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Constructing a Spatially Resolved Methane Emission Inventory for the Barnett Shale Region

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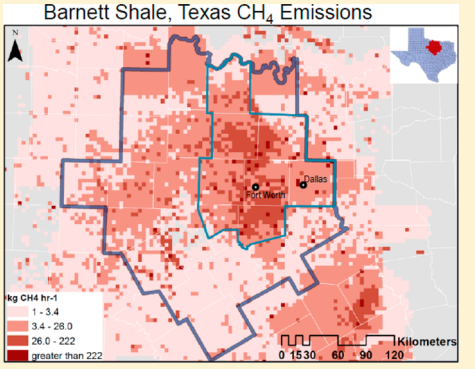
Supporting Information

ABSTRACT: Methane emissions from the oil and gas industry (O&G) and other sources in the Barnett Shale region were estimated by constructing a spatially resolved emission inventory. Eighteen source categories were estimated using multiple data sets, including new empirical measurements at regional O&G sites and a national study of gathering and processing facilities. Spatially referenced activity data were compiled from federal and state databases and combined with O&G facility emission factors calculated using Monte Carlo simulations that account for high emission sites representing the very upper portion, or fat-tail, in the observed emissions distributions. Total methane emissions in the 25-county Barnett Shale region in October 2013 were estimated to be 72,300 (63,400−82,400) kg CH_4 h^{-1}. O&G emissions were estimated to be 46,200 (40,000−54,100) kg CH_4 h^{-1} with 19% of emissions from fat-tail sites representing less than 2% of sites. Our estimate of O&G emissions in the Barnett Shale region was higher than alternative inventories based on the United States Environmental Protection Agency (EPA) Greenhouse Gas Inventory, EPA Greenhouse Gas Reporting Program, and Emissions Database for Global Atmospheric Research by factors of 1.5, 2.7, and 4.3, respectively. Gathering compressor stations, which accounted for 40% of O&G emissions in our inventory, had the largest difference from emission estimates based on EPA data sources. Our inventory's higher O&G emission estimate was due primarily to its more comprehensive activity factors and inclusion of emissions from fat-tail sites.



INTRODUCTION

Fossil fuel substitutions resulting from the recent growth of natural gas production have the potential to immediately reduce CO_2 emissions and long-term climate impacts, but emissions of methane from the natural gas supply chain may also increase short-term climate impacts.[1] Several recent studies have used different methodologies to estimate the magnitude of oil and gas industry (O&G) methane emissions.[2−6] Top-down approaches, which quantify emissions from a region using atmospheric measurements of well mixed air, have inferred higher O&G methane emissions than bottom-up approaches, which estimate regional emissions by constructing inventories based on activity factors and emission factors.[7,8] Reported differences may result in part from top-down studies incorrectly attributing emissions to O&G sources or sampling during times when short-term events are occurring at a different rate than predicted by inventories. Additionally, bottom-up studies may underestimate emissions due to incomplete activity factors or emission factors based on measurements that exclude the fat-tail of a skewed emission rate distribution — relatively rare sources that contribute a large fraction of total emissions. Coordinated top-down and bottom-up measurements are needed to reconcile the two methods and more accurately estimate methane emissions.[8,9] Development of a detailed emission inventory composed of both more complete activity factors and more representative emission factors is a critical step in top-down/bottom-up reconciliation.

The Barnett Shale of north-central Texas was the first shale basin to be developed for natural gas with a combination of

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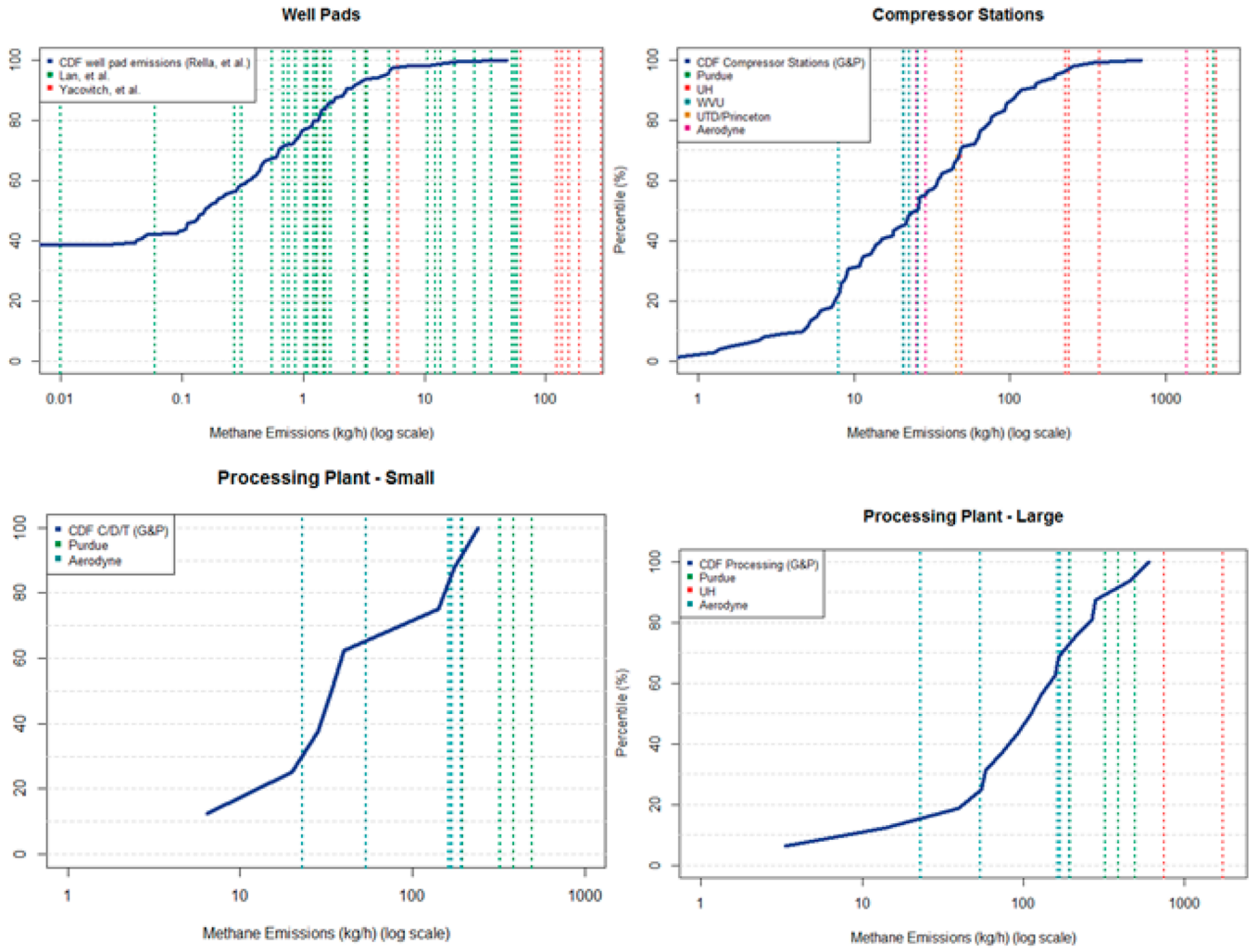

Figure 1. Sampled emission rate distributions by O&G sector with superimposed values of Barnett Coordinated campaign measurements. The blue lines are the cumulative distribution functions of sampled distributions used in Monte Carlo simulations, which include production site measurements made using unbiased sampling during the campaign[28] and a national data set of gathering stations and processing plants.[31] The vertical lines are emission rates of sites measured during the Barnett Campaign using sampling biased toward high emission sites.[27,29,30] The values exceeding the maximum of the sampled distributions are used as fat-tail site distributions in the Monte Carlo simulations.

horizontal drilling and hydraulic fracturing. Development peaked in 2008 with over 4,000 drilling permits issued and then declined to less than 1,000 issued permits 2013.[10] Barnett Shale production peaked at 5.7 billion cubic feet (Bcf) natural gas day^{-1} in 2012 and 28,000 barrels (Mbbl) day^{-1} hydrocarbon liquids (oil and natural gas condensate) in 2011,[10] but the basin is expected to remain a major contributor to U.S. natural gas production through 2030.[11] A mature field is ideal for investigating long-term methane emissions from O&G sites in the production phase but provides fewer opportunities to observe emission events from well development activities, which can be challenging to characterize due to their short duration and spatiotemporal heterogeneity.[12]

The Barnett Shale region contains most of the Dallas−Fort Worth−Arlington Metropolitan Statistical Area, which has a population over 6.5 million and includes many urban methane sources such as landfills. The region also includes extensive rural land use and over 1 million cattle. O&G air pollution sources in the region have been extensively studied; for example, a criteria and hazardous air pollutant emission inventory was developed by the Texas Commission on Environmental Quality (TCEQ),[13] and field measurements of pollutant emissions, including methane, were commissioned by the City of Fort Worth.[14]

During the two week period of October 16—30, 2013, ten research teams performed multiscale measurements in the Barnett Shale region to quantify methane emissions from O&G and other sources including landfills (the Barnett Coordinated Campaign). This paper uses bottom-up measurements from the Barnett Coordinated Campaign and other available data to construct a spatially resolved methane emission inventory (4 km × 4 km grid cells) for the 25-county Barnett Shale region defined by the Texas Railroad Commission.[10] Natural gas production site emission estimates were characterized in Zavala-Araiza et al.[15] Our bottom-up emission estimates were compared to alternative emission inventories we developed from commonly cited sources: the United States Environmental Protection Agency (EPA) Greenhouse Gas Reporting Program (GHGRP),[16] EPA United States Greenhouse Gas Inventory (GHGI),[17] and the Emissions Database for Global Atmospheric Research v4.2 (EDGAR).[18] These inventories have been shown to produce lower methane emissions rates than top-down studies regionally and nationally.[4,7,8,19] The top-down estimates made as part of the Barnett Coordinated Campaign and an

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additional two week period in March 2013 included the core production area of the Barnett Shale but did not include all of the 25-county area of this inventory.[20,21] The gridded inventory constructed for this paper can be used to estimate emissions in other spatial domains in the Barnett region including areas measured by top-down methods.

METHODS AND DATA

A spatially resolved methane emissions inventory for the 25-county Barnett Shale region was constructed using a combination of bottom-up approaches to estimate emissions from O&G and other sources. Emissions from O&G facilities (production sites, compressor stations, and processing plants) were estimated with emission factors calculated using Monte Carlo simulations, which account for the uncertainty associated with the variability of measured site emission rates.[22] Other emission sources were estimated using data from the GHGRP, GHGI, and published literature. Activity factors were spatially referenced to estimate emissions within grid cells, similar to the approach used in Jeong et al.[23] The 4 km × 4 km grid cells conform to the Comprehensive Air Quality Model Texas domain with extensions.[24] Emissions are grouped into three classes: O&G (active well to customer meter), other thermogenic (fossil sources not included in GHGI natural gas and petroleum systems), and biogenic. Emissions are reported as central estimates with 95th percent confidence intervals; total and category subtotal uncertainties are estimated by quadrature summation of the uncertainties in each source category.

Activity Factors. The number and location of O&G and other methane-emitting facilities were compiled from multiple state and federal databases. Facilities with annual greenhouse gas emissions of ≥25,000 t carbon dioxide equivalents (CO_2e) are required to submit annual emissions to the GHGRP.[16] These sites were classified as gas transmission, gas processing, gas gathering, landfills, or other industrial sites based on the GHGRP subpart under which they report. Additional O&G sources were identified using two data sets from the Texas Commission on Environmental Quality (TCEQ): the 2009 Barnett Shale Area Special Inventory (BSASI)[13] and the air permit database.[25] These sites were classified based on equipment type and facility name. Compressor stations were classified as gathering (upstream of processing) or transmission (downstream of processing) based on their proximity to gathering and transmission pipelines. O&G well locations were obtained from DI Desktop[26] and clustered into production sites as described in Zavala-Araiza et al.[15] Google Earth imagery was used to quality control reported spatial coordinates, manually locate sites without reported coordinates, and remove duplicate and decommissioned sites. The Supporting Information (SI) includes additional details on the compilation and classification of activity factors (section SI1), a map (Figure SI1), and a spreadsheet with facility locations.

Monte Carlo Simulations of O&G Emissions. For each O&G facility type, emission factors with a 95th percent confidence interval were calculated with Monte Carlo simulations that drew from facility-specific emission rate distributions assembled from measurements made during the Barnett Coordinated Campaign[27,28] and a recent national study on methane emissions from gathering and processing facilities.[31] Two emission rate distributions were used for each Monte Carlo simulation. The first emission rate distribution, defined as the "sampled distribution", was constructed from data collected by unbiased sampling of the Barnett region or the national population. Due to the positively skewed emission rate distribution of many O&G facility types,[3,28,31] the mean emission rate of a random sample may underestimate the average emission rate of the entire population if the sample size is insufficient to fully capture the highest end, or fat-tail, of the distribution. Figure 1 compares the sampled distributions by facility type to emission rates observed during the Barnett Coordinated Campaign using sampling methods biased toward higher emission sites.[27,29,30] These other Barnett data sets include measurements exceeding the maximum of the sampled distributions, which indicates unbiased sampling did not fully capture the fat-tail. To account for the effect of these high-emitting sites, we constructed a second emission rate distribution, defined as the "fat-tail site distribution", from data representing sites with emission rates exceeding the maximum value in the unbiased, sampled distribution of each facility type. A two-step Monte Carlo simulation was performed with the first step drawing from the sampled distribution and the second step drawing from the fat-tail site distribution. For each facility type, the probability of drawing from the fat-tail distribution was a best estimate based on the number of observed fat-tail sites compared to the total sites in the region. We ran sensitivity tests using a range of probabilities (0—5%) to test the effect of this assumption. Each Monte Carlo simulation included 10,000 iterations of random selection with replacement from one of the two emission rate distributions for every facility in the 25-county region. The 95th percent confidence interval of regional emission estimates was determined by the 2.5th percentile and 97.5th percentile of the 10,000 iterations. Facility-specific emission factors were calculated for each facility type by dividing the regional emission estimates by the number of facilities in the region (Figure SI2 illustrates the method). Spatially resolved O&G facility emissions were estimated by applying the emission factors to the spatially referenced activity data.

Compressor station emissions were estimated with two-phase Monte Carlo simulations drawing from site emission rate distributions constructed using data from a national study of gathering and processing facilities (Mitchell et al.)[31] and the Barnett Coordinated Campaign.[27,29,30] Gathering stations comprised over 90% of the compressor stations in the region. Transmission stations and storage facilities were treated identically to gathering compressor stations since they have similar equipment and installed engine horsepower. The sampled distribution was constructed using a national data set of 100 gathering stations with compression or a combination of compression and dehydration equipment with site emissions ranging from of 0 to 700 kg CH_4 h^{-1} and averaging 55 kg CH_4 h^{-1}.[31] The fat-tail site distribution included four gathering sites measured during the campaign ranging from 1,360 to 2,120 kg CH_4 h^{-1}.[27,29,30] The probability to draw from the fat-tail site distribution was set at 1%, which is equivalent to 2 to 3 compressor stations in the Barnett region with fat-tail emission rates at any moment in time. This probability was chosen based on the observation of four fat-tail sites over a 15-day period out of a population of 276 facilities. Multiple simulations were run with the probability of selecting from the fat-tail site distribution ranging from 0 to 5% to test the sensitivity of the outcome to the 1% assumption.

Processing plant emissions were estimated following a similar approach as compressor stations. Monte Carlo simulations drew from two sets of site emission rate distributions

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constructed using data from Mitchell et al.[31] for the unbiased sample distribution and the Barnett Coordinated Campaign[29,30] for the fat-tail site distribution. Because of the wide variation in processing plant size and complexity, processing plants were subdivided into two classes: large if they reported to the GHGRP or small if they did not. The average installed horsepower of Barnett plants (large = 21,000 HP, small = 8,000 HP) supports this division. The sampled distribution for large processing plants was constructed from a national data set of 16 processing plants with site emissions ranging from 4 to 600 kg CH_4 h^{-1} and averaging 170 kg CH_4 h^{-1}.[31] The sampled distribution for small processing plants was constructed from a national data set of nine gathering stations containing a combination of compression, dehydration, and treatment (C/D/T) equipment with site emissions ranging from 7 to 240 kg CH_4 h^{-1} and averaging 78 kg CH_4 h^{-1}.[31] Although C/D/T sites were not defined as processing plants by Mitchell et al., they have similarities to small plants including gas treatment and comparable installed horsepower (5000 HP).[31] Five processing plants measured during the campaign were used in the fat-tail distributions.[29,30] For large plants, the fat-tail distribution used two measurements exceeding the sampled distribution (750 and 1,720 kg CH_4 h^{-1}). For small plants, the fat-tail distribution used three measurements exceeding the sampled distribution (320, 390, and 490 kg CH_4 h^{-1}). The two higher values in the large plant fat-tail distribution were not used for small plants because they would require unreasonably high leak rates for these smaller throughput facilities. The probability of selecting from the fat-tail site distributions was set at 2%, which is equivalent to a single processing plant in the region with a fat-tail emission rate at any one moment. Multiple simulations were run with the probability of selecting from the fat-tail site distributions ranging from 0 to 5% to test the sensitivity of the outcome to the 2% assumption.

Production site emissions were estimated using a more complex approach that defined fat-tail sites based on proportional loss rates (methane emitted relative to methane produced). The method is briefly described below with additional details in the SI (section S2); the full method and results are found in Zavala-Araiza et al.[15] Activity factors were based on estimated O&G production site counts. Emission factors were derived with Monte Carlo simulations drawing from site emission rate distributions constructed using data from 226 sites measured during the Barnett Coordinated Campaign.[27−29] Activity and measurement data were divided into cohorts based on gas production and production-normalized emissions. As described in Zavala-Araiza et al.,[15] the sites with the highest proportional loss rates were defined as γ-sites; a fat-tail probability of 0.25% for γ-sites was chosen, and a sensitivity analysis was performed to test the effect of differing probabilities on estimated emissions. Zavala-Araiza et al.[15] reports emissions only for gas-producing sites. For this paper, gas-producing site emissions were divided into gas sites and oil sites based on the well type reported in DI Desktop.[26] In addition, emissions from oil sites with no gas production were estimated using an emission factor of 5.14×10^{-3} kg CH_4 h^{-1} $well^{-1}$ based on the Petroleum Systems stripper well emission factor in the GHGI.[17]

Other O&G Sources. Production site emissions estimated with the Monte Carlo simulations only included emissions during the operation phase. Additional emissions can occur episodically during drilling, completion flowback, or maintenance activities. Completion flowback emissions, which occur when a well is vented after hydraulic fracturing to prepare for routine production, were estimated for 73 individual well completions that occurred during the Barnett Coordinated Campaign based on well locations and completion start dates from DI Desktop.[26] In summary, emissions were estimated based on initial gas production with an assumption that gas wells, but not oil wells, controlled emissions due to federal regulations.[32] The average of the daily completion emission estimates during the campaign was used as the central estimate, and the minimum and maximum daily estimates were used as the lower and upper bound estimates. The detailed methods are described in SI Section SI3.

Gathering and transmission pipeline emissions were estimated from pipeline mileage and per mile emission factors. GIS shapefiles of gathering and transmission pipelines from DI Desktop[26] were joined with the grid to determine the miles of transmission and gathering pipelines in each grid cell. Emissions were estimated using the GHGI emission factors from the production and transmission and storage sectors.[17] Gathering pipelines used an emission factor of 4.7×10^{-2} kg CH_4 h^{-1} $mile^{-1}$ based on Midcontinent production sector emission factors for pipeline leaks, pipeline blowdowns, and mishaps. Transmission pipelines used an emission factor of 7.1×10^{-2} kg CH_4 h^{-1} $mile^{-1}$ based on transmission and storage sector emission factors for pipeline leaks and pipeline venting. Uncertainty was based on EPA's uncertainty estimate (+30%/−19%) for GHGI Natural Gas Systems.[17]

Natural gas distribution emissions were estimated using data from a recent national study of methane emissions from local distribution pipelines and metering and regulating (M&R) stations (Lamb et al.).[33] In summary, activity factors were based on data reported by Atmos Energy, which is the utility serving the vast majority of customers in the Barnett region. Emissions from sources not measured in Lamb et al.[33] were estimated using GHGI national emissions[17] prorated by activity factors. The detailed methods are described in SI Section SI4. The upper confidence limit uncertainty (+71%) was based on the emission factor uncertainties of Lamb et al.;[33] for the lower confidence limit, EPA's uncertainty estimate (−19%) for GHGI Natural Gas Systems[17] was used since Lamb et al. only report upper confidence limits.

Other Thermogenic Sources. Abandoned well emissions were estimated using well counts and a per well emission factor. The locations of inactive and plugged and abandoned wells in the Barnett region were obtained from DI Desktop.[26] For the subset of wells without coordinates, activity data were aggregated by county. The emission factor and uncertainty is based on the observed average emission rate of nine abandoned wells in the Marcellus Shale, 1.1×10^{-2} (+100%/−50%) kg CH_4 h^{-1} $well^{-1}$.[34]

Emissions from other industrial sources reporting to the GHGRP were based on reported 2013 emissions.[16] Annual emissions were converted to kg CH_4 h^{-1} by assuming a constant emission rate. Uncertainty for industrial source emissions was assumed to be +138%/−58%, which is the uncertainty of the combustion emission factor used to estimate GHGRP emissions.[35]

Residential and commercial end use emissions from leaks past the meter and incomplete combustion of natural gas by heaters and appliances were estimated using October 2013 gas deliveries to residential and commercial customers. Barnett gas consumption was estimated by prorating statewide monthly gas deliveries[36] by 2010 population.[37] For the central estimate, it

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Table 1. Activity Factors, Methane Emissions, and Percent of Emissions from Fat-Tail Sites by Source Category for the 25-County Barnett Shale Region[a]

source	activity factor	emissions (kg CH_4 h^{-1})	contribution from fat-tail sites (%)
gas production sites	15,044 well pads	16,400 (15,400−17,300)	11% (8−13%)
oil production sites	5,842 well pads	1,800 (1,700−1,900)	
well completions	38 gas wells	150 (30−290)	
	36 oil wells		
gathering compressor stations	259 facilities	18,700 (12,900−26,000)	33% (14−51%)
gathering pipelines	20,100 miles	940 (760−1,200)	
processing plants	22 large plants	5,500 (3,700−8,100)	11% (4−21%)
	16 small plants		
transmission and storage compressor stations	17 facilities	1,600 (1,100−2,200)	33% (14−51%)
transmission pipelines	3,300 miles	230 (190−300)	
local distribution	5,700 M&R stations; 11,700 pipeline leaks	920 (750−1,600)	
O&G subtotal		**46,200 (40,000−54,100)**	19% (14−26%)
abandoned wells	57,600 wells	630 (320−1,300)	
residential and commercial end use	5.6 MMcf/h gas delivered	160 (30−1,600)	
industrial facilities	56 facilities	60 (30−110)	
onroad vehicles (natural gas)	0.3 MMcf/h gas delivered	14 (7−68)	
onroad vehicles (gasoline and diesel)	65 billion vehicle miles traveled/yr	150	
geological seepage	57,900 km^2	1,100	
thermogenic subtotal		**48,400 (42,100−56,400)**	18% (14−26%)
landfills	21 GHGRP landfills	11,300 (5,000−16,900)	
	712 other landfills		
livestock	980,00 beef cattle	11,900 (9,500−14,300)	
	190,00 dairy cattle		
wastewater treatment	5,730,000 people	760 (560−670)	
biogenic subtotal		**24,000 (17,200−30,100)**	
emissions total		**72,300 (63,400−82,400)**	12% (9−15%)
% O&G		64% (52−78%)	
% thermogenic		67% (55−81%)	
% biogenic		33% (23−43%)	

[a]Numbers in parentheses are the 95th confidence interval. Estimates assumes a 0.25% fat-tail probability for production sites,[15] 1% fat-tail probability for compressor stations, and 2% fat-tail probability for processing plants.

was assumed that 0.16% of delivered gas was emitted, which is based on measurements of five California residences.[38] For the lower bound, a leak rate of 0.028% was based on the GHGI emission factor for residential and commercial stationary combustion.[17] For the upper bound, a leak rate of 1.6% was based on a Boston study that reported 2.7% of delivered gas was emitted−a state emission inventory estimated emissions from other sources in the Boston region were equivalent to 1.1% of delivered gas, so this leak rate assumes that the remainder of emissions in that study were due to residential and commercial end use.[39]

Methane emissions from gasoline and diesel onroad vehicle were based on county-level annual emissions reported in the 2011 National Emissions Inventory.[40] These emissions were estimated by the EPA using the MOVES2010b model.[41] No data were found on the uncertainty of these emission estimates, so uncertainty was conservatively excluded.

Natural gas vehicle emissions were estimated at the county-level based on the volume of natural gas delivered as vehicle fuel in October 2013 and assumed leak rates. The state-wide fuel delivery (210 MMscf)[36] was prorated by county-level vehicle miles traveled.[42] Emissions were assumed to equal 1% of fuel delivered with an uncertainty bound of 0.5—5%. This assumption is highly uncertain but has minor impact on the overall inventory due to the low usage of natural gas vehicles.

Geologic seepage emissions were estimated using a per area emission factor of 0.0184 kg CH_4 h^{-1} km^{-2}, which is based on a global average net flux of 4.42 mg CH_4 day^{-1} m^{-2} for microseepage and an assumption of 90% methanotrophic consumption.[43] This source category is highly uncertain, but no data were found to quantify the uncertainty; therefore, uncertainty was conservatively excluded.

Biogenic Sources. Emissions from landfills reporting to the GHGRP were based on reported 2013 emissions.[16] Annual emissions were converted to kg CH_4 h^{-1} by assuming a constant emission rate. We identified 712 additional landfills by querying TCEQ municipal solid waste permit data.[44] Based on the EPA estimate that 82% of landfill emissions are from facilities reporting to the GHGRP,[45] emissions from the nonreporting landfills were estimated by allocating 18% of GHGRP landfill emissions evenly among the 712 facilities. Uncertainty was based on EPA's uncertainty estimate (+49%/−56%) for GHGI landfills.[17] This uncertainty does not account for potential temporal variability in landfill emissions due to factors such as changing atmospheric pressure.

Livestock emissions from cattle manure management and enteric fermentation were estimated using activity data from the United States Department of Agriculture (USDA) and TCEQ and emission factors from the GHGI. Confined animal feeding operation (CAFO) locations and head counts of beef cattle, milking dairy cattle, and nonmilking dairy cattle were obtained from the TCEQ water quality general permit database.[46] County-level 2013 head counts of beef cattle, dairy cattle, and unspecified cattle were obtained from the USDA National

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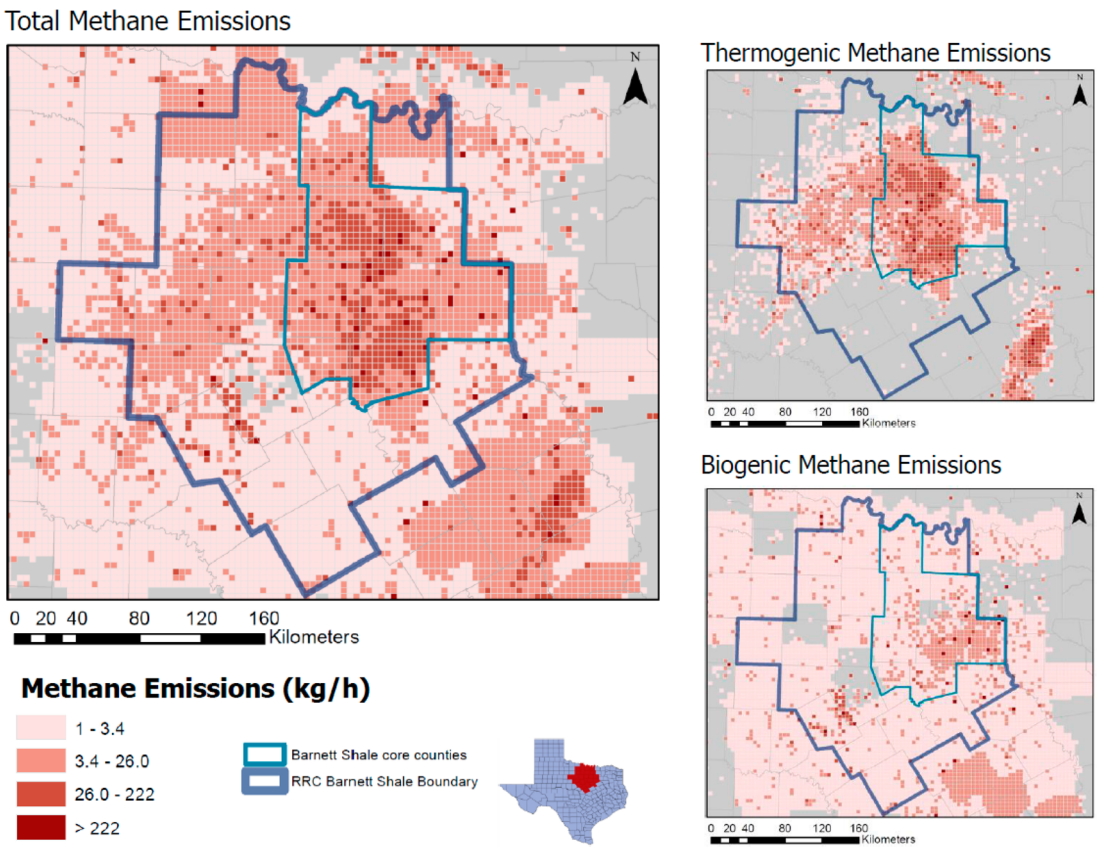

Figure 2. Spatially resolved methane emissions of the Barnett Shale region showing total, thermogenic, and biogenic emissions in 4 km × 4 km grid cells. Total emissions are reported in Table 2. The purple line is the boundary of the 25-county Barnett Shale region, and the blue line is the boundary of the 8-county core production area.

Agricultural Statistics Service database.[47] Unspecified cattle were assumed to be beef cattle. Dairy and beef cattle populations were further classified into detailed animal types by assuming the same proportion as the Texas 2012 cattle population used in the GHGI.[17] USDA county-level head counts were adjusted downward to account for the CAFO population in each county, which were treated separately as point sources. Enteric fermentation and manure management emission factors for beef cattle and dairy cattle animal types were derived from the GHGI Texas activity data and emissions.[17] Livestock methane emissions were estimated by multiplying the animal type head counts and the GHGI animal type emission factors for enteric fermentation and manure management. Livestock emission uncertainty was based on the Intergovernmental Panel on Climate Change Tier 2 methodology uncertainty (±20%),[48] which is similar to the approach used in the GHGI.[17]

Domestic wastewater treatment emissions were estimated from GHGI 2013 national emissions.[17] Population data from the 2010 US Census were used to prorate national emissions.[37] Population was spatially allocated based on census tract population data.[49] Uncertainty was based on EPA's uncertainty estimate (+2%/−39%) for GHGI Wastewater Treatment.[17]

Other potential methane sources in the region (e.g., reservoirs, wetlands, abandoned coal mines) were assumed to have negligible emissions.

Spatially Resolved Emission Inventory. Emissions data included sources with three levels of spatial resolution. GHGRP facilities, O&G facilities, landfills, and CAFOs were referenced to a specific latitude/longitude. Population-based and area-based emission estimates of natural gas distribution, wastewater treatment, residential and commercial end use, and geologic seepage were attributed to 4 km × 4 km grid cells based on the fractional area and population of each cell. Vehicle and a subset of abandoned well and livestock emissions were estimated at the county level with emissions spatially distributed across the grid proportional to the fraction of county land area in each cell. In addition to generating a gridded emission inventory by source category, emissions were estimated for the 25-county Barnett Shale region based on the spatial intersection of the grid cells and county boundaries.

Alternative Emission Inventory Estimates. Alternative O&G emission inventories were constructed using data from GHGRP, GHGI, and EDGAR. Emission data were scaled to account for the different spatial domains as described briefly below. The GHGRP inventory was based solely on 2013 reported emissions from regional O&G point sources and onshore production basins, which only includes facilities meeting the 25,000 t CO_2e reporting threshold.[16] The GHGI inventory was based on 2013 national emissions from Natural Gas Systems and Petroleum Systems[17] with individual source categories prorated by the ratio of Barnett region and national parameters such as gas production and transmission pipeline miles. The EDGAR inventory was based on EDGAR v4.2 2010 emissions from the gas production/distribution and oil production/refineries sectors.[19] Emissions were converted from 0.1° × 0.1° cells to the 25-county region using the spatial intersection of the cells and county boundaries. EDGAR 2010 emissions were extrapolated to 2013 using the 2013/2010 ratio of Barnett region gas and oil production from DI Desktop.[26] A

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Table 2. Comparison of the 25-County Barnett Region O&G Methane Emission Inventories from This Paper to Alternative Inventories Constructed from GHGI, GHGRP, and EDGAR[16−19]

	October 2013 emissions (kg CH_4 h^{-1})			
sector	Lyon et al. (95th percent CI)	GHGI	GHGRP	EDGAR
production	18,400 (17,100−19,500)	12,700	14,550	
gathering	19,600 (13,700−27,200)	2,700	4	
processing	5,500 (3,700−8,100)	8,700	800	
transmission and storage	1,800 (1,300−2,500)	2,700	300	
local distribution	920 (750−1,600)	4,300	1,350	
total O&G	**46,200 (40,000−54,100)**	**31,000**	**17,000**	**10,800**

detailed description of the methods used to construct each methane emission inventory for the 25-county Barnett Shale region in October 2013 is included in SI Section SI5.

RESULTS AND DISCUSSION

Barnett Region Emission Estimates. Estimated total emissions in the 25-county Barnett Shale region for October 2013 are 72,300 kg CH_4 h^{-1} (95th percent confidence interval = 63,400−82,400 kg CH_4 h^{-1}). O&G sources are estimated to emit 46,200 (40,000−54,100) kg CH_4 h^{-1} or 64% (52−78%) of total emissions (Table 1). Thermogenic sources, which include additional emissions from abandoned well, natural gas end use, and geologic seepage, are 48,400 (42,100−56,400) kg CH_4 h^{-1} or 67% (55−81%) of total emissions. Gathering compressor stations and active well pads are the largest emission sources, contributing 26% and 25% of total emissions, respectively. Livestock and landfills are the largest biogenic emission sources, contributing about 16% each. A core region of eight counties responsible for 94% of gas production and 43% of oil production contributes 67%, 77%, and 75% of the total, O&G, and thermogenic emissions, respectively, in the Barnett region (Figure 2).

Fat-tail sites contribute 19% (14−26%) of O&G emissions and 12% (9−15%) of total emissions in our reported inventory estimate, which assumes fat-tail emission rates at 0.25% of production sites,[15] 2% of processing plants, and 1% of compressor stations. At these probabilities, there would be approximately 50 production sites, 1 processing plant, and 2 to 3 compressor station with fat-tail emission rates somewhere in the Barnett region at any moment in time. The research teams were able to identify and measure these sites despite their limited numbers in a large region by utilizing specific sampling strategies (e.g., aircraft-based surveys targeting sites with high methane enhancements[30]). O&G site emission factors are dependent on the selected fat-tail site probability (Figure SI3). If the probability of fat-tail sites were reduced by half, O&G emissions would decrease by 8%, while at double the probability, O&G emissions would increase by 16%. Additional sensitivity analyses for production site emissions are reported in Zavala-Araiza et al.[15]

Fat-tail sites do not necessarily have persistently high emissions but may represent short-term emission events caused by maintenance activities or malfunctions. For production sites, fat-tail γ-sites included emission rates up to 287 kg CH_4 h^{-1}, approximately six times higher than the maximum emission rate observed using unbiased sampling.[27−29] An effort to identify high emitting sites in the Marcellus Shale region observed average emissions of 850 kg CH_4 h^{-1} at seven multiwell sites in the drilling phase, which the authors attributed to the conveyance of methane from overlying coal formations through the wellbore.[6] The high emission rates observed during the Barnett Coordinated Campaign do not appear to be related to drilling or hydraulic fracturing due to the infrequent occurrence of these activities during the campaign, but they may be caused by major malfunctions at production sites (e.g., stuck separator dump valve).[15] Another possibility is that measurements occurred during maintenance events such as venting to unload liquids accumulated in the wellbore. The median emission rate of unloading event from 107 wells in a nationwide study was equivalent to 257 kg CH_4 h^{-1}, similar to our fat-tail production site emission rates.[50] Based on the low number of unloading events reported to the GHGRP in the Barnett region,[16] emissions associated with liquids unloading are unlikely to be a major emission source in this case but may be substantial in regions with frequent unloading events such as the San Juan Basin.

For compressor stations and processing plants, the maximum fat-tail emission rates were 2,040 and 1,720 kg CH_4 h^{-1}, respectively. These emission rates are higher than the maximum annual average 2013 facility emissions reported to the GHGRP for transmission (520 kg CH_4 h^{-1}) and processing (1,050 kg CH_4 h^{-1}).[16] However, the GHGRP reports almost 2,400 unique blowdown (emptying or depressurizing a gas-filled vessel) events nationally exceeding 1,000 kg CH_4 total emissions in 2013, including over 800 events exceeding 10,000 kg CH_4.[16] Since the typical duration of these events range from minutes to hours, short-term blowdown events could cause fat-tail magnitude emission rates observed at compressor stations and processing plants during the Barnett Coordinated Campaign, but the probability of their observation is likely low. Additionally, GHGRP protocols may not capture high emissions from some malfunctions. For example, a recent national study of 45 transmission and storage compressor stations found two sites with emissions up to 1,000 kg h^{-1} likely caused by leaking isolation valves; the GHGRP-compliant on-site surveys reported emissions 2−3 orders of magnitude lower.[51]

For our study, we define fat-tail sites as those with emission rates above the sampled distribution, but this does not indicate that they are the only high emission sites. The sampled distributions are positively skewed and include sites with high emission rates, some of which had substantial tank venting due to equipment issues.[31] Consequently, there is a larger population of sites than the fat-tail sites in our analysis that contribute a large fraction of regional emissions and have avoidable, excess emissions.[15]

Barnett Shale O&G wells produced 5.6 Bcf day^{-1} natural gas and 54.5 Mbl oil and condensate day^{-1} in October 2013.[26] Assuming a constant production rate and weighted average gas composition of 88.5% methane by volume, our O&G emission estimate is equivalent to 1.2% (1.0−1.4%) of gas production. If oil production site emissions (4% of O&G total) are excluded,

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Table 3. Comparison of Activity Factors (AF) and Emissions Factors (EF) for the 25-County Barnett Region from This Paper, GHGI, and GHGRP[c]

		Lyon et al.	GHGI	GHGRP
active oil and gas wells (excludes completions)	AF	29,900 wells 20,900 well pads	34,800 wells	15,900 wells[b]
	EF	0.61 (0.57−0.64) kg CH_4 h^{-1} $well^{-1}$ 0.87 (0.82−0.92) kg CH_4 h^{-1} pad^{-1}	0.35 kg CH_4 h^{-1} $well^{-1}$	0.91 kg CH_4 h^{-1} $well^{-1}$
gathering stations (excludes pipelines)	AF	259 stations	1 large station 782 total stations[a]	54 stations
	EF	72 (50−100) kg CH_4 h^{-1}	1.7 kg CH_4 h^{-1}[a]	0.06 kg CH_4 h^{-1}
processing plants	AF	38 plants (16 small and 22 large)	55 plants	22 plants
	EF	average: 145 (84−231) kg CH_4 h^{-1} small: 84 (45−133) kg CH_4 h^{-1} large: 190 (112−301) kg CH_4 h^{-1}	159 kg CH_4 h^{-1}	36 kg CH_4 h^{-1}
transmission and storage stations (excludes pipelines)	AF	17 stations	25 stations	5 stations
	EF	72 (50−100) kg CH_4 h^{-1}	103 kg CH_4 h^{-1}	61 kg CH_4 h^{-1}

[a]GHGI only includes station counts for large gathering compressor stations. An alternative estimate of total station counts is based on the number of compressor engines. The emission factor is based on this alternative AF. [b]GHGRP data does not include well counts. Activity data was estimated from the county-level well counts reported in DI Desktop for GHGRP reporting operators. [c]GHGI factors are derived from national emissions and activity factors prorated by production, gas processed, and pipeline miles.[17] GHGRP factors are counts and average 2013 emissions of reporting facilities in the Barnett region.[16]

then the natural gas leak rate decreases to 1.1% (1.0% − 1.3%). Allocating emissions between natural gas and hydrocarbon liquids on an energy basis according to the methods of Zavala-Araiza et al.[52] attributes 95% of emissions to natural gas, resulting in a similar adjusted leak rate.

Comparing Inventories. The O&G emission inventory reported here for the Barnett region is a factor of 1.5 (1.3−1.7) greater than the emissions estimated from GHGI 2013 national emissions (46,200 versus 31,000 kg CH_4 h^{-1}; Table 3). This difference is similar to the ratio of ∼1.5 between top-down and GHGI estimates of total US methane emissions reported in Miller et al.[7] and Brandt et al.[8] Comparing the inventory reported here to an estimate based on GHGRP 2013 emissions (17,000 kg CH_4 h^{-1}) yields a factor of 2.7 (2.4−3.2) higher emissions from our inventory, comparable to the ratio of 2.5 between aircraft-based mass balance and GHGRP estimates of methane emissions from O&G activities in the Denver-Julesburg basin.[4] Comparing our inventory to an estimate derived from EDGAR 2010 emissions (10,800 kg CH_4 h^{-1}) yields a factor of 4.3 (3.7−5.0) higher emissions from our inventory, similar to the ratio of 4.9 ± 2.6 between O&G emission estimates for the south-central US based on atmospheric data versus EDGAR v4.2.[7]

Production sector emissions based on GHGI and GHGRP emissions are 31% and 21% lower than our estimate, respectively. Comparing the underlying activity factors and emission factors allows for a more detailed assessment of the inventories (Table 3). GHGI and GHGRP site emission factors are not directly reported in these data sources but derived by dividing total emissions by the number of sites. For the production sector, our activity and emission factors are converted from a per site basis to a per well basis using a factor of 1.4 wells $site^{-1}$ based on the 25-county average. Compared to our estimates for production sites excluding completions, the GHGI activity and emission factor are 16% higher and 43% lower, respectively. The GHGI emission factor may be lower because the data underlying the GHGI, which is from a 1990s study,[53] is not representative of current operational practices. The GHGRP has the opposite trend of the GHGI with a 47% lower activity factor and 49% higher emission factor than our estimate. The GHGRP emissions and activity factor are expected to be lower since the data only include facilities meeting the 25,000 t CO_2e reporting threshold. The higher GHGRP emission factor could be due to either reporting facility wells having higher emissions than the regional average or an overestimation of reported emissions caused by GHGRP methods. Our well pad emission factor (0.87 kg h^{-1}) is between the geometric means of Midcontinent region (0.54 kg h^{-1}) and Barnett well pads (1.19 kg h^{-1}) from two recent studies,[2,3] supporting the consistency of our estimates.

The largest difference among this work and other inventories is for the gathering sector; the GHGI and GHGRP emissions estimates are a factor of 7.3 and 4,900 lower than our estimate, respectively. Since the GHGI groups gathering within the production sector, we disaggregated emissions by assuming all compressor and pipeline emissions are associated with the gathering sector. The GHGI activity factor for gathering stations, which only includes large stations, greatly underestimates the number of facilities in the Barnett region. An alternative GHGI station activity factor can be estimated from the GHGI production sector compressor engine activity factor by assuming 3.1 compressors per station based on the average from Mitchell et al.[31] This alternative activity factor is three times higher than our facility count, which is probably because the study used to develop the GHGI compressor engine activity factors grouped together production site wellhead compressors and gathering station compressors.[53] Using this high GHGI activity factor, the GHGI emission factor is still 42 times lower than our emission factor. The GHGRP gathering station activity factor and emission factor are 4.8 and over 1,200 times lower than our factors, respectively. GHGRP Subpart W currently exempts gathering stations from reporting vented and fugitive methane emissions.[54] Therefore, GHGRP data only includes gathering facilities reporting combustion emissions under Subpart C.[55] Reporters are required to use a default methane emission factor based on natural gas turbines, which is known to be at least 2 orders of magnitude too low for reciprocating engines used by the vast majority of gathering stations.[56]

For the processing sector, GHGI emissions are 58% higher than our estimate. The GHGI emission factor is within 10% of

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our average processing plant emission factor; therefore, the higher emissions are caused primarily by a 45% higher activity factor. GHGRP emissions are a factor of 6.9 lower than our estimate—some of this difference is due to 16 of 38 plants that are not required to report to GHGRP, but the largest difference is from the emission factors. The average reported emissions of GHGRP processing plants is a factor of 5.5 lower than our large processing plant emission factor, which may be due to the exclusion of certain emission sources (e.g., tanks) from GHGRP reporting requirements.[54]

For the transmission and storage sector, the inventory comparisons have similar trends as processing. GHGI emissions are 50% higher than our estimate. The GHGI transmission and storage compressor station activity factor and emission factor (excluding pipeline emissions) are 47% and 43% higher than our factors, respectively. GHGRP emissions are a factor of 6.0 lower than our estimate, which primarily is due to about 70% of facilities not being required to report to the GHGRP. The average reported emissions of GHGRP facilities is 15% lower than our emission factor, which may be due to the exclusion of certain compressor engine operating mode emissions from GHGRP reporting requirements.[54] Our compressor station emission factor is within 10% of the value used for California compressor stations in Jeong et al.[23] The average emissions of five compressor stations and storage facilities measured during the Barnett Coordinated Campaign with on-site, leak and loss audits was 19 kg CH_4 h^{-1}.[57] This value is a factor of 3.8 lower than our emission factor but within 30% of the median value of our compressor station sampled distribution, which demonstrates how emissions could be underestimated if an emission factor is based on a small sample size of a skewed distribution.

For the distribution sector, our emission estimates are a factor of 4.7 and 1.5 lower than the GHGI and GHGRP estimates, respectively. This is due to our study using emission factors from a recent national study that reported emissions from pipelines and M&R stations have decreased since the 1990s when a previous study collected measurements used to develop the GHGI and GHGRP emission factors.[53]

Our detailed, spatially explicit methane emission inventory for the Barnett Shale region illustrates the limitations of relying on commonly used data sources such as GHGI and GHGRP to estimate regional emissions. The GHGI Natural Gas Systems section relies primarily on national emission factors developed in the 1990s to estimate natural gas industry emissions[53] and may not reflect regional differences or recent changes in emission profiles. The GHGRP only includes emissions from facilities meeting a reporting threshold and excludes most emissions from the gathering sector and certain emission sources; therefore, it is inherently an underestimate of emissions and should not be viewed as a complete emission inventory. EPA has recently made changes to improve the completeness of the GHGI and GHGRP and has proposed adding gathering facilities to the GHGRP.[58]

Our methane inventory estimates higher emissions than other inventories predominantly due to two reasons: more complete, region-specific activity factors and the inclusion of emissions from fat-tail sites. Our comprehensive search of multiple data sources revealed a substantially higher count of O&G facilities than was contained in any single data source, particularly with regards to gathering compressor stations. Relatively rare, high emitting fat-tail sites such as those observed during the Barnett Coordinated Campaign were estimated to contribute 19% of regional O&G emissions. Our estimate of total methane emissions from the 25-county Barnett Shale region, 72,300 (63,400−82,400) kg CH_4 h^{-1}, is not statistically significantly different from a top-down, aircraft-based estimate from the Barnett Coordinated Campaign,[21] 76,000 ± 13,000 kg CH_4 h^{-1}, which quantified emissions in areas intermediate to the 8-county core production area and 25-county region. The bottom-up estimate of thermogenic emissions from O&G and other fossil sources, 48,400 (42,100−56,400) kg CH_4 h^{-1}, is within the uncertainty bounds of the top-down estimate, 60,000 ± 11,000 kg CH_4 h^{-1}, of fossil emissions determined from source apportionment based on airborne ethane observations during the campaign.[20,21] Future studies comparing top-down and bottom-up emission estimates should ensure that emission inventories rely on comprehensive activity factors and contemporary emission factors that account for the highest emitting sites. Such efforts are likely to result in better agreement between top-down and bottom-up methods than previously has been reported.

■ ASSOCIATED CONTENT

Supporting Information

Additional information as described in the text. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/es506359c.

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Notes

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■ REFERENCES

(1) 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.* **2012**, *109* (17), 6435−6440, DOI: 10.1073/pnas.1202407109.

(2) Allen, D. T.; Torres, V. M.; Thomas, J.; Sullivan, D. W.; Harrison, M.; Hendler, A.; Herndon, S. C.; Kolb, C. E.; Fraser, M. P.; Hill, A. D.; et al. Measurements of methane emissions at natural gas production sites in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 17768−17773, DOI: 10.1073/pnas.1304880110.

(3) Brantley, H. L.; Thoma, E. D.; Squier, W. C.; Guven, B. B.; Lyon, D. Assessment of Methane Emissions from Oil and Gas Production

DOI: 10.1021/es506359c
Environ. Sci. Technol. 2015, 49, 8147−8157

Pads using Mobile Measurements. *Environ. Sci. Technol.* **2015**, *49*, 3219−3227, DOI: 10.1021/es5052809.
(4) 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*, 6836−6852, DOI: 10.1002/2013JD021272.
(5) Wecht, K. J.; Jacob, D. J.; Frankenberg, C.; Jiang, Z.; Blake, D. R. Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data. *J. Geophys. Res.: Atmos.* **2014**, *119*, 7741−7756, DOI: 10.1002/2014JD021551.
(6) Caulton, D. R.; Shepson, P. B.; Santoro, R. L.; Sparks, J. P.; Howarth, R. W.; Ingraffea, A. R.; Cambaliza, M. O. L.; Sweeney, C.; Karion, A.; Davis, K. J.; et al. Toward a better understanding and quantification of methane emissions from shale gas development. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 6237−6242, DOI: 10.1073/pnas.1316546111.
(7) Miller, S. M.; Wofsy, S. C.; Michalak, A. M.; Kort, E. A.; Andrews, A. E.; Biraud, S. C.; Dlugokencky, E. J.; Eluszkiewicz, J.; Fischer, M. L.; Janssens-Maenhout, G.; et al. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2013**, *110*, 20018−20022, DOI: 10.1073/pnas.1314392110.
(8) Brandt, A. R.; Heath, G. A.; Kort, E. A.; O'Sullivan, F.; Pétron, G.; Jordaan, S. M.; Tans, P.; Wilcox, J.; Gopstein, A. M.; Arent, D.; et al. Methane Leaks from North American Natural Gas Systems. *Science* **2014**, *343* (6172), 733−735, DOI: 10.1126/science.1247045.
(9) Allen, D. T. Methane emissions from natural gas production and use: reconciling bottom-up and top-down measurements. *Curr. Opin. Chem. Eng.* **2014**, *5*, 78−83, DOI: 10.1016/j.coche.2014.05.004.
(10) Texas Commission on Environmental Quality. Barnett Shale Information; TCEQ: Austin, TX, 2014. http://www.rrc.state.tx.us/oil-gas/major-oil-gas-formations/barnett-shale-information/ (accessed Apr 20, 2015).
(11) Browning, J.; Ikonnikova, S.; Gülen, G.; Tinker, S. Barnett Shale Production Outlook. *SPE Econ. Manage.* **2013**, *5*, 89−104.
(12) Zavala-Araiza, D.; Sullivan, D. W.; Allen, D. T. Atmospheric hydrocarbon emissions and concentrations in the Barnett shale natural gas production region. *Environ. Sci. Technol.* **2014**, *48*, 5314−5321, DOI: 10.1021/es405770h.
(13) Texas Commission on Environmental Quality. Barnett Shale Area Special Inventory, Phase 2; TCEQ: Austin, TX, 2014. https://www.tceq.texas.gov/assets/public/implementation/air/ie/pseiforms/Barnett%20Shale%20Area%20Special%20Inventory.pdf (accessed Apr 20, 2015).
(14) Eastern Research Group. City of Fort Worth Natural Gas Air Quality Study; City of Fort Worth: Fort Worth, TX, 2011. http://fortworthtexas.gov/gaswells/air-quality-study/final/ (accessed Apr 20, 2015).
(15) Zavala-Araiza, D.; Lyon, D.; Alvarez, R. A.; Palacios, V.; Harriss, R.; Lan, X.; Talbot, R.; Hamburg, S. P. Towards a Functional Definition of Methane Super-Emitters: Application to Natural Gas Production Sites. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/acs.est.5b00133.
(16) United States Environmental Protection Agency. Greenhouse Gas Reporting Program; EPA: Washington, DC, 2014. http://ghgdata.epa.gov/ghgp/main.do (accessed Nov 1, 2014).
(17) United States Environmental Protection Agency. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990−2013; EPA: Washington, DC, 2015. http://www.epa.gov/climatechange/ghgemissions/usinventoryreport.html (accessed Apr 20, 2015).
(18) European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL). Emission Database for Global Atmospheric Research, release version 4.2; 2011. http://edgar.jrc.ec.europa.eu (accessed Apr 20, 2015).
(19) 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*, 6898−6903, DOI: 10.1002/2014GL061503.
(20) Karion, A.; Sweeney, C.; Kort, E. A.; Shepson, P. B.; Brewer, A.; Cambaliza, M. O. L.; Conley, S.; Davis, K. J.; Deng, A.; Hardesty, M.; et al. Aircraft-based estimate of total methane emissions from the Barnett Shale region. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/acs.est.5b00217.
(21) 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**, DOI: 10.1021/acs.est.5b00219.
(22) Roy, A. A.; Adams, P. J.; Robinson, A. L. Air pollutant emissions from the development, production, and processing of Marcellus Shale natural gas. *J. Air Waste Manage. Assoc.* **2013**, *64*, 19−37, DOI: 10.1080/10962247.2013.826151.
(23) Jeong, S.; Millstein, D.; Fischer, M. L. Spatially explicit methane emissions from petroleum production and the natural gas system in California. *Environ. Sci. Technol.* **2014**, *48*, 5982−5990, DOI: 10.1021/es4046692.
(24) Texas Commission on Environmental Quality. Comprehensive Air Quality Model Domain Shapefile TX 4 km; TCEQ: Austin, TX, 2014. ftp://amdaftp.tceq.texas.gov/pub/TX/map/ (accessed Apr 20, 2015).
(25) Texas Commission on Environmental Quality. Air Permit Database; TCEQ: Austin, TX, 2014. http://www2.tceq.texas.gov/airperm/ (accessed Apr 20, 2015).
(26) Drillinginfo. DI Desktop; Drillinginfo: Austin, TX, 2015. http://www.didesktop.com/ (accessed March 1, 2015).
(27) Yacovitch, T. I.; Herndon, S. C.; Pétron, G.; Kofler, J.; Lyon, D.; Zahniser, M. S.; Kolb, C. E. Mobile Laboratory Observations of Methane Emissions in the Barnett. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/es506352j.
(28) Rella, C. W.; Tsai, T.; Botkin, C.; Crosson, E.; Steele, D. Measuring Emissions from Oil and Natural Gas Well Pads Using the Mobile Flux Plane Technique. *Environ. Sci. Technol.* **2015**, *49*, 4742−4748, DOI: 10.1021/acs.est.5b00099.
(29) Lan, X.; Talbot, R.; Laine, P.; Torres, A. Characterizing Fugitive Methane Emissions in the Barnett Shale Area Using a Mobile Laboratory. *Environ. Sci. Technol.* **2015**, accepted for publication, DOI: 10.1021/es5063055.
(30) Lavoie, T. N.; Shepson, P. B.; Cambaliza, M. O. L.; Stirm, B. H.; Karion, A.; Sweeney, C.; Yacovitch, T. I.; Herndon, S. C.; Lan, X.; Lyon, D. Aircraft-Based Measurements of Point Source Methane Emissions in the Barnett Shale Basin. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/acs.est.5b00410.
(31) Mitchell, A. L.; Tkacik, D. S.; Roscioli, J. R.; Herndon, S. C.; Yacovitch, T. I.; Martinez, D. M.; Vaughn, T. L.; Williams, L.; Sullivan, M.; Floerchinger, C.; et al. Measurements of methane emissions from natural gas gathering facilities and processing plants: Part 2. measurement results. *Environ. Sci. Technol.* **2015**, *49*, 3219−3227, DOI: 10.1021/es5052809.
(32) Standards of Performance for New Stationary Sources: Subpart OOOO—Standards of Performance for Crude Oil and Natural Gas Production, Transmission and Distribution. Public Law 77 FR 49542, 2012. http://www.ecfr.gov/cgi-bin/text-idx?SID=true&node=sp40.7.60.oooo (accessed Apr 20, 2015).
(33) Lamb, B. K.; Edburg, S. L.; Ferrara, T. W.; Howard, T.; Harrison, M. R.; Kolb, C. E.; Townsend-Small, A.; Dyck, W.; Possolo, A.; Whetstone, J. R. Direct Measurements Show Decreasing Methane Emissions from Natural Gas Local Distribution Systems in the United States. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/es505116p.
(34) Kang, M.; Kanno, C. M.; Reid, M. C.; Zhang, X.; Mauzerall, D. L.; Celia, M. A.; Chen, Y.; Onstott, T. C. Direct measurements of methane emissions from abandoned oil and gas wells in Pennsylvania. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 201408315 DOI: 10.1073/pnas.1408315111.
(35) Eastern Research Group. Emission Factor Documentation for AP-42 Section 1.4 Natural Gas Consumption; EPA: Washington, DC, 1998. http://www.epa.gov/ttnchie1/ap42/ch01/bgdocs/b01s04.pdf (accessed Apr 20, 2015).

(36) Energy Information Administration. Natural Gas Consumption by End Use; EIA: Washington, DC, 2014. http://www.eia.gov/dnav/ng/ng_cons_sum_a_EPG0_vgt_mmcf_m.htm (accessed Apr 20, 2015).
(37) United States Census Bureau. 2010 Census Summary File 1; US Census: Washington, DC, 2011. http://www2.census.gov/census_2010/04-Summary_File_1/Texas/ (accessed Apr 20, 2015).
(38) Walpert, T. H. Measuring Residential Natural Gas Leakage and Its Impact on CH_4 Emission Inventories in California. Senior Thesis, University of California-Berkeley, Berkeley, CA, 2013. http://nature.berkeley.edu/classes/es196/projects/2013final/WalpertT_2013.pdf (accessed Apr 20, 2015).
(39) McKain, K.; Down, A.; Raciti, S. M.; Budney, J.; Hutyra, L. R.; Floerchinger, C.; Herndon, S.; Nehrkorn, T.; Zahniser, M. S.; Jackson, R. B.; et al. Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts. *Proc. Natl. Acad. Sci. U. S. A.* **2015**, *112*, 1941−1946, DOI: 10.1073/pnas.1416261112.
(40) United States Environmental Protection Agency. *2011 National Emissions Inventory*; EPA: Washington, DC, 2013. http://www.epa.gov/ttn/chief/net/2011inventory.html (accessed Apr 20, 2015).
(41) United States Environmental Protection Agency. Motor Vehicle Emission Simulator (MOVES) Version 2010b; EPA: Washington, DC, 2010. http://www.epa.gov/otaq/models/moves/moves-docum.htm (accessed Apr 20, 2015).
(42) Texas Commission on Environmental Quality. Air Modeling FTP site, MOVES output files; TCEQ: Austin, TX, 2014. ftp://amdaftp.tceq.texas.gov/pub/Mobile_EI/Trends/mvs/ (accessed Apr 20, 2015).
(43) Etiope, G.; Klusman, R. W. Geologic emissions of methane to the atmosphere. *Chemosphere* **2002**, *49*, 777−789, DOI: 10.1016/S0045-6535(02)00380-6.
(44) Texas Commission on Environmental Quality. Inventory of Closed Municipal Solid Waste Landfills; TCEQ: Austin, TX, 2014. http://www.tceq.state.tx.us/permitting/waste_permits/waste_planning/wp_closed_lf_inv.html (accessed Apr 20, 2015).
(45) United States Environmental Protection Agency. *Technical Support Document for the Landfill Sector: Proposed Rule for Mandatory Reporting of Greenhouse Gases*; EPA: Washington, DC, 2009. http://www.epa.gov/ghgreporting/documents/pdf/archived/tsd/TSD-Landfill-sector.pdf (accessed Apr 20, 2015).
(46) Texas Commission on Environmental Quality. Water Quality General Permits & Registration Search; TCEQ: Austin, TX, 2014. http://www2.tceq.texas.gov/wq_dpa/index.cfm (accessed Apr 20, 2015).
(47) National Agriculture Statistics Service. Texas Cattle Inventory by County; United States Department of Agriculture: Washington, DC, 2014. http://www.nass.usda.gov/Statistics_by_State/Texas/Publications/County_Estimates/ce_tables/cecatt1.htm (accessed Apr 20, 2015).
(48) Dong, H.; Mangino, J.; McAllister, T. A.; Hatfield, J. L.; Johnson, D. E.; Lassey, K. R.; de Lima, M. A.; Romanovskaya, A. 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Vol. 4: Agriculture, Forestry, and Other Land Use, Chapter 10: Emissions from Livestock and Manure Management; IPCC: Paris, France, 2006. http://www.ipcc-nggip.iges.or.jp/public/2006gl/pdf/4_Vol.4/V4_10_Ch10_Livestock.pdf (accessed Apr 20, 2015).
(49) United States Census Bureau. 2013 TIGER/Line® Shapefiles: Census Tracts; US Census: Washington, DC, 2013. https://www.census.gov/cgi-bin/geo/shapefiles2013/layers.cgi (accessed Apr 20, 2015).
(50) Allen, D. T.; Sullivan, D. W.; Zavala-Araiza, D.; Pacsi, A. P.; Harrison, M.; Keen, K.; Fraser, M. P.; Daniel Hill, A.; Lamb, B. K.; Sawyer, R. F.; et al. Methane Emissions from Process Equipment at Natural Gas Production Sites in the United States: Liquid Unloadings. *Environ. Sci. Technol.* **2014**, *49*, 641−648, DOI: 10.1021/es504016r.
(51) Subramanian, R.; Williams, L.; Vaughn, T. L.; Zimmerle, D.; Roscioli, J. R.; Herndon, S. C.; Yacovitch, T. I.; Floerchinger, C.; Tkacik, D. S.; Mitchell, A. L.; et al. Methane emissions from natural gas compressor stations in the transmission and storage sector: Measurements and comparisons with the EPA greenhouse gas reporting program. *Environ. Sci. Technol.* **2015**, DOI: 10.1021/es5060258.
(52) Zavala-Araiza, D.; Allen, D. T.; Harrison, M.; George, F. C.; Jersey, G. B. Allocating Methane Emissions to Natural Gas and Oil Production in Shale Formations. *Environ. Sci. Technol.* **2015**, *3*, 492−498, DOI: 10.1021/sc500730x.
(53) Harrison, M. R.; Shires, T.; Wessels, J. K.; Cowgill, R. M. *Methane emissions from the natural gas industry*; EPA: Washington, DC, 1996. http://www.epa.gov/methane/gasstar/tools/related.html (accessed Apr 20, 2015).
(54) Mandatory Greenhouse Gas Reporting: Subpart W - Petroleum and Natural Gas Systems. Public Law 75 FR 79140, 2010. http://www.ecfr.gov/cgi-bin/retrieveECFR?gp=&SID=d7a88dd57a772702658ef74704f9f395&n=pt40.21.98&r=PART&ty=HTML#sp40.21.98.w (accessed Apr 20, 2015).
(55) Mandatory Greenhouse Gas Reporting: Subpart C - General Stationary Fuel Combustion Sources. Public Law 78 FR 71952, 2013. http://www.ecfr.gov/cgi-bin/text-idx?SID=d7a88dd57a772702658ef74704f9f395&node=sp40.21.98.c (accessed Apr 20, 2015).
(56) United States Environmental Protection Agency. AP-42, Fifth ed., Vol. I, Chapter 3: Stationary Internal Combustion Sources, Part 3.2: Natural Gas-fired Reciprocating Engines; EPA: Washington, DC, 2000. http://www.epa.gov/ttn/chief/ap42/ch03/final/c03s02.pdf (accessed Apr 20, 2015).
(57) Johnson, D.; Covington, A.; Clark, N. Methane emissions from leak and loss audits of natural gas compressor stations and storage facilities. *Environ. Sci. Technol.* **2015**, accepted for publication, DOI:10.1021/es506163m.
(58) 40 CFR Part 98 Greenhouse Gas Reporting Rule: 2015 Revisions and Confidentiality Determinations for Petroleum and Natural Gas Systems; Proposed Public Law, 2014. http://www.gpo.gov/fdsys/pkg/FR-2014-12-09/pdf/2014-28395.pdf (accessed Apr 20, 2015).

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