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Environmental Science & Technology

Article

Cite This: Environ. Sci. Technol. 2019, 53, 4619−4629

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Characterizing Regional Methane Emissions from Natural Gas Liquid Unloading

George G. Zaimes,[†] James A. Littlefield,[*,†] Daniel J. Augustine,[†] Gregory Cooney,[†] Stefan Schwietzke,[‡,§,#] Fiji C. George,[∥] Terri Lauderdale,[⊥,∇] and Timothy J. Skone[†]

[†]National Energy Technology Laboratory, 626 Cochrans Mill Road, P.O. Box 10940, Pittsburgh, Pennsylvania 15236, United States

[‡]NOAA Earth System Research Laboratory, 325 Broadway, Boulder, Colorado 80305, United States

[§]Cooperative Institute for Research in Environmental Sciences, University of Colorado, 216 UCB, Boulder, Colorado 80309, United States

[∥]Cheniere Energy, Inc., 700 Milam Street, Suite 1900, Houston, Texas 77002 United States

[⊥]AECOM, 9400 Amberglen Boulevard, Austin, Texas 78729, United States

Supporting Information

ABSTRACT: A "bottom-up" probabilistic model was developed using engineering first-principles to quantify annualized throughput normalized methane emissions (TNME) from natural gas liquid unloading activities for 18 basins in the United States in 2016. For each basin, six discrete liquid-unloading scenarios are considered, consisting of combinations of well types (conventional and unconventional) and liquid-unloading systems (nonplunger, manual plunger lift, and automatic plunger lift). Analysis reveals that methane emissions from liquids unloading are highly variable, with mean TNMEs ranging from 0.0093% to 0.38% across basins. Automatic plunger-lift systems are found to have significantly higher per-well methane emissions rates relative to manual plunger-lift or non-plunger systems and on average constitute 28% of annual methane emissions from liquids unloading over all basins despite representing only ∼0.43% of total natural gas well count. While previous work has advocated that operational malfunctions and abnormal process conditions explain the existence of super-emitters in the natural gas supply chain, this work finds that super-emitters can arise naturally due to variability in underlying component processes. Additionally, average cumulative methane emissions from liquids unloading, attributed to the natural gas supply chain, across all basins are ∼4.8 times higher than those inferred from the 2016 Greenhouse Gas Reporting Program (GHGRP). Our new model highlights the importance of technological disaggregation, uncertainty quantification, and regionalization in estimating episodic methane emissions from liquids unloading. These insights can help reconcile discrepancies between "top-down" (regional or atmospheric studies) and "bottom-up" (component or facility-level) studies.

INTRODUCTION

Technological advancements in horizontal drilling and hydraulic fracturing contributed to accelerated production and use of natural gas resources in the United States and have fundamentally transformed the U.S. energy landscape.[1−3] Recent efforts to improve the economic efficacy and environmental performance of natural gas systems have focused on identifying cost-effective technologies and best practices that simultaneously enhance operational efficiency and reduce emissions of methane, the primary constituent of natural gas and a potent greenhouse gas, across all stages of the natural gas supply chain.[4] However, methane emissions from natural gas systems are highly variable and dependent on multiple factors including throughput, reservoir type, well age, equipment and processing technology, geography, and operator practices.[5,6] Consequently, studies have reported divergent results regarding the methane emissions from natural gas systems, driven by differences in analytic methods, system boundaries, and modeling assumptions.[7,8] As such, prioritizing strategies for efficiency improvement and emissions mitigation requires (1) a robust understanding of the regional variability in methane emissions across natural gas infrastructure and reservoir types using a common set of system boundary and modeling assumptions and (2) a mechanistic understanding of operating conditions within facilities.[9]

Historically, two types of frameworks have been applied to estimate methane emissions from natural gas operations: (1)

Received: October 1, 2018
Revised: February 15, 2019
Accepted: March 13, 2019
Published: March 29, 2019

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"top-down" methods[9−19] that rely on atmospheric measurements via air sampling from aircraft, tower, and ground instrumentation as well as satellite and remote sensing approaches; and (2) "bottom-up" methods[20−24] that use measurements at the component or facility level, or engineering calculations based on first principles. Bottom-up methods are leveraged by the U.S. Environmental Protection Agency (EPA) in developing national greenhouse gas emissions inventories.[25] Brandt et al. performed a meta-analysis to systematically compare CH_4 emissions from North American natural gas systems using scientific literature complied over 20 years.[26] Their analysis revealed a distinct asymmetry in reported methane emissions, with top-down studies consistently estimating higher methane emissions relative to bottom-up studies and suggest that U.S. national emissions inventories under-estimate methane emissions from the natural gas supply chain. However, reconciliation between "top-down" and "bottom-up" studies is technically challenging as data reported across studies is often spatiotemporally incongruent and may contain systematic bias.[27,28]

Sources of variability and bias common to "top-down" or "bottom-up" studies include but are not limited to data representativeness, sample size, sampling bias, methodological variations, temporal and geographic incongruency, system boundaries, and modeling assumptions. Regional, "top-down" studies are often constrained in spatiotemporal resolution, with a high volume of measurement samples taken over short-duration sampling campaigns.[26,29,30] Further, atmospheric measurements taken during aircraft fly-overs are typically performed mid-day, when sufficient atmospheric mixing has occurred, and the planetary boundary layer is well-developed. Due to the time-envelope for measurement sampling, airplane fly-overs may detect diurnal peak emissions from episodic sources (such as liquid unloading) that occur during day-time operations, as compared with nominal averages.[9] Additionally, apportioning methane emissions to natural gas operations is technically challenging, as biogenic and other non-natural gas industrial sources of methane (e.g., natural seeps or concurrent oil operations) constitute a source of uncertainty and can confound source attribution. Despite these technical difficulties, "top-down" methods are an essential counterpoint to component-level measurements given their independence from industry reported emissions and bottom-up calculations. Bottom-up estimates using engineering design equations may lack the technological and geographic specificity to accurately model methane emissions from all emission sources, and studies that rely on normalized or average values fail to capture variability in emissions estimates. Furthermore, empirical evidence has suggested that methane emissions from natural gas systems are extremely heterogeneous, with a small portion of natural gas activities responsible for a disproportionately large fraction of total emissions;[31] these infrequent but high-emitting activities or sites are colloquially referred to as "super-emitters".[32,33] Bottom-up studies with small sample sizes may underrepresent the influence of super-emitters in emission inventories.

Recent efforts to reconcile these discrepancies have resulted in the hybridization of top-down and bottom-up methods.[34] Schwietzke et al. 2017 performed the first comprehensive comparison study, using consistent spatiotemporally resolved "top-down" and "bottom-up" methods, to estimate methane emissions from natural gas operations in the Fayetteville Shale play.[9] Schwietzke et al. 2017 found that operator reported episodic emissions from natural gas liquids unloading account for a substantial portion (up to ∼33%) of observed midday CH_4 emissions measured via airplane fly-over. These findings suggest that episodic methane emissions from liquids unloading can (1) partially rectify discrepancies between top-down and bottom-up estimates and (2) explain daily variability in measured atmospheric methane concentrations. Additionally, in cases in which the plunger system or other artificial lift technology malfunctions, the blowdown characteristics may exhibit the emissions profile of a manually unloaded well, resulting in higher than designed emission rates, although the event type may be reported to monitoring programs according to its technology type (e.g., gas lifts, plunger-lifts, etc.). Consequentially, "bottom-up" emissions inventories using only reported design specifications may underestimate cumulative emissions from these sources. As component-level inventories are primarily utilized to inform national policy, an improved "bottom-up" synthesis of methane emissions from natural gas liquids unloading that accounts for technological differentiation across plunger-types, regionality, and uncertainty is critical for accurate emissions characterization and for guiding effective methane-mitigation strategies.

Natural Gas Liquid Unloading. Natural gas liquids unloading is a process used to remove liquids (produced water, oil, or condensate) that may accumulate in the well production tubing downhole. Accumulation of liquids in the wellbore can occur due to a variety of reasons including temporal changes in the produced gas-to-liquid ratio, decreasing produced gas velocity, and declining reservoir pressure.[35] The hydrostatic head created by the accumulation of liquids in the wellbore can restrict or inhibit gas flow; thus, intermittent liquids unloading is necessary to restore and maintain normal gas production. Three liquids unloading systems are analyzed in this work: nonplunger systems, manual plunger-lift systems, and automatic plunger-lift systems. Due to data limitations, automatic nonplunger systems are not considered in the analysis. In nonplunger systems, the well operator manually vents a well to the atmosphere, a process referred to as well blowdown, to remove liquid build-up in the wellbore and re-establish gas flow. Plunger-lift systems utilize gas pressure buildup in the casing-tubing annulus to drive a mechanical plunger and lift the column of accumulated fluid out of the well. In manual plunger-lift systems, a well operator manually initiates the plunger-lift cycle, while in automatic plunger-lift systems, the plunger-lift cycle is automated via the use of control technologies.

Research Objectives. An increasing body of literature suggests that episodic methane emissions from natural gas liquids unloading are substantial[20] and potentially under-characterized in the literature.[9] As such, this study performs a robust "bottom-up" synthesis of methane emissions from natural gas liquids unloading that accounts for technological differentiation across plunger-types, regionality, and uncertainty quantification under a consistent and common set of system boundary and modeling assumptions.

In this work, a "bottom-up" probabilistic model was developed to characterize basin-level methane emissions from six discrete liquids unloading scenarios consisting of a combination of well types (conventional and unconventional), and liquid-unloading systems (nonplunger, manual plunger lift, and automatic plunger lift), evaluated over 18 U.S. natural gas basins. The primary objectives of this study are as follows: (1) compare methane emissions from liquids unloading across 18

DOI: 10.1021/acs.est.8b05546
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U.S. natural gas basins, (2) characterize variability in methane emissions metrics across multiple geospatial scales, (3) determine key factors that drive liquids unloading emissions at the component-level, (4) quantify the contribution of six discrete liquids unloading scenarios to basin-wide methane emissions, and (5) benchmark the results against emissions reported in the 2016 Greenhouse Gas Reporting Program (GHGRP).

METHODS

This work quantifies throughput normalized methane emissions (TNME) from natural gas liquids unloading activities in 2016 for the following 18 natural gas basins in the United States: Anadarko, Appalachian, Appalachian (Eastern Overthrust), Arkla, Arkoma, Chautauqua, Denver–Julesberg, East Texas, Fort Worth Syncline, Green River, Gulf Coast, Permian, Piceance, San Juan, South Oklahoma Folded Belt, Strawn, Uinta, and Williston. These basins were selected as they represent the most productive U.S. onshore natural gas production regions and account for over 80% of total U.S. natural gas production in 2016.[36] In this work, TNME is defined as the ratio of methane emissions (kilograms of CH_4) from liquids unloading that are apportioned to the natural gas supply chain, via energy-based allocation, to the methane component of produced natural gas (kilograms of CH_4); see eq 1:

$$\text{TNME\%} = \frac{\text{methane emissions (kg CH}_4\text{)}}{\text{natural gas produced (kg CH}_4\text{)}} \quad (1)$$

Per-well methane emissions (kilograms of CH_4 per well per year) for nonplunger and plunger-lift systems are developed by modifying equations W-8 and W-9 from the EPA's GHG reporting rule in 40 CFR 98 Subpart W[37] and provided in eqs 2 and 3, respectively:

$$E_{\text{non-plunger}} = [(V \times (0.37 \times 10^{-3}) \times \text{CD}^2 \times \text{WD} \times \text{SP}) + V(\text{SFR} \times (\text{HR} - 1.0) \times Z)] \times V_{\text{frac,CH}_4} \times \text{density}_{\text{CH}_4} \times \text{Alloc}_{\text{CH}_4} \quad (2)$$

$$E_{\text{plunger-lift}} = [(V \times (0.37 \times 10^{-3}) \times \text{TD}^2 \times \text{WD} \times SP) + V(\text{SFR} \times (HR - 0.5) \times Z)] \times V_{\text{frac,CH}_4} \times \text{density}_{\text{CH}_4} \times \text{Alloc}_{\text{CH}_4} \quad (3)$$

Key parameters used in estimating methane emissions from liquids unloading include: venting frequency (V), casing diameter (CD) or tubing diameter (TD), well depth (WD), shut in pressure (SP), standard flow rate (SFR), venting duration (HR), volumetric fraction of methane in produced natural gas ($V_{\text{frac,CH}_4}$), volumetric density of methane ($\text{density}_{\text{CH}_4}$), and fraction of methane emissions from liquids unloading that are allocated to the methane component of natural gas ($\text{Alloc}_{\text{CH}_4}$), summarized in Table S1. Multiple data sources including the American Petroleum Institute (API) and America's Natural Gas Alliance (ANGA) survey,[38] DrillingInfo (DI) Desktop,[36] the 2016 Greenhouse Gas Reporting Program (GHGRP),[39] and peer-reviewed literature are used to parametrize key model inputs. Additionally, data used to populate the model span multiple tiers of data resolution from the production site, facility level, county level, basin level, National Energy Modeling System (NEMS) regions, and global level.

Basin-Level Methane Emissions. Basin-level methane emissions were quantified for six discrete liquids unloading scenarios, consisting of a combination of two well types (conventional and unconventional), and three liquids unloading systems (nonplunger, manual plunger-lift, and automatic plunger-lift). To preserve data quality, methane emissions from natural gas liquids unloading were calculated at the county-level and subsequently aggregated to the basin-level. Counties were mapped to basins using a standardized cross-walk developed by the Environmental Protection Agency (EPA),[40] and natural gas basins were defined based on American Association of Petroleum Geologists (AAPG) geologic province codes.[41]

Let ($E_{i,j}$) denote the per-well methane emissions associated with liquids unloading, and ($W_{i,j}$) denote the venting well count for the ith liquids unloading scenario and j^{th} county, with $i = 1 \cdots 6$ and $j = 1 \cdots n$, wherein n is the number of counties in the select basin. Basin-level ($E_{\text{CH}_4,\text{basin}}$) methane liquids unloading emissions were calculated as the sum of the product of the per well methane emissions from liquids unloading and venting well count over all i and j; see eq 4:

$$E_{\text{CH}_4,\text{basin}} = \sum_{i=1}^{6} \sum_{j=1}^{n} E_{i,j} \times W_{i,j} \quad (4)$$

Emissions Attribution: Energy-Based Allocation. Natural gas is often co-produced with liquid hydrocarbons from the same well,[42] thus allocating emissions from natural gas operations across the entire product slate [e.g., methane, natural gas liquids, condensates (C_{2+}), and crude oil] is critical for accurate emissions attribution and for demarcating the environmental burdens between petroleum and natural gas supply chains. This work used the life cycle assessment (LCA) framework for apportioning emissions across multi-output product systems, i.e., via attributing emissions to products in proportion to their relative fraction of total economic value, energy, mass, or other underlying physical relationship.[43] In this work, energy-based allocation was performed at the county level and used to apportion methane emissions from liquids unloading across the methane component of produced natural gas (CH_4), natural gas liquids and condensates (C_{2+}), and co-produced oil. In energy-based allocation, emissions are weighted across products based on the relative energy content (e.g., BTU) of each product stream. Additional details of the allocation methodology and procedure are provided in section S4 of the Supporting Information.

Monte Carlo Simulation. Monte Carlo simulation (10 000 iterations) was performed to generate statistical bounds for basin-level TNME. Probability distribution functions (PDFs) were fit to sample data for key parameters in eqs 2 and 3. Multiple distributions were tested against the sample data and compared across several goodness of fit (GOF) indicators including the Akaike information criteria (AIC), Bayesian information criteria (BIC), Chi-squared statistic, Kolmogorov–Smirnov (K–S) statistic, and Anderson Darling (AD) statistic. Several heuristics were used to pick the "best-fit" distribution ensuring that: (1) the distribution is physically relevant, (2) the distribution is well-ranked across GOF indicators, and (3) there is precedence in the literature for using said distribution to characterize the underlying data.

DOI: 10.1021/acs.est.8b05546
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Additional information is provided in section S5 of the Supporting Information.

■ DATA ACQUISITION

DI Desktop. Data for 2016 natural gas operations including cumulative gas produced, cumulative oil produced, well count, well depth, date of first production, producer type, and drilling type was obtained from DrillingInfo (DI) desktop.[36] Data sourced from DI desktop was screened and cleaned using a series of search criteria and filters to remove extraneous data uncharacteristic of natural gas activities that may be commingled in the data set (such as petroleum operations):

- contains at least one of the following producer types: "GAS", "OIL", "O&G", or "CBM";
- well production occurs between the time horizon of January 1, 2016 and December 31, 2016;
- for producer types "OIL" and "O&G", wells with a gas-to-oil ratio less than 100 MCF bbl^{-1} were excluded from the analysis;[44] and
- wells with null cumulative gas production were excluded from the analysis.

In this work, wells were classified as conventional or unconventional based on producer type and drilling type, consistent with prior published literature.[44] Wells with producer type Coal Bed Methane (CBM) were classified as unconventional, otherwise wells were characterized based on drilling type. DI desktop characterizes wells across four drilling types: vertical, horizontal, directional, or unknown. Vertical wells were classified as conventional, while wells with drilling type horizontal or directional were flagged as unconventional. Wells with drilling type unknown were considered conventional if the date of first gas production occurred in or prior to 2002, while wells that produce post 2002 were considered unconventional. As large-scale unconventional natural gas production first became a commercial reality in the early 2000s,[1] a cutoff year of 2002 was chosen to assign unknown wells into conventional or unconventional categories. In this work, the classification of conventional or unconventional is important and used to map key well characteristics (e.g., casing diameter, tubing diameter, etc.) across data sets, and thus not merely for categorical differentiation.

Data obtained from DI desktop (e.g. cumulative gas production, oil production, etc.) was aggregated to the county-level and stratified by well type (conventional or unconventional). County-level SFR (scf NG $well^{-1}$ h^{-1}) was derived based on county-level cumulative natural gas production, well count, and well type. Triangular distributions for well depth, stratified by well type, were constructed at the county-level via considering the min, max, and expected value (average) depth across the subset of conventional and unconventional wells for each county. For instances in which well depth is unavailable or not reported for a select county, triangular distributions were developed using basin-level min, max, and expected values. Additional information regarding the screening procedure used to clean data from DI desktop as well as relevant basin-level summaries are provided in section S3 of the Supporting Information.

Previous Estimates. Allen et al. 2015 provide estimates of liquids unloading event frequency and duration for non-plunger, manual, and automatic plunger-lift wells based on measurements collected at the production site.[20] Additionally, the authors report the volumetric fraction of methane in produced gas for each of the sampled wells, and relevant metadata including study region (Appalachia, Mid-Continent, Rocky Mountain, Gulf Coast), well direction, etc. In this work, data reported in Allen et al. 2015 was used to parametrize probability distributions for the venting frequency (V) and venting duration (HR) of liquids unloading systems. The mean venting frequency for nonplunger, manual, and automatic-plunger lift systems obtained via Monte Carlo simulation was 39, 14, and 2450 vents per well per year, respectively. Triangular distributions were used to model the volumetric fraction of methane in natural gas (V_{frac,CH_4}), stratified by NEMS region, based on data reported in Allen et al. 2015. It was assumed that the volumetric methane concentration of natural gas is dependent on the geographic region but invariant to liquids unloading system or well type. Values for V_{frac,CH_4} were calculated at a NEMS level and mapped to county-level data obtained from DI desktop. Additional details regarding V and HR are provided in section S5 in the Supporting Information, and information for V_{frac,CH_4} is provided in section S8 of the Supporting Information.

GHGRP. The EPA's Greenhouse Gas Reporting Program (GHGRP) collects facility-level greenhouse gas data from major industrial sources across the United States. The 2016 GHGRP data set contains key information for natural gas liquids unloading activities including facility-level averages for well parameters (CD, TD, WD, and SP), number of wells that vent for liquids unloading, cumulative number of liquids unloading events, and natural gas emissions from liquids unloading reported during the 2016 period. In this work, data from the 2016 GHGRP was used to (1) populate triangular distributions for well CD and TD stratified by well type and (2) determine the fraction of wells that vent for liquids unloading stratified by well type and liquids unloading system. The latter was applied to county-level well count obtained from the DI desktop to derive county-level venting well count ($W_{i,j}$). This approach implicitly assumes that the fraction of wells that vent for liquids unloading sampled in the GHGRP are representative of the well population and, thus, can be used to characterize county-level data obtained from DI desktop.

Onshore natural gas facilities reported by the GHGRP are mapped to natural gas basins using AAPG geologic provinces codes and are classified as conventional or unconventional based on formation type. Facilities with the formation types "coal seam" or "shale gas" are classified as unconventional, while the types "high permeability gas", "oil", or "other tight reservoir rock" are flagged as conventional. Facility-level average CD and TD reported in the GHGRP are mapped to county-level data obtained from DI desktop. Triangular distributions for CD and TD, stratified by well type, were constructed at the county-level via considering the min, max, and expected value (venting well weighted average) across all facilities that lie within said county. In cases in which there is no direct mapping between facility-level data reported in the GHGRP and county-level data obtained from DI desktop, triangular distributions for CD and TD were developed using basin-wide min, max and expected values. It is important to note that the 2016 GHGRP data set is largely limited to CD and TD, with sparse to no coverage for WD or SP.

The 2016 GHGRP broadly classifies wells that vent for liquids unloading into nonplunger and plunger-lift categories but does not distinguish between manual or automatic-plunger-lift subtypes. To disaggregate the plunger-lift category,

DOI: 10.1021/acs.est.8b05546
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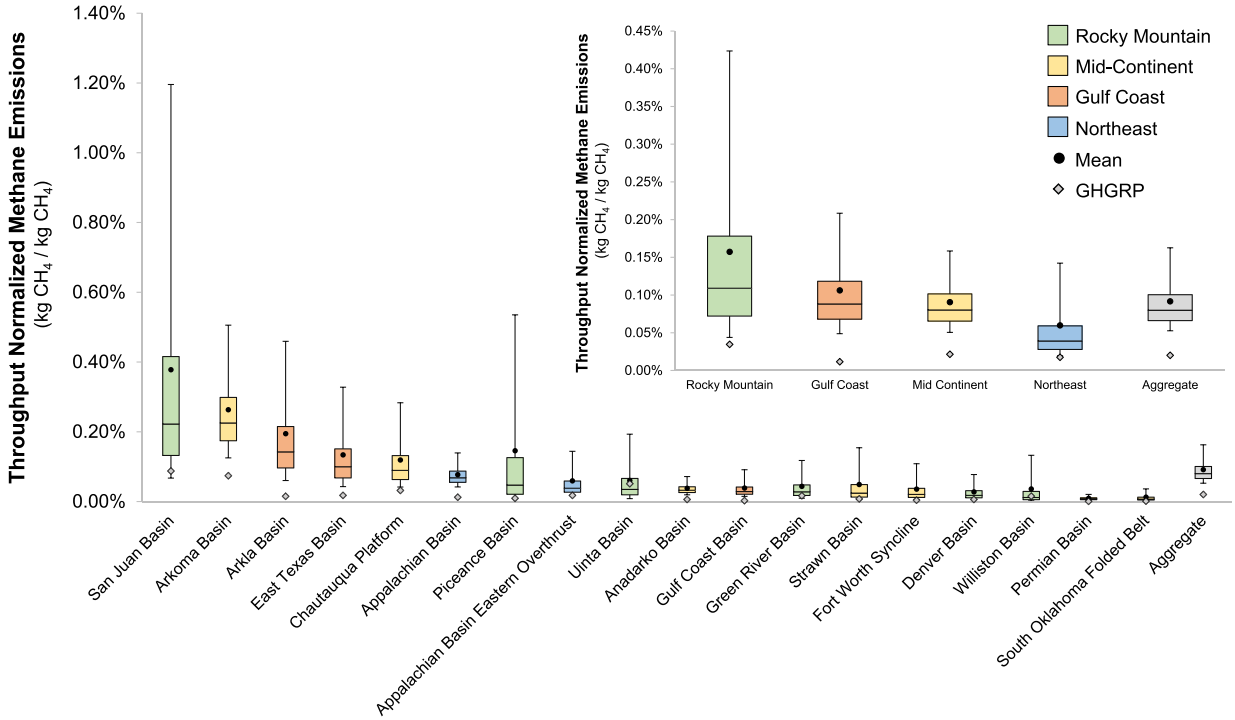

Figure 1. Throughput-normalized methane emissions (TNME) stratified by basin and NEMS regions. Error bars represent the 5th and 95th percentiles obtained via Monte Carlo simulation.

a stochastic subroutine was developed to parse venting plunger-lift well count reported by the GHGRP into constituent venting manual and automatic well counts. Liquids unloading event count for plunger-lift systems, reported by the GHGRP, are modeled as the linear combination of per-well venting frequency and venting well count for manual and automatic plunger-lift systems. Probability distributions for the venting frequency of manual plunger-lift and automatic plunger-lift systems were parametrized using data reported in Allen et al.,[20] and randomly sampled via Monte Carlo simulation. Manual and automatic plunger-lift venting well counts were determined via solving a system of conditional linear equations, using venting plunger-lift well count and event count reported by GHGRP as model constraints. In this approach, variability in per-well venting frequency for manual and automatic plunger systems is propagated into uncertainty in venting well counts. The fraction of wells that vent for liquids unloading, stratified by well type and liquids unloading system, was obtained by taking the ratio of county-level venting well count to county-level total well count sampled in the GHGRP. Additional information is provided in section S6 of the Supporting Information

API/ANGA Survey. The 2012 API/ANGA survey provides high-level data on natural gas production activities and equipment emission sources representative of industrial practices and contains information for 91 000 wells operated by over 20 companies.[38] API and ANGA report aggregate data for wells that vent for liquids unloading including well CD, TD, WD, and SP across five NEMS regions (Northeast, Gulf Coast, Mid-Continent, Southwest, and Rocky Mountain), two well types (conventional and unconventional), and two broad liquids unloading systems (nonplunger and plunger-lifts). In this work, data from the API/ANGA survey was used to characterize well SP, and fill data gaps may arise in the 2016 GHGRP data set. Triangular distributions were used to characterize well CD, TD, WD, and SP based on min, max, and expected values (venting well weighted average) using data reported from API/ANGA. Additional information is provided in section S10 in the Supporting Information

Data Limitations. In this work data from Allen et al. (2015) was used to model liquids unloading venting frequency instead of data reported to EPA's Greenhouse Gas Reporting Program (GHGRP) and was chosen for several reasons: (1) Allen et al. provides a stratification of venting episodes across manual and automatic plunger-lift systems. This level of stratification is not possible using GHGRP because only an aggregate plunger category is reported. (2) Allen et al. provides the requisite data necessary for developing probability distributions for unloading frequencies, thus allowing stochastic modeling of variability, a key research objective of this work. However, there are several limitations in using data from Allen et al. to model 2016 liquids unloading venting frequency. First, data from Allen et al. is reflective of liquids unloading event counts recorded in 2012 and, thus, is not congruent with the temporal window of the analysis (i.e., 2016 liquids unloading activities). This is important because changes in industry practices have resulted in a general decrease in venting frequency and unloading emissions at a national level over the 2012 to 2016 time-period. Second, during site selection, Allen et al. preferentially selected regions with substantive emissions from liquids unloading and the sampling of wells with high unloading frequencies. Despite these limitations, Allen et al. provides the best available data to meet the research objectives of this study. Additionally, it is important to note that Allen et al. is not the sole data source used for characterizing unloading activity. The 2016 GHGRP is used by the model to inform the fraction of wells that vent for liquids unloading in 2016, while Allen et al. is used to develop probability distributions for key parameters (venting duration, venting frequency) and split

DOI: 10.1021/acs.est.8b05546
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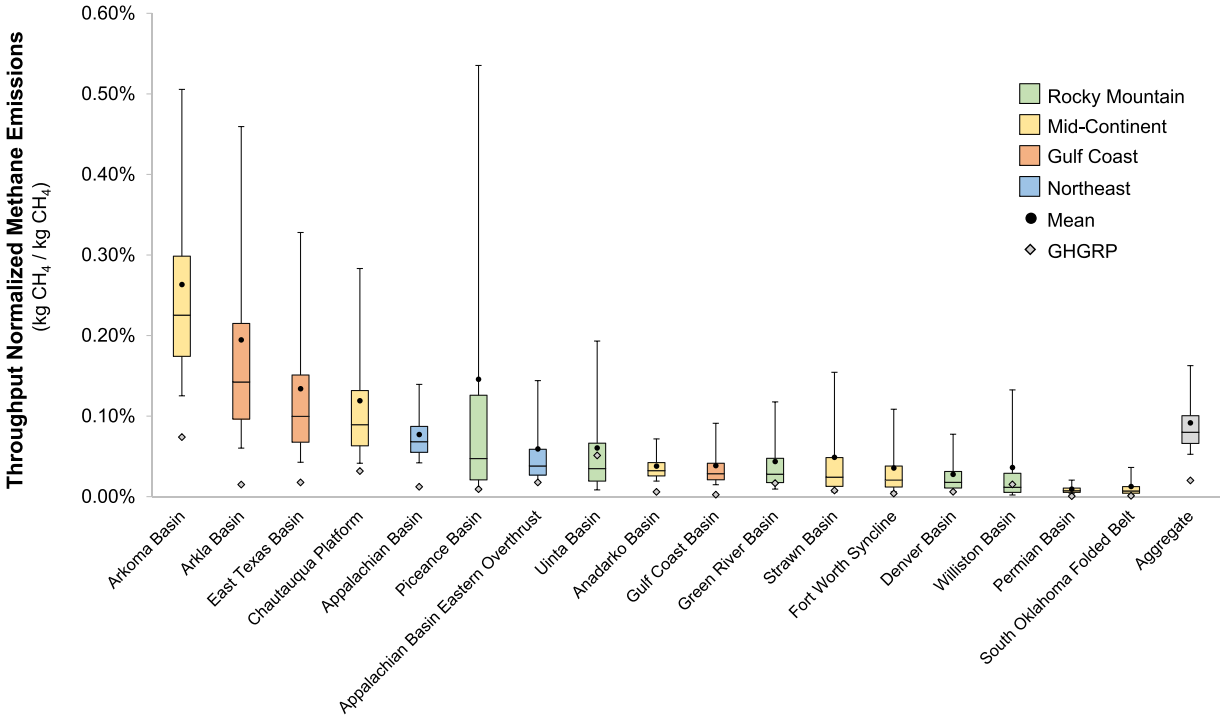

Figure 2. TNME stratified by basin (excluding San Juan). Error bars represent the 5th and 95th percentiles obtained via Monte Carlo simulation.

Table 1. Comparison between the 2016 GHGRP and This Work

Parameter	2016 GHGRP	NETL (This work)
Method for Calculating Emissions	GHGRP estimates methane emissions from liquids unloading using a combination of calculation methods 1 as well as calculation methods 2 & 3.	This work estimates methane emissions from liquids unloading using calculation methods 2 & 3. However, it is important to note that calculation methods 2 & 3 are used to estimate ~99% of methane emissions from liquids unloading in the 2016 GHGRP, thus this work is largely consistent with the GHGRP dataset.
Technological Characterization of Plunger-lift systems	The GHGRP characterizes wells that vent for liquids unloading into non-plunger or plunger-lift categories.	This work characterizes wells that vent for liquids unloading into non-plunger, manual-plunger lift, and automatic-plunger lift categories.
Uncertainty and Variability Quantification	The GHGRP uses average values for well parameters (casing diameter, tubing diameter. etc.) reported at the facility sub-basin level, and thus does not capture variability and uncertainty in emissions estimates.	This work uses Monte Carlo simulation to quantify variability and uncertainty in key parameters (casing diameter, tubing diameter, well depth, venting frequency, venting duration, etc). Data used to define the venting frequency for liquids unloading systems is obtained from Allen et al. 2015. However, it is important to note the relatively small sample size for the venting frequency of non-plunger (N=32), manual plunger-lift (N=50), and automatic plunger-lifts (N=25).
Well Coverage	The GHGRP reports the average number of wells that vent for liquids unloading, at the facility-level. However, the GHGRP is constrained by the reporting threshold (e.g. only sources that emit ~25,000 metric tons or more of carbon dioxide equivalent per year in the United States are required to report to the GHGRP). In total, 40,418 wells vent for liquid unloading across the 18 basins evaluated in this work.	This work uses data from drilling information (DI) desktop to quantify key flows from natural gas activities. Data from DI desktop is used due to its extensive coverage of natural gas and petroleum operations, and to overcome limitations imposed by the GHGRP reporting threshold. In this work, the mean number of wells that vent for liquids unloading across the 18 basins is ~50,393.

venting activity between nonplunger, manual, and automatic technologies.

It is important to note that CBM well characteristics tend to be dissimilar to other nonconventional well types due to differences in geomorphology and drilling practices (e.g., CBM wells are characteristically shallow and vertical, whereas hydraulically fractured wells are typically long and horizontal). However, as the majority of CBM wells are concentrated in a small number of counties in the San Juan basin, the county-level aggregation performed in this work curtails the extent that CBM well characteristics are blended with other unconventional sources.

DOI: 10.1021/acs.est.8b05546
Environ. Sci. Technol. 2019, 53, 4619−4629

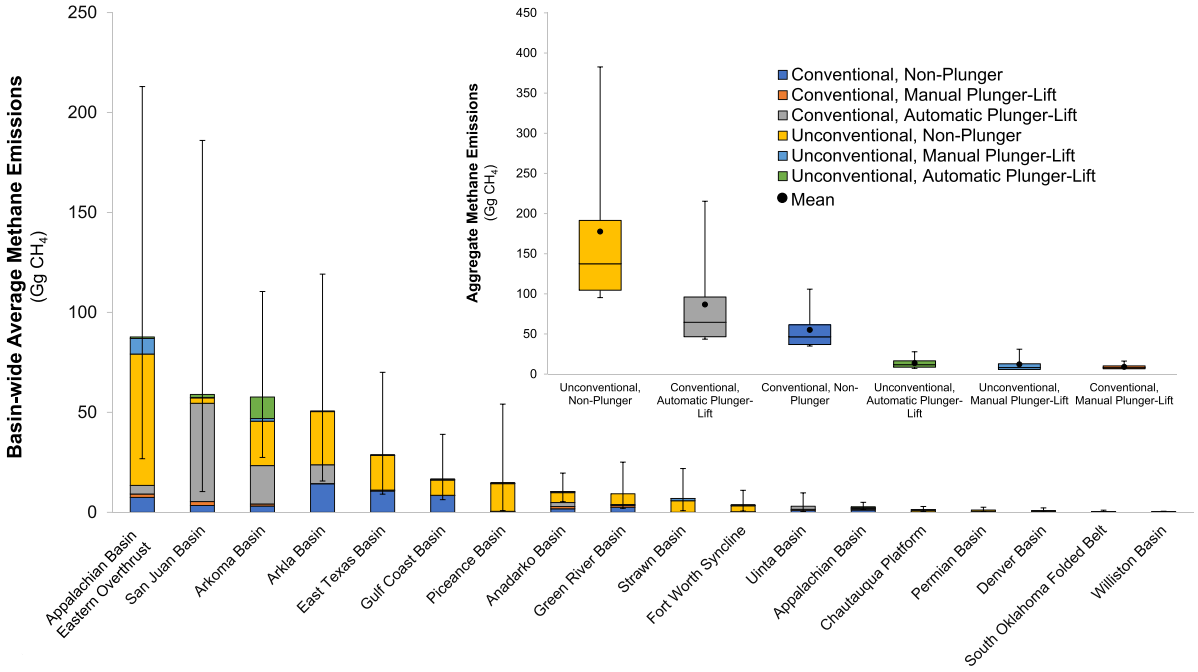

Figure 3. Contribution of liquids unloading scenarios to basin-wide average methane emissions. Error bars represent the 5th and 95th percentiles obtained via Monte Carlo simulation.

■ RESULTS AND DISCUSSION

Figure 1 plots the TNME for 18 natural gas basins and four NEMS regions in the United States and compares the results against a composite $TNME_{GHGRP}$ inferred from the 2016 GHGRP. $TNME_{GHGRP}$ are calculated via eq 1 and derived from several data-elements reported to the GHGRP, including (1) facility-level natural gas production, (2) salable oil production, and (3) methane emissions from liquids unloading, as well as an assumed volumetric concentration of methane in produced natural gas (V_{frac,CH_4}). Further, in calculating $TNME_{GHGRP}$, allocation factors are derived at the facility-level, additional information regarding data acquisition and modeling of $TNME_{GHGRP}$ is provided in section S11 of the Supporting Information. The results presented herein are derived based on allocated methane emissions; see the Supporting Information for additional details.

Analysis reveals that methane emissions from liquids unloading are highly variable both within and among basins. Mean TNME range from 0.0093% to 0.38% across basins, with the San Juan and Permian basins displaying the highest and lowest TNME, respectively. On average, nonplungers, manual, and automatic plunger-lift systems constitute 66%, 6.0%, and 28% of aggregate methane emissions over all basins. Further, conventional and unconventional venting wells represent, on average, 43% and 57% of cumulative liquids unloading methane emissions across all basins, respectively. Analysis at the NEMS level indicate that the Rocky Mountain region has the highest median TNME followed by the Gulf Coast, mid-continent, and northeast. The high TNME for the Rocky Mountain region is primarily driven by significant methane emissions from the Four Corners area. For ease of readability, the TNME for all basins, excluding San Juan, is provided in Figure 2. TNMEs constructed in this work are generally higher than composite $TNME_{GHGRP}$ inferred from the GHGRP and suggest that current bottom-up national emissions inventories underestimate methane emissions from liquids unloading. Key drivers for differences in the absolute methane emissions between this work and the GHGRP include: (i) differences in the analytic approach for calculating methane emissions, (ii) technological characterization of plunger-lift systems, (iii) deterministic versus stochastic modeling, and (iv) data inputs and coverage; see Table 1.

Figure 3 plots the contribution of each liquids unloading scenario to average basin-wide cumulative methane emissions. The top five natural gas basins ranked by average cumulative methane emissions are the Appalachian (Eastern Overthrust), San Juan, Arkoma, Arkla, and East Texas basins. The contribution of emissions by liquids unloading scenario is found to be exceedingly heterogeneous, with unconventional nonplunger systems responsible for over 75% of total CH_4 emissions in the Appalachian basin (Eastern overthrust), while conventional automatic plunger-lifts dominate the emissions profile for the San Juan basin, accounting for 83% of basin-wide methane emissions. Analysis at the component level indicates that methane emissions from liquids unloading are highly sensitive to venting frequency as well as casing diameter and tubing diameter. The venting frequency for automatic-plunger lift systems was found to follow a heavy tailed distribution (Weibull distribution), resulting in a skewed emissions distribution profile. Furthermore, due to their comparatively higher venting frequency, automatic plunger-lift systems were found to have significantly higher per-well methane emission rates relative to nonplunger or manual plunger-lift systems despite exhibiting a lower venting duration. On average, venting automatic plunger-lift systems constitute 28% of cumulative methane emissions from liquids unloading over all 18 basins in 2016, yet represent only ~0.43% of total natural gas well population. These results are consistent with Allen et al., which find that wells with high venting frequencies (>100 vents per well per year) account for the majority of emissions from liquids unloading. Furthermore, uncertainty in automatic venting well count had a nontrivial impact on methane emissions estimates and is a key source of emissions

DOI: 10.1021/acs.est.8b05546
Environ. Sci. Technol. 2019, 53, 4619−4629

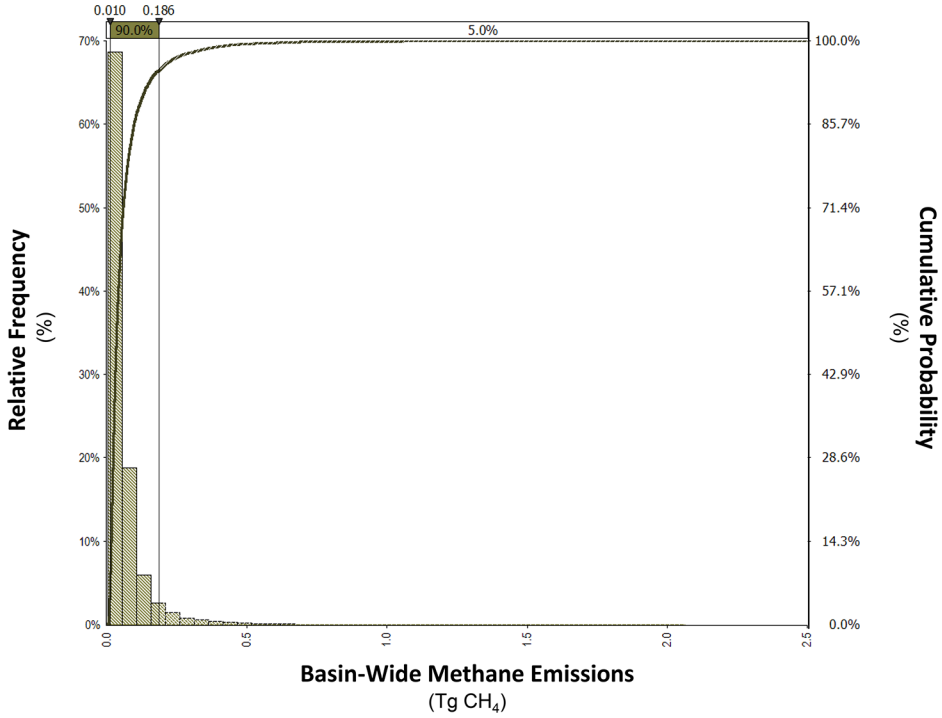

Figure 4. Frequency distribution and overlaid cumulative probability function for methane emissions from liquids unloading in the San Juan basin.

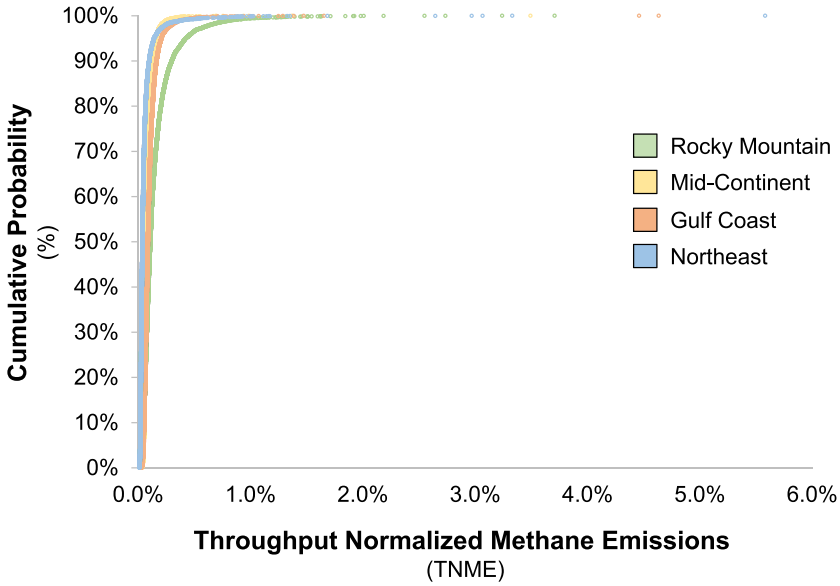

Figure 5. Cumulative distribution function: TNME by NEMS regions.

uncertainty. Although automatic plunger-lift systems constitute a disproportionate amount of methane emissions from liquids unloading, it is erroneous to conclude that automated plunger lifts are causally responsible for higher emissions. Automatic plunger-lifts may be preferentially installed on wells that require more frequent unloading due to well characteristics such as high liquids content, well age, etc. and not necessarily due to inherent properties of the unloading technology. Establishing causality between automatic plunger-lifts systems and higher emissions requires consideration of the counter-factual scenario, e.g., quantifying the emissions profile of the well absent (or prior to) the installation of automatic plunger-lift technology.

The frequency distribution and cumulative distribution function (CDF) for basin-wide methane emissions from liquids unloading in the San Juan basin are plotted in Figure 4. Cumulative methane emissions from the San Juan basin exhibit a heavy-tail distribution, indicating the presence of super-emitters, e.g., sites or activities that constitute a small fraction of the total population but account for a disproportionately large volume of total emissions. The relatively high number of venting automatic plunger-lift wells as well as the skewed emissions distribution from automatic-plunger systems drives the heavy tail distribution for the San Juan basin, resulting in comparatively higher methane emissions per unit produced natural gas (kg CH_4), relative

DOI: 10.1021/acs.est.8b05546
Environ. Sci. Technol. 2019, 53, 4619−4629

to other basins. The mean and median TNME for San Juan is 0.38% and 0.22%, respectively; divergence between mean and median estimates arises due to the positive skewness of the heavy-tail distribution. Figure 5 plots the CDF for TNME by NEMS regions. The results indicate that the Rocky Mountain region has a highly skewed CDF relative to other NEMS regions, due to the disproportionate influence of high-emitting sites in the San Juan basin. Furthermore, cumulative methane emissions from the Rocky Mountain region are on average ~4.0 times higher than those reported in the 2016 GHGRP. These findings corroborate similar studies that have (1) shown that methane emissions from fossil activities in the Four Corners region are likely under-estimated in emissions inventories (e.g., EDGAR v4.2 or GHGRP)[45] and (2) indicated that methane emissions in the Four Corners region, among other regions, follow a log-normal, heavy-tail distribution.[46]

While previous work has advocated that operational malfunctions and abnormal process conditions explain the existence of super-emitters in the natural gas supply chain,[32,47] the findings from this work show that super-emitters can arise naturally due to variability in underlying component processes. Specifically, randomly sampling from probability distributions for key parameters (see Table S1) can give rise to instantiations in which the superposition of parameter values leads to an extremum emissions rate. Additionally, in natural gas processes, such as liquids unloading, in which underlying parameter distributions are nonsymmetric and heavy-tailed, and the equations used for emissions prediction are nonlinear; the expected value from stochastic model simulation can be highly divergent from deterministic results. This is important for national emissions programs, which typically rely on point-estimates and deterministic modeling in developing bottom-up emissions inventories. Further, coarse technological aggregation often implemented by emissions monitoring programs to reduce the dimensionality of the reported data may obfuscate important emissions drivers. In the context of natural gas liquids unloading, automatic plunger-lift systems are found to have emissions signatures characteristic of super-emitters and are a key emissions source. As such, it is recommended that emissions monitoring programs report emissions from liquids unloading across nonplunger, manual plunger, and automatic plunger-lift categories. Additionally, bottom-up emission inventories can be improved via (1) the implementation of stochastic modeling, and (2) a robust characterization of the regional and temporal variability in liquids unloading venting frequency and venting duration disaggregated by unloading technology. In this work, methane emissions from liquids unloading were found to be highly variable, and on average are ~4.8 times higher than those inferred from the 2016 Greenhouse Gas Reporting Program. In the context of EPA's Greenhouse Gas Inventory (GHGI), which relies on GHGRP data for liquids unloading, a 4.8 factor increase would increase the 2016 liquids unloading emissions in EPA's 2018 GHGI from 133 to 638 kilotons per year. In turn, this would increase EPA's inventory of 2016 natural gas system emissions by 8% (from 6541 to 7046 kilotons of CH_4 per year). This scaling approximates the extent to which revised reporting and inventory methods could change the liquids unloading emissions in the GHGI; it does not account for year-to-year variability in unloading emissions and assumes that the basins outside the scope of this analysis have liquids unloading characteristics similar to the 18-basin aggregate.

This work presented a bottom-up, engineering-based, stochastic model that provided a deeper understanding of the methane emissions variability from liquids unloading in select natural gas production basins. This work was inspired in part by Schweitzke et al.'s 2017 study in the Fayetteville Shale, which provided a mechanistic understanding that bridged aerial measurements and production site activities.[9] The results presented herein are compelling in the context of other aerial measurement studies including Petron et al. and Karion et al. that observed high methane emission rates in the Colorado Denver-Julesburg Basin and Uintah County, Utah, respectively.[14,18] Additionally, this work replicates Allen et al.'s finding that automatic plunger lifts are a key contributor to liquids unloading emissions, but unlike Allen et al., our work concludes that reporting programs potentially understate total unloading emissions from liquids unloading.[20] This work demonstrates that when routine events are a function of multiple variables in a stochastic environment, the resulting emissions may be distributed in ways that are characteristic of super-emitters. We cannot rule out abnormal process events as an explanation for super-emitters[32] (as proposed by Zavala et al.) but have provided an alternative perspective that points to routine operations. In totality, these insights help rectify discrepancies between "top-down" and "bottom-up" studies and highlight the importance of technological disaggregation, uncertainty quantification, and regionalization in developing robust bottom-up estimates of methane emissions from high-emitting episodic sources.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b05546.

Additional details on experimental methods and analyses (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: (412)-386-7560; e-mail: james.littlefield@netl.doe.gov.

ORCID

James A. Littlefield: 0000-0002-2861-7438

Gregory Cooney: 0000-0001-5853-5102

Present Address

#Environmental Defense Fund, 2060 Broadway, Boulder, CO 80302, United States

Funding

This analysis was prepared by the Office of Fossil Energy for the United States Department of Energy (DOE), National Energy Technology Laboratory (NETL). This work was completed under DOE NETL contract no. DE-FE0025912. This funding supported contributions by G.G.Z., J.A.L., G.C., D.J.A., and T.J.S. The other authors received no compensation for their contributions to this work.

Notes

The authors declare no competing financial interest.

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■ ACKNOWLEDGMENTS

We graciously thank our colleagues Ambica Koushik Pegallapati, Tyler J. Hengen, Selina Roman-White, and Aranya

Venkatesh for their discussion and feedback on an earlier version of the manuscript. Additionally, we thank the two anonymous reviewers for their technical feedback, insightful comments, and suggestions. This study was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

■ DEDICATION

This paper is dedicated to the memory of Terri Lauderdale. Her unparalleled expertise on methane emissions research and reporting programs was a valuable contribution to this work and many other papers of national significance. Terri was a pleasure to work with and will be missed dearly.

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