gas and ozone precursor. Quantifying methane emissions is critical for projecting and mitigating changes to climate and air quality. Here we present CH_4 observations made from space combined with Earth-based remote sensing column measurements. Results indicate the largest anomalous CH_4 levels viewable from space over the conterminous U.S. are located at the Four Corners region in the Southwest U.S. Emissions exceeding inventory estimates, totaling 0.59 Tg CH_4/yr [0.50–0.67; 2σ], are necessary to bring high-resolution simulations and observations into agreement. This underestimated source approaches 10% of the EPA estimate of total U.S. CH_4 emissions from natural gas. The persistence of this CH_4 signal from 2003 onward indicates that the source is likely from established gas, coal, and coalbed methane mining and processing. This work demonstrates that space-based observations can identify anomalous CH_4 emission source regions and quantify their emissions with the use of a transport model.

1. Introduction

Understanding the global CH_4 budget has proven particularly elusive in recent years. Following a rapid decrease of the atmospheric burden's growth rate [*Dlugokencky et al.*, 1994] an apparent approach to steady state occurred in the late 1990's to early 2000's, with significant interannual variations attributed to sources including wetlands or fires [*Ringeval et al.*, 2010; *Wang et al.*, 2004]. The atmospheric burden then began growing again in 2007 [*Rigby et al.*, 2008; *Dlugokencky et al.*, 2009; *Nisbet et al.*, 2014]. Multiple studies have attempted to discover the cause for this renewed growth—determining whether this is an Arctic signal [*O'Connor et al.*, 2010; *Kort et al.*, 2012], renewed tropical wetland emissions [*Bousquet et al.*, 2011], attributable to increased anthropogenic emissions [*Bergamaschi et al.*, 2013], or some combination of these is critical for future climate projections and potential mitigation actions.

The explosive growth of unconventional gas recovery by high-volume hydraulic fracturing (fracking) has transformed the natural gas industry, vastly increasing accessible reserves in the U.S. Estimates of CH_4 emissions associated with this new extraction technique vary widely [*Howarth et al.*, 2011; *Cathles et al.*, 2012; *Howarth et al.*, 2012; *Levi*, 2012], with significant implications on the climate impact of hydraulic fracturing. Atmospheric studies over North America, considering overall emissions from basin to continental scale, have systematically pointed to underestimates through inventories [*Kort et al.*, 2008, 2010; *Hsu et al.*, 2010; *Petron et al.*, 2012; *Wennberg et al.*, 2012; *Wunch et al.*, 2009; *Brandt et al.*, 2014]. Other studies [*Katzenstein et al.*, 2003; *Miller et al.*, 2013] have used ground and airborne observations focused on the central U.S. to highlight that oil and gas activities in this region likely are underrepresented. A European study has demonstrated remote sensing from aircraft can quantify CH_4 emissions from coalbeds [*Krings et al.*, 2013]. Here we ask if space-based observations of atmospheric CH_4 can provide top-down constraints to identify sources and quantitatively assess this atmosphere-inventory discrepancy. We analyze regional-scale atmospheric CH_4 observations from space- and Earth-based instruments. With this multiinstrument remote sensor suite we discover a regional signature of large CH_4 emissions not seen in prior studies. We quantitatively use the observed CH_4 enhancement to demonstrate that emissions associated with established fossil fuel extraction activities (not associated with recent high-volume hydraulic fracturing activities) are significantly underestimated over large scales.

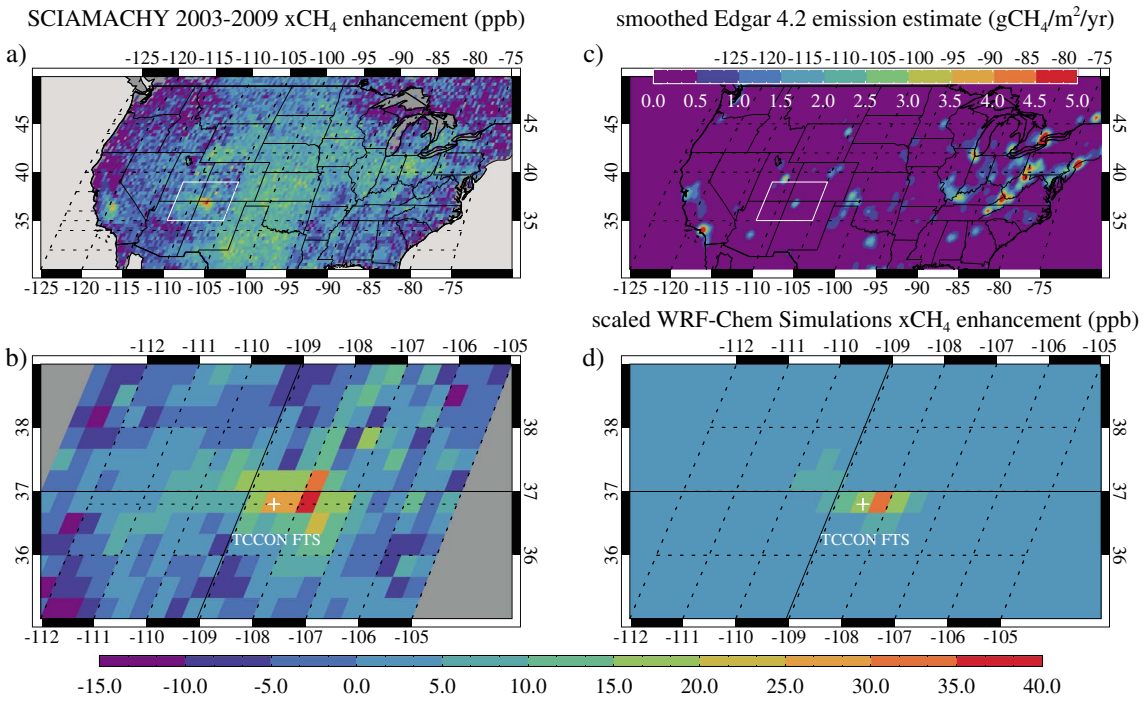

Figure 1. Column methane anomalies and emissions over the conterminous U.S. (a) Average SCIAMACHY anomaly from 2003 to 2009 gridded at 1/3° resolution. (b) Average SCIAMACHY anomaly over just the Four Corners region from 2003 to 2009. (c) EDGAR v4.2 gridded methane emissions (smoothed with a Gaussian filter). (d) Gridded WRF-Chem simulated methane anomaly using 3.5 times EDGAR v4.2 emissions for the Four Corners region.

2. Space-Based Observations

First, we consider space-based observations of column-averaged CH_4 mole fractions retrieved from spectra collected by the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) instrument from 2003 to 2009 [*Frankenberg et al.*, 2011]. Focusing on North America, averaging for the entire time period, and removing topographic impacts on the retrieval (see methods, Figure S1, and Text S1 in the supporting information discussion), we produced a map of CH_4 anomalies (Figure 1a, enhancement over topographical average). The largest local enhancement over this time frame is located over the Four Corners region of the U.S. (where Arizona, New Mexico, Colorado, and Utah all meet, Figure 1b). Other regions show elevated CH_4 levels as well: notably the Texas/Oklahoma region and central California. These elevated levels are likely associated with anthropogenic emissions from oil, gas, ruminants, and agriculture, as noted in recent focused studies on these regions [*Katzenstein et al.*, 2003; *Kort et al.*, 2008; *Miller et al.*, 2013]. Methane signals in these regions are weaker (~50% of Four Corners) and less persistent (not present in all seasons and years) than those observed at Four Corners; thus, it is more difficult to reliably constrain source strength and perform attribution. The Four Corners region shows the strongest column anomaly in the conterminous U.S. (Figures 1a and 1b). The substantive enhancement persists robustly through all seasons and years (Figure S2), within the 2003–2009 time period. Even though there is increased variability in the column enhancement after 2006 (possibly linked to SCIAMACHY signal degradation), there is no statistically significant change when comparing the enhancement from 2003 to 2005 and 2007 to 2009 (Figures S3 and S4). The larger interannual variability observed post-2006 may be indicative of annual emissions fluctuations in Four Corners, but the more robust multiyear comparison shows no trend, and degraded retrieval quality in SCIAMACHY data after 2005 warrants some caution in interpreting the enhanced variability. A strong source has persisted at Four Corners from 2003 through 2009 in all seasons (noting uncertainty on the source 2007–2009 is much greater than 2003–2005, owing to detector degradation).

3. Inventory Emissions

A substantive source of CH_4 for the Four Corners region is included in bottom-up inventory estimates but has not been validated. For the region exhibiting the large CH_4 anomaly, within −109.6°W to −107.0°W and 36.2°N

to 37.4°N, EDGAR v4.2 [*European Commission, Joint Research Centre/Netherlands Environmental Assessment Agency*, 2010] estimates 0.168 Tg CH_4 in 2008, primarily attributed to natural gas production, processing, and distribution. EDGAR emissions in this region change little both spatially and in magnitude from year to year. There are extensive gas, oil, coal, and coalbed CH_4 harvesting activities in the region southeast of Four Corners, centered on the San Juan Basin in New Mexico, along with associated gas processing and compressing facilities. This basin has been particularly important as a source of coalbed CH_4 production. The San Juan Basin has been the largest coalbed CH_4 production site in the U.S., with cumulative production in the 1990's exceeding all other U.S. coalbed CH_4 production combined [*Moore*, 2012]. High-volume hydraulic fracturing did begin in the basin circa 2009, but coalbed CH_4 remains the dominant natural gas source (natural gas produced from shale in 2012 remained at less than 1% that produced by coalbed methane in western NM). Operators in Four Corners report higher emissions than any other basin in the new U.S. Environmental Protection Agency (EPA) greenhouse gas reporting program (GHGRP) subpart W [*U.S. Environmental Protection Agency*, 2013]. GHGRP reported methane emissions (including any producers whose total emissions exceed 25,000 MMT CO_2 equivalent) for the San Juan basin in 2012 total 0.33 Tg CH_4/yr. EDGAR, which is a global product, thus underpredicts emissions in the San Juan basin relative to reported emissions (0.168 versus 0.33 Tg). In our analysis, we use EDGAR and its spatial distribution, but also note findings relative to the basin specific estimate from GHGRP.

4. High-Resolution Simulations

To determine what emissions rate is consistent with our atmospheric observations, we performed high-resolution regional simulations with the Weather Research and Forecasting Chemical transport model (WRF-Chem; see methods) [*Grell et al.*, 2005]. WRF-Chem simulations were performed for six separate 6 day periods (totaling 36 simulated days) encompassing January–February, March–April, May–June, July–August, September–October, and November–December of 2012. These time frames were chosen to cover the full range of seasonal dynamics, facilitating an even-handed annual comparison with SCIAMACHY observations. Simulations were performed in 2012 to match ground-based observations (which began in 2011) enabling both rigorous assessment of representativeness and direct simulation comparison with ground-based observations. Comparison of simulated days in 2012 with data from the ground-based Total Carbon Column Observing Network (TCCON) observations from the same days and for all of 2012 (Figure 3, Text S1 in the supporting information discussion on representativeness) demonstrates representativeness is robust. We performed forward model simulations of expected CH_4 enhancements (the hourly averaged value above the afternoon minimum) from the inventoried sources in EDGAR v4.2. Comparisons of the WRF-Chem simulations with SCIAMACHY observations (Figures 1d and 2) show that the model captures the observed spatial structure, and a significant linear relationship exists between model and observations ($r = 0.43$). This comparison is done with data averaged on $0.5 \times 0.5°$ grid, considering bimonthly average (i.e., January/February) for SCIAMACHY data from 2003 to 2009 and averaged WRF values for the corresponding simulation time (i.e., January/February) in 2012, as illustrated in Figure S2. Spatially shifting the simulations relative to observations demonstrates optimal correlation occurs with minimal offsetting (Figure S5), indicating that source location and transport in simulations are representative of the observations. The combination of these findings thus supports quantitative comparisons of the average of WRF simulations from 2012 with the average SCIAMACHY observations from 2003 to 2009. We calculate the slope accounting for uncertainty in both the ordinate and abscissa (Text S1 in the supporting information discussion) and find a slope of 3.5 ± 0.5 (2σ). This slope represents a scaling factor: simulated emissions need to be 3.5 times greater than in the prior model to agree with the SCIAMACHY observations (Figure 1d).

This is a substantive discrepancy with the prior inventory, and implies the Four Corners region alone emitted on average 0.59 Tg CH_4/yr (0.50–0.67 Tg, 2σ; includes all sources) from 2003 to 2009. This value is 1.8 times greater than GHGRP reported emissions for the region, and 3.5 times greater than EDGAR v4.2. The most recent U.S. EPA inventory released in 2013 estimates total U.S. emissions associated with natural gas as 7.7 Tg CH_4/yr (for 2008, *U.S. Environmental Protection Agency* [2013]). If the U.S. EPA inventory is accurate, this suggests that the Four Corners region is responsible for the equivalent of almost 10% of U.S. CH_4 emissions from natural gas systems (Figure 2b). In this case, compared to estimated emissions from natural gas systems, coal mining, and petroleum systems combined, the Four Corners region alone represents almost 5% of total U.S. emissions for those sectors (Figure 2b).

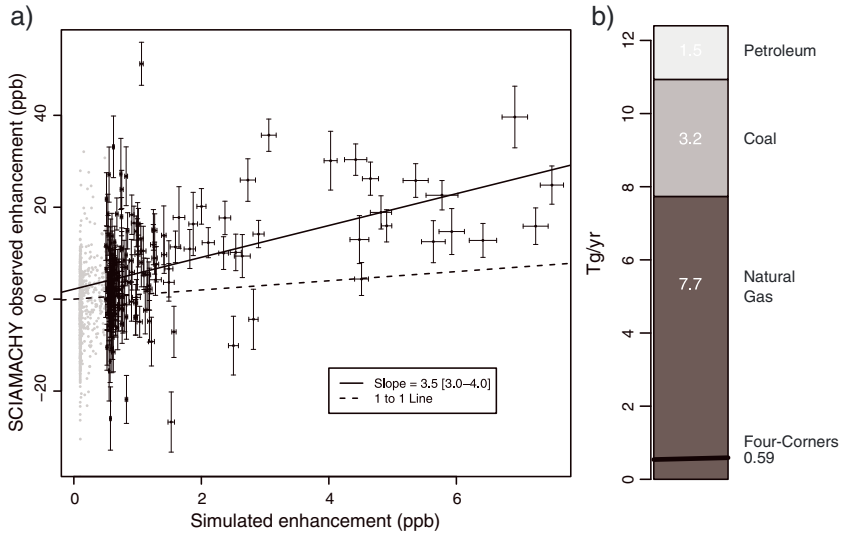

Figure 2. Methane enhancement for Four Corners region and U.S. EPA inventory estimates. (a) Observed (SCIAMACHY 2003–2009 average) and simulated (WRF-Chem) methane over Four Corners, with slope (calculated from 163 points) shown in solid line. Gray data points were excluded from the fit. (b) *U.S. Environmental Protection Agency*, 2013 inventory (for 2008) estimate of methane emissions associated with petroleum systems, coal mining, and natural gas systems, with our finding for the Four Corners region noted, representing nearly 10% of total U.S. methane emissions from natural gas systems according to this inventory.

5. Ground-Based Validation

This space-based finding is substantiated with independent ground-based observations at Four Corners performed at much higher temporal frequency than SCIAMACHY that capture the diurnal variations in column methane. As part of the Total Carbon Column Observing Network (TCCON) [*Wunch et al*. [2011]; *Lindenmaier et al*., 2014], an upward looking Fourier transform spectrometer was located near Four Corners (Figures 1b and 1d) between 1 March 2011 and 30 September 2013. This site is within the CH_4 anomaly observed by SCIAMACHY and predicted by WRF-Chem simulations. The TCCON site collected daytime observations under sunny conditions of column-averaged dry air CH_4 molar fraction tied to the World Meteorological Organization scale [*Wunch et al*., 2010].

A distinct diurnal cycle is measured in the total column observations (Figure 3, high solar zenith angle observations removed, plotting only 11 A.M. to 5 P.M. local). The column value peaks at 16–17 UTC (11 A.M. to noon local time), when winds are originating from the East/Southeast sector. More than half of the observing days show a diel cycle with column enhancements of at least 20 ppb (defined as A.M. peak to P.M. minimum, equivalent to daily maxima-minima), with 20% of days observing enhancements of 50 ppb or greater (Figure S7). The mean diurnal column enhancement for both the simulated days, and all days with observations in 2012, exceed 30 ppb (Figure 3). Occasional outlier days exhibited enhancements exceeding 100 ppb. The TCCON data are consistent with observations made from SCIAMACHY, independently confirming the presence of a strong CH_4 source that produces

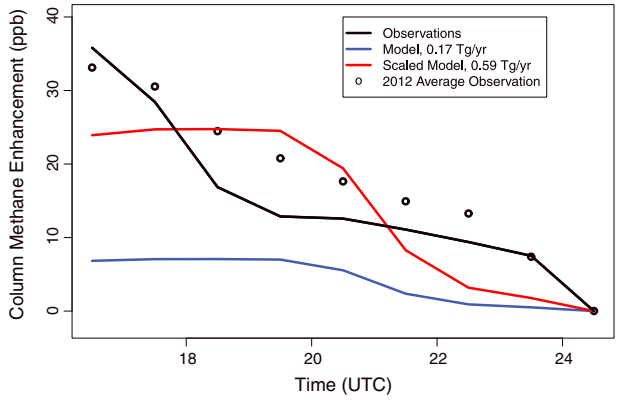

Figure 3. Column Methane Enhancement at Four Corners TCCON site. Observed (black line), simulated with EDGAR v4.2 (blue line), and simulated with EDGAR v4.2 scaled by 3.5 (red line) for the corresponding days of simulations. Black open circles indicate observed data points considering all 2012 data.

substantial column enhancements. In the TCCON data, this is evident in the diurnal cycle. The signal observed at the TCCON site is more sensitive to local winds that are more challenging to accurately simulate than the averaged SCIAMACHY observations. The WRF model simulations show fidelity in capturing the diurnal structure, exhibiting similar morning to afternoon dynamics. This suggests the simulations, when considered in aggregate, are capturing the regional dynamics and distribution of emission sources reasonably well. However, the magnitude of emissions is too low. In fact, the application of the scaling factor found from independent SCIAMACHY observations (3.5) brings the simulated diurnal cycle into agreement on days with corresponding observations in 2012 (Figure 3). This finding independently substantiates and strengthens the conclusion that Four Corners CH_4 emissions are ~0.59 Tg CH_4/yr, confirming the satellite finding and suggesting that enhanced emissions persisted through 2012.

6. Conclusion

Based on our finding of a large and consistent regional atmospheric CH_4 signal from 2003 onward, we conclude that long-established fossil fuel extraction, at least in the Four Corners region, likely has larger CH_4 emissions, and subsequent greenhouse gas footprint, than accounted for in current inventories. Other studies [*Katzenstein et al*., 2003; *Miller et al*., 2013] have highlighted that oil and gas activities likely are underrepresented but have primarily focused on oil and gas activities in the south central U.S. and could not resolve localized features such as Four Corners. This study pioneers the use of space-borne CH_4 retrievals to identify and quantify localized emissions from fossil fuel activities. Our study also shows that observational gaps can lead to large biases in inverse CH_4 estimates, since our increased Four Corners estimate is in disagreement with the results of inverse modeling that predicted lower emissions without data in this region [*Miller et al*., 2013]. We consider more than seven years of observations of oil/gas/coal activities, demonstrating a consistent emissions pattern over almost a decade (including a prefracking time frame) using a method not susceptible to bias caused by short time or limited area sampling. Our findings underscore the need to develop accurate baselines for fugitive CH_4 emissions from established gas activities (particularly, coalbed CH_4 extraction) to assess both changes from growing unconventional shale gas mining and short-term radiative forcing mitigation strategies through reductions in CH_4 emissions. The likely imminent growth of hydraulic fracturing in the Four Corners region makes our results timely for establishing a baseline and comparative analysis of fugitive leaks from new extraction technologies. Furthermore, we have demonstrated that space-based observations of CH_4 can be used to identify regions of anomalously large CH_4 emissions, quantitatively inform emission rates, and guide ground-based follow-up studies. If future space-based CH_4 observations are made with greater spatiotemporal coverage, it is likely more regions of anomalous emissions could be identified and have their respective emissions quantified from space.

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