Environmental Science & Technology

Open Access

pubs.acs.org/est

Article

Informing Methane Emissions Inventories Using Facility Aerial Measurements at Midstream Natural Gas Facilities

Jenna A. Brown, Matthew R. Harrison, Tecle Rufael, Selina A. Roman-White, Gregory B. Ross, Fiji C. George, and Daniel Zimmerle*

Cite This: https://doi.org/10.1021/acs.est.3c01321

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: Increased interest in greenhouse gas (GHG) emissions, including recent legislative action and voluntary programs, has increased attention on quantifying and ultimately reducing methane emissions from the natural gas supply chain. While inventories used for public or corporate GHG policies have traditionally utilized bottom-up (BU) methods to estimate emissions, the validity of such inventories has been questioned. Therefore, there is attention on utilizing full-facility measurements using airborne, satellite, or drone (top-down (TD)) techniques to inform, improve, or validate inventories. This study utilized full-facility estimates from two independent TD methods at 15 midstream natural gas facilities in the U.S.A., which were compared with a contemporaneous daily inventory assembled by the facility operator, employing comprehensive inventory methods. Estimates from the two TD methods statistically agreed in 2 of 28 paired measurements. Operator inventories, which included extensions to capture sources beyond regular inventory requirements and integration of local measurements, estimated significantly lower emissions than the TD estimates for 40 of 43 paired comparisons. Significant disagreement was observed at most facilities, both between the two TD methods and between the TD estimates and operator inventory. These findings have two implications. First, improving inventory estimates will require additional on-site or ground-based diagnostic screening and measurement of all sources. Second, the TD full-facility measurement methods need to undergo further testing, characterization, and potential improvement specifically tailored for complex midstream facilities.

KEYWORDS: *methane emissions, oil and gas, aerial measurements, measurement-informed inventories, bottom-up inventories, emission inventories, top-down measurements*

1. INTRODUCTION

Numerous U.S. domestic and international initiatives[1,2] have increased focus on greenhouse gas (GHG) emissions reduction. In the U.S., the National Climate Task Force has set groundbreaking goals to reduce domestic GHG emissions 50−52% below 2005 levels by 2030.[3] These efforts have focused attention on reducing methane emissions; methane is the second most common greenhouse gas after carbon dioxide (CO_2) and has a global warming potential (GWP) 86[4] times that of CO_2 on a 20-year basis.

The oil and gas (O&G) supply chain represents a substantial source of methane emissions, notably from the production, transport, and use of natural gas.[5−7] The natural gas supply chain is commonly divided into production, midstream, and distribution sectors; this study considers only facilities in the midstream sector; a related paper discusses similar work in production.[8] The midstream sector is commonly further divided into gathering and processing (G&P) and transmission and storage (T&S) segments. Midstream facilities are more complex and often have larger structures and buildings than production and distribution facilities.

Nearly all midstream facilities include gas compression equipment augmented by inlet and interstage separators that remove liquids from gas streams and tanks to store liquids. In many cases, the largest methane emitters at midstream facilities are compressors and compressor drivers.[5,9,10] Gas processing plants and some gathering compressor stations include additional processing equipment to upgrade gas to pipeline quality, such as dehydrators, acid gas removal units, and associated flares and tanks. Most midstream facilities also include miscellaneous equipment to support pipelines, fuel systems, and similar functions.[11] Finally, storage facilities also include wells and

Received: February 17, 2023
Revised: August 27, 2023
Accepted: August 29, 2023

wellhead equipment to store gas in underground reservoirs. This study includes eight transmission compressor stations, five gathering compressor stations, one gas processing plant, and one underground storage facility.

Multiple recent studies have characterized methane emissions from the O&G sector, primarily focusing on emissions in production basins. Broadly, these studies indicate a tendency for inventories to underestimate emissions relative to facility-scale or regional estimates of emissions.[7,12−18] The persistent disagreement between inventories and measurement-based estimates has placed additional focus on changing traditional inventory methods to improve emission estimates. New regulatory measures that include methane fees, coupled with public commitments by companies to environmental, social, and governance (ESG) programs, have also raised pressure on inventories to produce accurate and defensible results.

Recent U.S. legislative action includes the 2022 Inflation Reduction Act (IRA),[19] which funds a number of initiatives to reduce GHG emissions. As with most governmental initiatives, the IRA relies on the inventories as the basis for GHG accounting; for the U.S., these inventories are the Environmental Protection Agency (EPA) Greenhouse Gas Reporting Program (GHGRP)[20] and the Greenhouse Gas Inventory (GHGI).[5] Both are considered activity-based BU inventories and have been used as the basis for public policy decisions.[21] By 2024, the IRA requires the EPA to revise its GHGRP reporting methods and establish methane emission fees based on empirical data that "accurately reflect the total methane emissions".[19] The IRA also allows owners and operators of applicable facilities to submit empirical emissions data for compliance with the methane fees. As an indicator of broad support for direct measurements, in 2023, corporate investors filed 10 resolutions[22] calling for direct measurements instead of conventional inventory estimates of emissions. The combination of investor action and regulatory changes indicates broad support for the direct measurements of emissions. One promising approach in developing empirical data sets to enhance inventories at a facility-scale is to use multiscale measurements.[8] These approaches are broadly identified as measurement-informed inventory (MII) methods. While variations exist among proposed methods,[23−25] all typically include a three-step process:

1. Traditional BU inventory methods (activity counts × emission factors) and supplemental measurements are used to produce a reference estimate of emissions, herein termed the "operator estimated inventory (OEI)". In most cases, inventories are enhanced by comprehensively identifying all possible sources and utilizing per-source measurements, when available, in place of emission factors.

2. Independent TD estimates are performed using methods that measure full-facility emissions (either directly or by summing emissions from major sources). Since these methods have limitations, including method detection limits (MDLs) that vary with environmental conditions, some of these methods may not necessarily capture all emissions from a facility. Therefore, some of these methods are often supplemented with other information, such as per-source measurements and/or estimations based on BU emission factors, to produce a complete estimate of emissions or an adjusted TD estimate.

3. The OEI and TD estimates are aligned to the same time basis and compared to assess inventory completeness and inform changes to improve inventory results.

In this work, we will examine these three steps by focusing on the time period when measurements underlying the TD estimates were conducted, typically one working day (the time basis in step 3, above). This represents a subset of a complete MII process, which must also account for source intermittency and episodic events, like blowdowns or upset conditions, changes in the operational state of facilities, etc. By focusing on the measurement period, TD and BU methods can be compared with minimal complications, highlighting how information can be exchanged between these methods to improve emissions estimates. While theoretically estimating the same emissions, fundamental differences between TD and OEI methods complicate comparisons for midstream facilities. Three key differences must be considered.

First, the TD methods typically measure over short time periods, from seconds to hours. In contrast, OEI methods sum a set of point-source estimates, which are inherently based upon observations made over different time frames, from hourly logs of operating conditions to short-duration point measurements of individual sources. The averaging in inventory methods works well for long-duration (typically annual) emissions estimates but may not directly account for the specific conditions when TD methods were sampling.

Second, OEI methods rely on emission factors when measurements cannot be made. These factors typically originate from prior field studies conducted years earlier on facilities operated by multiple O&G operators. While well-performed studies include the full range of emissions from each source category, emission distributions are inherently averages across facility types, operating methods, failure modes, etc. Further, OEI methods use only the mean emission factor and therefore do not account for extremes in emission rates that may exist on any one facility or for any one source.

Third, midstream facilities frequently change operational state, primarily by starting, stopping, or changing load on compressors or processing equipment. These state changes drive substantial variation in emissions from the facility.[26] For example, a state simulation conducted on a midsized gathering compressor station could see methane emissions vary by 350% due solely to compressors changing operating mode (Supporting Information (SI) Section S-3.3). State changes may also occur frequently, often multiple times per day.

The work conducted for this study was part of a larger Quantification, Monitoring, Reporting, and Verification (QMRV) program.[27] The QMRV research and development (R&D) program includes modules for all sectors of the natural gas supply chain, and each module defines three phases:

- A baseline phase resulting in an TD-OEI comparison, as described above.
- An enhanced monitoring phase, where operators monitor emissions, estimate/update monthly OEIs, and monthly MIIs are computed.
- An end-of-project verification phase, when another TD-OEI facility comparison is done, and final MII analysis is completed.

Emissions data were collected at 15 facilities, dispersed across four states, and operated by six midstream companies. This work analyzes results from the completed baseline phase.

2. METHODS

All facilities in this study were measured as found, during a preplanned measurement period, typically one work day. Facilities were operated as if measurements were not occurring, with the measurement teams working around any on-site activities, including maintenance work and operational changes.

Reconciliation is a process whereby the TD estimates are compared with the OEI. This requires three methodological elements. Characterizing the TD measurement methods, estimating uncertainty, and adjusting TD estimates are required to fully estimate facility emissions.

2.1. Measurement Methods. Bridger Photonics gas mapping laser imaging detection and ranging (LiDAR) (Solution 1)[28] and SeekOps flux plane mass balance drone (Solution 2)[29] were contracted to provide TD estimates at all facilities.

Solution 1 utilizes a downward-looking laser system that sweeps perpendicularly across the direction of flight of an aircraft and uses differential absorption to compute path-integrated methane concentration (ppm-m) from the aircraft to the ground. This system can detect concentrated (point source) emissions that produce sufficient imaging contrast to separate the emission plume from background methane concentrations. Solution 1 utilizes wind data, particularly wind speed, obtained from nearby weather station(s) to compute an emission rate from the plume image (i.e., two-dimensional concentration map). This approach eliminates the need to install an anemometer in the facility. Solution 1 also collected visual imagery and superimposed plume data on the photos to provide context for the detections.

In this study, Solution 1 typically screened (hereafter termed an "overflight") each facility twice during 1 day, typically making multiple passes to complete each overflight. Each facility overflight takes less than 1 h, while individual plumes are scanned in a few seconds. Since each overflight has multiple passes, an individual emitter may be characterized by several plumes. If the emitter appears in more than one pass, the emissions from that source are averaged and included only once. Therefore, the estimated facility emissions for Solution 1 are the result of adding estimates from multiple distinct plumes into one overflight estimate. In this study, each overflight, typically separated by several hours, is treated as an independent estimate.

Solution 2 uses a flux plane method with concentration data from a miniature, tunable laser spectroscopy sensor on a drone platform. The flux plane methodology is thoroughly discussed in the literature.[30,31] Briefly, the drone-mounted sensor measures methane concentration while making multiple downwind passes through the facility's emission plume at different heights, typically from near ground level to above and outside the plume, i.e., above any methane enhancements from the equipment being measured. Concentration measurements, multiplied by the normal of the wind speed through the plane of flight, are integrated across the plane of flight to calculate the emission rate.

Solution 2 deploys a ground meteorological station to collect wind speed data, which is supplemented by directional corrections of the drone while in flight. Solution 2 typically flies multiple flight planes within the facility to estimate emissions from subsets of equipment; the total facility estimate is the sum of all subsets. While each flux plane is completed in 20−40 min, it takes several hours to collect a full-facility estimate when measuring multiple subsets of the facility.

2.2. Solution Uncertainty. Both TD methods utilize proprietary algorithms to translate measurements from onboard instruments into emissions estimates. Uncertainty of each estimate is a function of multiple input uncertainties, ranging from instrument uncertainty (typically small) to larger wind field and algorithmic uncertainties. Therefore, this study uses uncertainty estimates from the controlled testing of the integrated solution.

Controlled release data from studies by Bell et al.[32] and Corbett et al.[30] were used to create an uncertainty model for Solution 1 and Solution 2, respectively. See SI Sections S-2.3 and S-2.5.

When Solution 2 subdivides the facility to create partial-facility estimates, analysts may subtract an emission estimate from one flux plane estimate from another flux plane estimate to account for upwind emissions. Subtraction impacts the uncertainty of the estimates. Since reports from the vendor do not indicate when subtraction was used, uncertainty was not adjusted for this impact.

Both methods are dependent on environmental and atmospheric conditions, resulting in detection and quantification limits; see SI Section S-2. For example, methane in compressor driver exhaust ("methane slip") may be too dilute to be visible to Solution 1's imaging, or emissions may pool or recirculate near large compressor buildings, possibly complicating or distorting either solution's quantification estimates. The controlled testing used single point sources in known locations that had less complex configurations than at midstream facilities, which have multiple, potentially overlapping and/or intermittent, emission sources and large structures that produce complex near-field winds. Therefore, the uncertainty models used here should be considered minimum estimates of uncertainty; in field conditions at complex midstream facilities, uncertainty is likely larger.

Finally, the methods differ on how brief, small emission events (e.g., intermittent gas releases from pneumatic controllers) are quantified. Timing of these events is unknown. Assuming random timing, some portion of these emissions will be transported by the wind and increase methane concentrations seen by the Solution 2 sensor, increasing the Solution 2 estimates. The impact is unknown; in this study, we assume these emissions are sufficiently random and mixed that Solution 2 detects methane enhancements that are representative of mean emissions from these sources. In contrast, these emission events create insufficient plumes to be detected during a Solution 1 overflight and have minimal impact on Solution 1 estimates. Therefore, to compare the two solutions, we add an estimate of these emissions to Solution 1.

In contrast, large episodic emissions, such as compressor unit blowdowns (depressurization of a compressor), can be identified by on-site observers. Since neither method can accurately estimate emissions from these events, estimates made during large episodic events are discarded, see SI Section S-3.1.

2.3. Aligning TD and OEI Estimates. A series of adjustments are required to bring the OEI and TD estimates to the same temporal basis and to accommodate differences in the detected emitters.

OEI methods estimate emissions by identifying emission sources and estimating emissions by one of two methods: (1) estimating activity data for each source and multiplying by an appropriate emission factor (e.g., pneumatic controller emissions) or (2) using a source-level on-site measurement (e.g., component leaks). Sources were either known (e.g.,

https://doi.org/10.1021/acs.est.3c01321
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Table 1. Facility Information

facility characteristics					compressor state in meas. period		estimates (kg/h)	
facility ID	supply chain sector[a]	compressor driver type[b]	number of compressor units at facility	engine class[c]	units operating[d]	state change[e]	operator estimated inventory (OEI)	measurement emissions check (MEC)[f]
A	G&P	recip	5	4SLB	yes	no	93.4	263 [193−331]
B	G&P	turbine	3		yes	no	26.2	545 [403−724]
C	T&S	turbine	4		yes	no	23	141 [115−165]
D	G&P	recip	11	4SRB	yes	no	59.3	63 [52−80]
E	G&P	recip	15	4SLB	yes	yes	117	694 [574−818]
F	G&P	recip	12	4SLB	yes	no	95.6	172 [124−216]
G	T&S	turbine	5		yes	no	18.6	57 [46−72]
H	G&P	recip	10	4SLB	yes	yes	58.6	70 [52−88]
I	T&S	turbine	1		yes	no	0.85	41 [30−54]
J	T&S	turbine	1		no	no	2.14	17 [13−21]
K	T&S	recip	8	2SLB	no	no	6.4	79 [65−97]
L	T&S	recip	5	4SLB	yes	yes	97.3	866 [689−1144]
M	T&S	recip	6	2SLB	yes	no	21.8	75 [59−92]
N	T&S	turbine	2		yes	no	13.8	40 [31−53]
O	T&S	recip	8	2SLB	no	no	62.4	300 [230−383]

[a]Supply chain sector of the facility: G&P = gas processing, T&S = transmission and storage. [b]The type of prime mover driving the compressor(s) at the facility: Recip = reciprocating (piston) engine, Turbine = combustion turbine. [c]For facilities with reciprocating (piston) engines, code indicates the type of engine: 2SLB = two-stroke, lean-burn, 4SLB = four-stroke, lean-burn, 4SRB = four-stroke, rich-burn. [d]Yes if any compressor units were operating during the measurement period. [e]Yes if any compressors changed state during the measurement period. [f]The MEC is typically the average of one Solution 2 estimate and two adjusted Solution 1 estimates.

compressor vents, blowdown stacks, etc.) or discovered via leak surveys; optical gas imaging (OGI) surveys and a Hi-Flow sampler were used in this phase of the project. In this study, source-level measurements were made where possible (SI Section S-1); other emitters were estimated using emission factors. OGI surveys were conducted throughout the day, typically measuring a source once (with the exception of the compressor-dependent emitters discussed below); therefore, all emitters were assumed to emit at a constant rate throughout the measurement period.

As noted earlier, a change in the operating state of the facility may change emissions significantly. The primary factors driving emissions at a facility are the online status of compressors or processing units, the load on these units, and changes to process settings. Emissions at the facility may also be influenced, typically to a lesser degree, by other process equipment, such as gas upgrading equipment. Prior studies have indicated that emissions driven directly by the operating state of compressor units often dominate overall emissions from midstream facilities.[9,10] Therefore, the compressor operating state provides a useful surrogate for the facility state at midstream facilities and are, of course, correlated with the throughput of compressor stations.

To align the estimates, three steps were taken. First, operators were requested to calculate daily OEI estimates so that individual operating states could be extracted. Operators also provided a log of when compressor units were operating and/or pressurized. This allowed the OEI to be decomposed into one OEI estimate for each operating state. Note that OEI estimates are assumed constant for each operating state; this effectively averages estimates for variable emitters (e.g., intermittent pneumatics or dump valves on compressor interstage separators).

Second, an independent observer from the study team noted compressor states and the time of large episodic emissions, such as blowdowns, compressor starts, etc. While episodic emissions need to be tracked and included in long-duration total emissions, comparisons performed here exclude episodic events from OEI estimates. TD estimates made during these episodic events were eliminated from the resulting comparisons. This simplification has no impact on TD-OEI comparisons; TD methods cannot reliably measure total emissions from episodic events, and the same engineering calculations would be used to estimate these emissions for both OEI and TD estimates.

Third, TD estimates were compared to OEI estimates in the same compressor operating state. For Solution 1 overflights, comparable states were identifiable. Solution 2 was measured over longer periods, increasing the likelihood that operating state would change during measurements. This type of impact was seen at 3 of the 15 facilities. Since operating state changes may have created substantial variations in emissions, the comparison weighted the OEI estimates by the time in each state over the measurement period (SI Section S-2.6). Using a weighted average OEI assumes that the total estimate of emissions from Solution 2 effectively averages emissions in a manner similar to the weighted average OEI. The validity of this assumption is impossible to test and is indicative of the challenges of comparing TD estimates to complex time-averaged OEIs in any MII program.

In addition to temporal alignment, estimates need to be adjusted to represent, but not double-count, all emissions from the facility. Adjustments are method-specific. For example, Solution 1's method typically omits emitters below the method's MDL; these need to be added to the Solution 1 estimate. Additionally, a quality review indicated that Solution 1 did not consistently detect and quantify methane in compressor driver exhaust (compare SI Figures S-2 vs S-3). An expert panel was utilized to estimate whether Solution 1's estimate included combustion slip (SI Section S-2.2). Results indicated that Solution 1 detected exhaust plumes at 3 of 8 facilities where substantial combustion slip would be expected due to the type and size of compressor drivers on the facility. If Solution 1 did not detect exhaust at a facility, emissions from combustion slip

https://doi.org/10.1021/acs.est.3c01321
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

were estimated using recent stack tests, if available, or emission factors, if not.

In theory, Solution 2 captures emissions from all emitters upwind of the flux plane that were active within the transport time of the emissions. However, exceptions exist for flights that did not extend high enough to traverse exhaust plumes from compressor drivers (Facility C). Since the drone transects are fast, additional uncertainty exists for unsteady emission sources near the flux plane (e.g., intermittent gas pneumatics) or in cases where wind near a building may attenuate or recirculate emissions. Unfortunately, no data exist to quantify these uncertainties. Finally, as noted earlier, multiple flux planes within one facility require adjustments by Solution 2's analysts to avoid double-counting emissions. In this study, we assume these corrections were perfect, and no emissions were transported through two flux planes without correction (SI Section S-2.4).

The adjustment process outlined here describes the necessary TD adjustments for midstream MII methods to construct complete estimates of emissions. Similar methods are likely required for other sectors with highly variable operating states.

2.4. Calculating the Measurement Emissions Check (MEC). After the TD methods were adjusted to represent a full-facility estimate, all independent estimates were averaged to produce the MEC: typically one Solution 2 estimate and two adjusted Solution 1 estimates for each facility (see SI Section S-3.1 for exceptions). The MEC serves as a useful basis for plotting (X-axis in figures below) and for normalizing comparisons (Y-axis). However, it is not used for comparisons. When comparing between any pair of estimates, each comparison was done with one TD method individually to highlight differences between the TD methods.

3. RESULTS AND DISCUSSION

Results are presented as a series of full-facility emission comparisons, with all independent estimates brought to a comparable basis, as described in Section 2. For these comparisons, adjusted Solution 1 and Solution 2 estimates are used, compared to each other and to the OEI. In the following discussion, the reader should note that any mention of Solution 1 refers to the adjusted estimate. Table 1 provides information about all of the enrolled facilities.

3.1. Solution Comparison. While most MII efforts would deploy only one TD method, this study had access to two TD technologies to support additional analysis. The methods selected for the study both create facility estimates but use distinctly different measurement instruments and analysis algorithms. Since both were deployed contemporaneously (same day in most cases; see SI Section S-3.1), these estimates can be compared to better inform the uncertainty of both methods.

Figure 1 uses a Bland−Altman difference plot to compare the two TD estimates. This analysis plots the relative difference between two estimates against a common estimate of emission rate. The facility MEC was used both for the X-axis and as an average estimate to normalize relative differences. Each point represents one estimate at one facility. Error bars indicate a 95% confidence interval for each estimate; see SI Sections S-2.3 and S-2.5. For each facility, there are two independent Solution 1 estimates, completed at different times, which appear as points that are immediately above each other. As depicted in the figure, as the MEC increases, the difference between the two Solution 1 estimates becomes more pronounced.

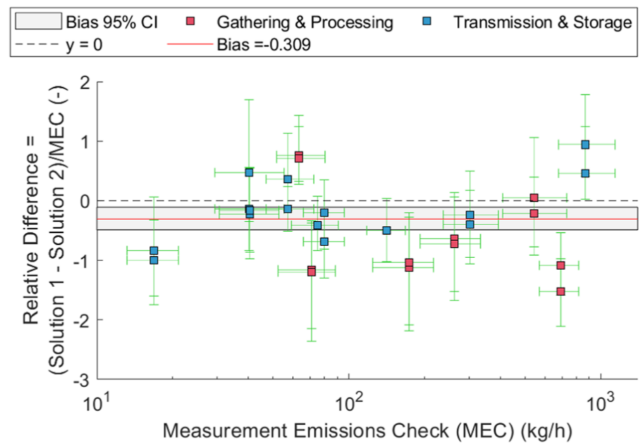


Figure 1. Horizontal axis is the MEC, an average of the TD estimates; the vertical axis is the relative difference between the adjusted solution 1 and solution 2 estimates for each facility. The gray box displays the 95% confidence interval over all facilities. The dashed line ($y = 0$) displays perfect agreement.

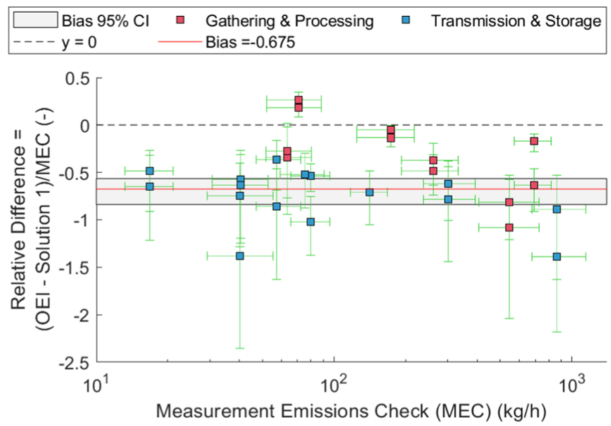
Aggregated across all facilities (gray box in figure), adjusted Solution 1 estimated emissions were 1,073 [387−1586] kg/h lower than Solution 2, a statistically significant difference of 31 [12−47%]. For pairwise comparisons at individual facilities, the two TD estimates statistically agree in 2 of 28 comparisons (Kolmogrov−Smirnov 2-sided, α = 0.05), with Solution 1 reporting emissions that were statistically lower than Solution 2 in 20 comparisons and higher in 6 comparisons. Results are compared using other methods in SI Section S-3.2.

Method disagreement could be compounded by changing the operating state during the measurement period. Eliminating the 3 facilities where the operating state changed during measurement increased the fraction of comparisons that agree by ≈3%, from 2 of 28 to 2 of 20, and decreased the fraction of comparisons that disagree by a similar amount; this is likely not a significant source of disagreement between methods. See Table 1, column "State Change" for the specific facilities.

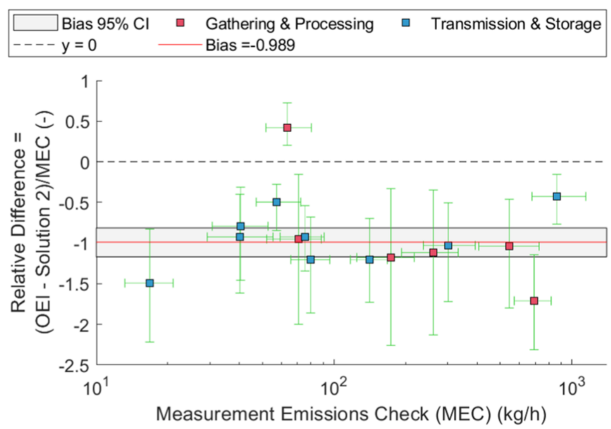
While emissions could change while the facility is in one operating state, operational knowledge indicates that large changes were unlikely. Therefore, since both methods measured when emissions were essentially stable, it is likely that the TD methods, in field conditions at midstream facilities, have uncertainties larger than indicated by controlled testing, likely due to multiple emission sources and complex wind fields at these facilities.

The above analysis of TD method uncertainty impacts the confidence in the TD estimation component of MII methods and offers guidance for calculating these results. Outside an R&D program, most MII programs would utilize only one TD estimate at a time. Therefore, uncertainty in the TD estimate would be inherently difficult to assess using one estimate at each facility. Inventories could be in or out of agreement with TD estimates due solely to uncharacterized uncertainty in the chosen TD method. Further, most reporting programs report only mean values without uncertainty and have no systematic method to capture uncertainty. Taking the current study as a test case, had all 15 facilities been part of a single report, the aggregate uncertainty in the TD methods, without reference to any inventory methods, would be at least 31%—the difference in the mean estimate of total emissions between the two TD methods.

https://doi.org/10.1021/acs.est.3c01321
Environ. Sci. Technol. XXXX, XXX, XXX−XXX



(a) Comparison of the OEI to the *adjusted* Solution 1. Where two points are vertically aligned, each represents a Solution 1 overflight, typically two per facility.



(b) Comparison the OEI to Solution 2

Figure 2. Comparison of the BU inventory (OEI) to each of the two TD methods: Panel (a) for adjusted solution 1 and panel (b) for solution 2. The horizontal axis is the MEC, an average of the TD estimates; the vertical axis is the relative difference between the OEI and adjusted TD estimate for each facility. The gray box displays the 95% confidence interval over all facilities. The dashed line ($y = 0$) displays perfect agreement.

These results indicate a need for better-controlled release and field testing of these methods at complex facilities to better characterize TD method uncertainties and a need for MII protocols to consider further assessment and reporting of TD uncertainty.

3.2. OEI Solution Comparison. The second analysis of importance is the comparison between the OEI i.e., the BU inventory and TD estimates for each facility and for all facilities aggregated together. This comparison uses the adjusted full-facility estimate from each TD method compared to the calculated OEI.

An OEI was estimated for each of the 15 facilities using traditional inventory methods augmented with supplemental sources, emission factors from recent studies, and other modifications to ensure the inventory captured all known sources (see the Emission Calculation Guidance Tool attached to the SI). The guidance tool informs how to augment routine emission factors with direct measurements for significant sources. For example, stack tests were used to directly measure methane slip in combustion exhaust in as-found conditions.

Therefore, the OEI process used in this study is among the most robust used in any program. Figure 2 illustrates the difference between the OEI and the TD estimates using the same plot format as Figure 1.

When comparing OEI to TD methods, it is important to note that OEI estimates have no stated uncertainty. Therefore, the uncertainty in all comparisons is exclusively from the uncertainty estimate of the TD method. For both TD methods, the operator's inventory is significantly lower than either of the TD methods at the vast majority of facilities; the OEI is statistically higher than Solution 2 in 1 of 15 comparisons and statistically higher than Solution 1 in 2 of 28 comparisons. Although TD estimates may have higher uncertainty than shown, logical increases in uncertainty are unlikely to close the gap between the TD and OEI estimates.

Given the diversity of reporters and facility types, the significant disagreement between BU inventories and TD estimates indicates systematic under-reporting in the OEI and/or issues in the TD measurement. Disagreements are neither company-specific nor facility type-specific.

The intent of the MII process is to utilize measurement-based estimates to improve the inventory process for a specific facility (or a small group of facilities) over results using traditional inventory methods. If successful, the improved inventory process would then be used to accurately estimate emissions for each of the facilities' operating states, over extended periods, the result needed for regulatory reporting, voluntary initiatives, and similar purposes. The question, then, is how do TD measurements inform inventories? In this study, the TD/BU

https://doi.org/10.1021/acs.est.3c01321
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

disagreement could originate from three potential causes (or any combination of the three):

1. Known sources are systematically underestimated in BU inventories. Midstream facility emissions are primarily dominated by a limited number of well-known sources (e.g., leakage through large valves, venting from compressor seals, etc.).[9,10,33] All such sources are included in the QMRV OEI calculation, using on-site measurements wherever possible or best-in-class emission factors when measurement was not possible. Therefore, for known sources to drive the TD/BU disagreement, both measurements and/or emission factors for a large number of key midstream sources would need to be systematically underestimated relative to common field conditions.

   While there is little evidence for systematic low bias, it cannot be ruled out. Some sources at facilities are difficult to measure, due to the size of equipment (e.g., a large diameter blowdown stack), accessibility of an emission location, or safety issues. For on-site measurements, the largest emitters often prove the hardest to measure.[10] Systematic errors in these measurements are possible and could contribute to the TD/BU disagreement. BU emission factors are typically developed from the results of field studies, and any given facility may have a different emissions profile than the field study. While this should not lead to systematic bias, bias cannot be ruled out. Additional on-site measurements and/or continuous metering of some sources may be required to characterize major emission sources across multiple operating states.
2. Uninventoried, large emission sources exist on many facilities. One emitter of this type was discovered on one facility during TD measurements (a leak in a fuel gas system, included in data presented here and corrected the day after identification). No similar large sources were found in the other 14 facilities. Therefore, if large emitters were to explain the gap between the TD estimates, multiple such sources must exist in most facilities, and these sources must have unknown characteristics that made them undetectable by Solution 1 overflights. While there is no evidence for this type of large, systematic emission source on midstream facilities, it cannot be ruled out. Per-source screening and measurements would likely be required to either identify these sources or increase confidence that they do not exist.
3. TD estimates are biased high, and this bias is systematic for this type of facility. As with the other potential causes, there is little evidence of systematic high bias in TD methods, in part because these methods have not been extensively tested on complex facilities like compressor stations under controlled conditions. Current controlled testing was performed at near-ideal conditions—single source, no nearby structures, etc. Winds may recirculate emissions near large structures (where both TD methods estimate emissions), or multiple nearby emission sources may complicate rate recovery algorithms.

Given the size of the TD/BU disagreement, all three of these causes deserve evaluation. The above analysis indicates that, for midstream facilities, an "informed" inventory will likely require a more comprehensive program than simply performing periodic TD measurements at a facility; full-facility estimates need to be augmented with additional, on-site diagnostic work and measurements. For example, many vents or other sources on midstream facilities could be metered and monitored over extended periods and compared to TD estimates.

3.3. Extending to Long-Duration Estimates.

The ultimate goal of the MII is to establish total facility emissions over extended periods; monthly or annual estimates are common. All analyses above consider the simplified case where (nearly) contemporaneous TD estimates were compared to an OEI estimate made for the facility. This comparison was typically limited to emissions measured in a single operating state; even when an operating state changed during the day (3 facilities), full estimates in each state were not possible. Emissions in operating states that were not active during measurements were not characterized by TD estimates nor measured by on-site methods. For example, a single compressor unit may be operating, pressurized but not operating, or depressurized. In each mode, different valves are open, and shaft or rod packing seals operate differently. Considering only compressors at a typical station, emissions may vary by 2.5−3.5 times due solely to changes in operating mode (see simulation in SI Section S-3.3).

As a result of state changes, MII methods must estimate emissions from operating conditions that were inactive during TD surveys or design TD surveys to better study the facility in all operating conditions. This implies that single snapshot TD estimates are unlikely to replace annual BU inventory methods for midstream facilities until additional testing is completed and that midstream facilities will require high-quality logging of operating state, per-unit and per-source emission factors, and/or per-source measurements for the foreseeable future. This can be in the form of advanced monitoring and tracking systems of a facility's operational states, supplemented by identifying, measuring, and including any unplanned emissions.

4. IMPLICATIONS

Inventories are an important tool for making policy and industrial decisions, and there must be confidence that these inventories are accurate. MII methods suggest that inventories can be verified by TD estimates that capture all emissions at a facility.[34] Results from this study confirm this reconciliation may be possible but indicate several nuances that must be considered when using TD estimates to verify inventories at midstream facilities. First, for the diverse set of facilities considered here, a simple comparison of TD estimates to a contemporaneous BU inventory yields the unsatisfying result that these estimates disagree with little guidance on how to eliminate the disagreement. Therefore, measurement-informed inventories will likely require both full-facility TD estimates and a battery of diagnostic measurements or monitoring on-site—both to identify causes of disagreement between TD and BU estimates made in the same operating state and to capture operating states not included in the TD estimates.

Second, in this study, the TD/BU disagreement is systematic: at 40 of 43 TD-OEI comparisons, the TD estimates are statistically higher than the contemporaneous inventory. For midstream facilities, there is no ready explanation of why this disagreement exists. While per-source measurements at facilities may under-report emissions due to challenges mentioned earlier, it is unlikely that these issues would explain the TD/BU disagreements seen in the study; at 35 of 43 comparisons, there is a disagreement by more than a factor of 2. All known large sources are included in the enhanced inventory process used here, and additional large sources were not identified by the source-locating TD method (Solution 1) in sufficient quantities

https://doi.org/10.1021/acs.est.3c01321
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

and sizes to account for the difference. Therefore, the results of this study indicate the need for more extensive on-site identification and measurement of midstream sources and/or more complete and representative emission factor data, coupled with better characterization of the TD method uncertainty for midstream facilities.

■ ASSOCIATED CONTENT

Supporting Information

The following files are available free of charge. The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.3c01321.

Emissions calculation guidance tool: contains improved calculation methodology for emission sources (XLSX)

Additional details on facilities; solution methods and uncertainty, and state simulation (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Daniel Zimmerle − *Energy Institute, Colorado State University, Fort Collins, Colorado 80524, United States;* orcid.org/0000-0003-2832-048X; Email: dan.zimmerle@colostate.edu

Authors

Jenna A. Brown − *Department of Mechanical Engineering, Colorado State University, Fort Collins, Colorado 80524, United States;* orcid.org/0000-0002-0518-7869

Matthew R. Harrison − *SLR International Corp., Houston, Texas 77036, United States*

Tecle Rufael − *SLR International Corp., Houston, Texas 77036, United States*

Selina A. Roman-White − *Cheniere Energy Inc., Houston, Texas 77002, United States*

Gregory B. Ross − *Cheniere Energy Inc., Houston, Texas 77002, United States*

Fiji C. George − *Cheniere Energy Inc., Houston, Texas 77002, United States;* orcid.org/0000-0001-8819-0838

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.3c01321

Funding

This work was funded by Cheniere Energy, Inc.

Notes

The authors declare the following competing financial interest(s): F.C.G., G.R., and S.R.W. are employees of Cheniere Energy Inc. D.Z. has current or recent research support from Cheniere and other natural gas operators, the US Department of Energy, and state and national regulators and has worked as a consultant for SLR International recently. SLR International performs work for Cheniere, other oil and gas industry clients, academic institutions, and industry research organizations.

■ ACKNOWLEDGMENTS

The authors acknowledge SLR for managing the field work and organizing logistics. The authors acknowledge partner operators who provided site access, operational data, and any other information needed for analysis.

■ REFERENCES

(1) Programme, U. N. E. Oil and Gas Methane Partnership: Reconciliation and Uncer-tainty in Methane Emissions Estimates for OGMP2.0.

(2) Environment, U. N. International Methane Emissions Observatory (IMEO), 2017. http://www.unep.org/explore-topics/energy/what-we-do/imeo. Wed, 10/11/2017 13:01.

(3) National Climate Task Force, 2021. https://www.whitehouse.gov/climate/.

(4) Myhre, G.; Shindell, D.; Bréon, F.-M.; Collins, W.; Fuglestvedt, J.; Huang, J.; Koch, D.; Lamarque, J.-F.; Lee, D.; Mendoza, B.; Nakajima, T.; Robock, A.; Stephens, G.; Zhang, H.; Aamaas, B.; Boucher, O.; Dalsøren, S. B.; Daniel, J. S.; Forster, P.; Granier, C.; Haigh, J.; Hodnebrog, Ø.; Kaplan, J. O.; Marston, G.; Nielsen, C. J.; O'Neill, B. C.; Peters, G. P.; Pongratz, J.; Ramaswamy, V.; Roth, R.; Rotstayn, L.; Smith, S. J.; Stevenson, D.; Vernier, J.-P.; Wild, O.; Young, P.; Jacob, D.; Ravishankara, A. R.; Shine, K. *Anthropogenic and Natural Radiative Forcing (Chapter 8)*, Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, 2013.

(5) E. Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2020, 2022. https://www.epa.gov/ghgemissions/draft-inventory-us-greenhouse-gas-emissions and sinks-1990-2020.

(6) Littlefield, J. A.; Marriott, J.; Schively, G. A.; Skone, T. J. Synthesis of Recent Ground-Level Methane Emission Measurements from the U.S. Natural Gas Supply Chain. *J. Cleaner Prod.* **2017**, *148*, 118−126, DOI: 10.1016/j.jclepro.2017.01.101.

(7) Alvarez, R. A.; Zavala-Araiza, D.; Lyon, D. R.; Allen, D. T.; Barkley, Z. R.; Brandt, A. R.; Davis, K. J.; Herndon, S. C.; Jacob, D. J.; Karion, A.; Kort, E. A.; Lamb, B. K.; Lauvaux, T.; Maasakkers, J. D.; Marchese, A. J.; Omara, M.; Pacala, S. W.; Peischl, J.; Robinson, A. L.; Shepson, P. B.; Sweeney, C.; Townsend-Small, A.; Wofsy, S. C.; Hamburg, S. P. Assessment of Methane Emissions from the U.S. Oil and Gas Supply Chain. *Science* **2018**, No. eaar7204, DOI: 10.1126/science.aar7204.

(8) Wang, J. L.; Daniels, W. S.; Hammerling, D. M.; Harrison, M.; Burmaster, K.; George, F. C.; Ravikumar, A. P. Multiscale Methane Measurements at Oil and Gas Facilities Reveal Necessary Frameworks for Improved Emissions Accounting. *Environ. Sci. Technol.* **2022**, *56*, 14743−14752.

(9) Zimmerle, D. J.; Williams, L. L.; Vaughn, T. L.; Quinn, C.; Subramanian, R.; Duggan, G. P.; Willson, B.; Opsomer, J. D.; Marchese, A. J.; Martinez, D. M.; Robinson, A. L. Methane Emissions from the Natural Gas Transmission and Storage System in the United States. *Environ. Sci. Technol.* **2015**, *49*, 9374−9383.

(10) Zimmerle, D.; Vaughn, T.; Luck, B.; Lauderdale, T.; Keen, K.; Harrison, M.; Marchese, A.; Williams, L.; Allen, D. Methane Emissions from Gathering Compressor Stations in the U.S. *Environ. Sci. Technol.* **2020**, *54*, 7552−7561.

(11) eCFR:: 40 CFR Part 98 − Mandatory Greenhouse Gas Reporting, 2009. https://www.ecfr.gov/current/title-40/chapter-I/subchapter-C/part-98.

(12) Brandt, A. R.; Heath, G. A.; Kort, E. A.; O'Sullivan, F.; Petron, G.; Jordaan, S. M.; Tans, P.; Wilcox, J.; Gopstein, A. M.; Arent, D.; Wofsy, S.; Brown, N. J.; Bradley, R.; Stucky, G. D.; Eardley, D.; Harriss, R. Methane Leaks from North American Natural Gas Systems. *Science* **2014**, *343*, 733−735.

(13) Pétron, G.; Karion, A.; Sweeney, C.; Miller, B. R.; Montzka, S. A.; Frost, G. J.; Trainer, M.; Tans, P.; Andrews, A.; Kofler, J.; Helmig, D.; Guenther, D.; Dlugokencky, E.; Lang, P.; Newberger, T.; Wolter, S.; Hall, B.; Novelli, P.; Brewer, A.; Conley, S.; Hardesty, M.; Banta, R.; White, A.; Noone, D.; Wolfe, D.; Schnell, R. 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.

(14) Rutherford, J.; Sherwin, E.; Ravikumar, A. P.; Heath, G. A.; Englander, J.; Cooley, D.; Lyon, D.; Omara, M.; Langfitt, Q.; Brandt, A. R. Closing the Methane Gap in US Oil Anad Natural Gas Production Emissions Inventories. *Nat. Commun.* **2021**, No. 4715, DOI: 10.1038/s41467-021-25017-4.

(15) Lyon, D. R.; Zavala-Araiza, D.; Alvarez, R. A.; Harriss, R.; Palacios, V.; Lan, X.; Talbot, R.; Lavoie, T.; Shepson, P.; Yacovitch, T. I.; Herndon, S. C.; Marchese, A. J.; Zimmerle, D.; Robinson, A. L.; Hamburg, S. P. Constructing a Spatially Resolved Methane Emission Inventory for the Barnett Shale Region. *Environ. Sci. Technol.* **2015**, *49*, 8147−8157.

(16) Vaughn, T. L.; Bell, C. S.; Pickering, C. K.; Schwietzke, S.; Heath, G. A.; Pétron, G.; Zimmerle, D. J.; Schnell, R. C.; Nummedal, D. Temporal Variability Largely Explains Top-down/Bottom-up Difference in Methane Emission Estimates from a Natural Gas Production Region. *Proc. Natl. Acad. Sci. U.S.A.* **2018**, *115*, 11712−11717.

(17) Cusworth, D. H.; Duren, R. M.; Yadav, V.; Thorpe, A. K.; Verhulst, K.; Sander, S.; Hopkins, F.; Rafiq, T.; Miller, C. E. Synthesis of Methane Observations Across Scales: Strategies for Deploying a Multitiered Observing Network. *Geophys. Res. Lett.* **2020**, *47*, No. e2020GL087869, DOI: 10.1029/2020GL087869.

(18) Barkley, Z. R.; Davis, K. J.; Feng, S.; Cui, Y. Y.; Fried, A.; Weibring, P.; Richter, D.; Walega, J. G.; Miller, S. M.; Eckl, M.; Roiger, A.; Fiehn, A.; Kostinek, J. Analysis of Oil and Gas Ethane and Methane Emissions in the Southcentral and Eastern United States Using Four Seasons of Continuous Aircraft Ethane Measurements. *J. Geophys. Res.: Atmos.* **2021**, *126*, No. e2020JD034194, DOI: 10.1029/2020JD034194.

(19) Ramseur, J. Inflation Reduction Act Methane Emissions Charge: In Brief, 2022. https://crsreports.congress.gov/product/pdf/R/R47206.

(20) US EPA, OAR. Greenhouse Gas Reporting Program (GHGRP), 2014. https://www.epa.gov/ghgreporting.

(21) Improving Characterization of Anthropogenic Methane Emissions in the United States. In *National Academies of Sciences, Engineering, and Medicine*; National Academies Press: Washington, D.C., 2018; p 24987.

(22) Welsh, H.; Passoff, M. *Proxy Preview 2023*, 2023.

(23) Veritas. 2021, https://www.gti.energy/veritas-a-gti-methane-emissions-measurement-and-verification-initiative/.

(24) Oil and Gas Methane Partnership (OGMP) 2.0 Framework. https://www.ccacoalition.org/en/resources/oil-and-gas-methane-partnership-ogmp-20-framework.

(25) MIQ. https://miq.org/.

(26) Nathan, B. J.; Golston, L. M.; O'Brien, A. S.; Ross, K.; Harrison, W. A.; Tao, L.; Lary, D. J.; Johnson, D. R.; Covington, A. N.; Clark, N. N.; Zondlo, M. A. Near-Field Characterization of Methane Emission Variability from a Compressor Station Using a Model Aircraft. *Environ. Sci. Technol.* **2015**, *49*, 7896−7903.

(27) A Closer Look at Quan, Monitoring, Reporting And, 2022. https://www.cheniere.com/our-responsibility/climate/closer-look-qmrv.

(28) Bridger, P. Gas Mapping LiDAR for Methane Detection. https://www.bridgerphotonics.com/gas-mapping-lidar-for-aerial-methane-detection.

(29) SeekOps, Technology - SeekOps. https://seekops.com/technology/.

(30) Corbett, A.; Smith, B. A Study of a Miniature TDLAS System Onboard Two Unmanned Aircraft to Independently Quantify Methane Emissions from Oil and Gas Production Assets and Other Industrial Emitters. *Atmosphere* **2022**, *13*, No. 804.

(31) Rella, C. W.; Tsai, T. R.; Botkin, C. G.; Crosson, E. R.; Steele, D. Measuring Emissions from Oil and Natural Gas Well Pads Using the Mobile Flux Plane Technique. *Environ. Sci. Technol.* **2015**, *49*, 4742−4748.

(32) Bell, C.; Rutherford, J.; Brandt, A.; Sherwin, E.; Vaughn, T.; Zimmerle, D. Single-Blind Determination of Methane Detection Limits and Quantification Accuracy Using Aircraft-Based LiDAR. *Elem.: Sci. Anth.* **2022**, *10*, No. 00080.

(33) Marchese, A. J.; Vaughn, T. L.; Zimmerle, D. J.; Martinez, D. M.; Williams, L. L.; Robinson, A. L.; Mitchell, A. L.; Subramanian, R.; Tkacik, D. S.; Roscioli, J. R.; Herndon, S. C. Methane Emissions from United States Natural Gas Gathering and Processing. *Environ. Sci. Technol.* **2015**, *49*, 10718−10727.

(34) Heath, G.; Warner, E.; Steinberg, D.; Brandt, A. *Estimating U.S. Methane Emissions from the Natural Gas Supply Chain. Approaches, Uncertainties, Current Estimates, and Future Studies*, 2015; p 1226158.