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A Methane Emission Estimation Tool (MEET) for predictions of emissions from upstream oil and gas well sites with fine scale temporal and spatial resolution: Model structure and applications

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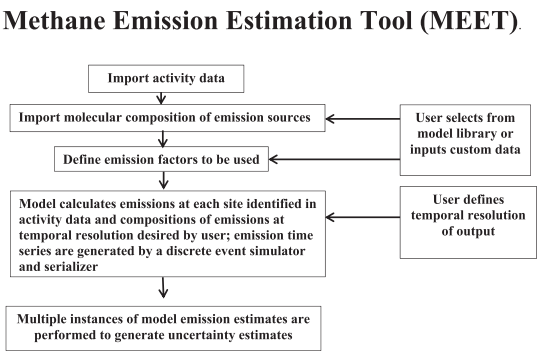
HIGHLIGHTS

- Emissions from oil and gas well sites exhibit considerable temporal variability.
- Short duration observations should be compared with fine time resolution inventories.
- Emission rates in fine time resolution inventories have multi-modal distributions.

GRAPHICAL ABSTRACT



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ABSTRACT

In comparing observation based methane emission estimates for oil and gas well sites to routine emissions reported in inventories, the time scale of the measurement should match the time scale over which the inventoried emissions are estimated. Since many measurements are of relatively short duration (seconds to hours), a tool is needed to estimate emissions over these time scales rather than the annual totals reported in most emission inventories. This work presents a tool for estimating routine emissions from oil and gas well sites at multiple time scales; emissions at well sites vary over time due to changes in oil and gas production rates, operating practices and operational modes at the sites. Distributions of routine emissions (expected and inventoried) from well sites are generally skewed, and the nature and degree to which the distributions are skewed depends on the time scales over which emissions are aggregated. Abnormal emissions can create additional skew in these distributions. At very short time scales (emissions aggregated over 1 min) case study distributions presented in this work are both skewed and bimodal, with the modes depending on whether liquid storage tanks are flashing at the time of the measurement and whether abnormal emissions are occurring. At longer time scales (emissions aggregated over 1 day) distributions of routine emissions simulated in this work can have multiple modes if short duration, high emission rate events, such as liquid unloadings or large abnormal emissions, occur at the site. Multiple applications of the methane emission estimation tool (MEET), developed in this work, are presented. These results emphasize the importance of developing detailed emission inventories, which incorporate operational data, when comparing measurements to routine emissions. The model described in this work supports such comparisons and is freely available.

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1. Introduction

Emissions of methane from sources in the oil and gas supply chain in the United States are reported in a number of inventories. These inventories provide estimates of methane emissions with varying degrees of spatial and temporal resolution. For example, the Greenhouse Gas Inventory (GHGI) for the United States, assembled by the U.S. Environmental Protection Agency (EPA) aggregates annual emissions of methane into broad oil and gas production regions (U.S. Environmental Protection Agency, 2021). The EPA Greenhouse Gas Reporting Program (GHGRP) also reports annual emissions of methane, but provides more detailed spatial resolution of some emissions (US Environmental Protection Agency, 2021). Recently, several teams of investigators have assembled methane emission inventories from oil and gas operations in the United States with much finer scale spatial and temporal resolution than is available in the GHGI or the GHGRP. Maasakkers et al. (2016) reported data from the 2012 US EPA Greenhouse Gas Inventory on a 0.1° by 0.1° spatial grid with a monthly temporal resolution. Allen et al. (2017) and Cardoso-Saldaña et al. (2019) reported hourly estimates of methane emissions for the Barnett Shale and Eagle Ford oil and gas production regions with resolution at individual well sites, gathering sites, processing sites and transmission sites, and with emissions attributed to specific types of equipment at well sites. Vaughn et al. (2018) produced hourly estimates of site level methane emissions for the Fayetteville dry gas production region, with detailed data on episodic emissions from oil and gas operations.

The temporal and spatial resolutions that are needed for emission inventories depend on how the inventories are to be used. For example, if the objective is to determine annual emissions of greenhouse gases to assess long term trends in national emission patterns, then the spatial and temporal resolution provided in the current US EPA GHGI is adequate. In contrast, if inventories are being used to reconcile emission estimates with ambient observations that have time resolutions that are daily averages, hourly averages, or even shorter duration, the spatial and temporal resolution of the inventory should match the spatial and temporal resolution of the observations. Data collected in the Fayetteville Shale (Vaughn et al., 2018), have demonstrated that this matching of spatial and temporal scales of inventory and observations can be critical to reconciling observations with inventories.

In addition to variations in spatial and temporal resolution, methane emission inventories can be constructed based on a variety of emission factors. The method used to scale up an emission measurement from a limited set of samples to a larger regional or national total is to multiply the average result or the distribution of results of the emission measurement by the number of times that emission occurs in the larger scale. Generally the emission measurement is thought of as an emission factor, and is applied to a particular source in some discreet increment (such as an emission per event, device, component, or location). The term used to scale up the emissions is called the activity factor, and is the count or population of the source or event at the scale of interest, such as regionally or nationally.

Depending on the scale at which activity and emission data are collected, the aggregation of individual emission sources can be done at an individual site that has multiple pieces of equipment, regionally over many sites, or at a national level. For methane emission inventories, extensive new measurements of emission and activity factors, at site, regional, and national levels, have been reported. Mechanisms for rapidly and transparently incorporating these measurements into scalable emission inventories are not generally available, however.

Emission inventories can also be informed by operational data. For example, the pressure and temperature at which a well site separator operates will determine the volume of methane that is flashed when liquids are sent from the separator to atmospheric pressure oil and water tanks. These tank flashes may or may not be released as emissions depending on whether emission control systems on the tanks are in place and have the capacity required to manage emissions from tanks. Similarly, a compressor operating at a continuous load will have very different emissions than a compressor operating at the same average flow, but having frequent start-ups and shut-downs, due to additional emissions from gas powered starter motors and blowdowns. Few current inventories have the capability to efficiently and transparently incorporate operational data.

Finally, most inventories that include methane emissions report only methane and not a broader range of chemical species that might be used in chemical fingerprinting of sources or in applications such as estimating emissions of volatile organic compounds (VOCs). The molecular and isotopic fingerprints of methane emission sources vary as gas proceeds through the oil and natural gas supply chains, and these distinctive molecular footprints can be useful in attributing sources of methane emissions (Allen et al., 2017; Cardoso-Saldaña et al., 2019; Allen, 2016). Therefore, even for methane-specific analyses, it is useful for methane inventories to report co-emitted species.

This work describes a Methane Emission Estimation Tool (MEET), which addresses these needs. MEET has the ability to estimate emissions over time scales ranging from seconds to years, the ability to include multiple different types of emission factors, the ability to incorporate key operational data and the ability to generate emission estimates for multiple light alkanes (ethane, propane, butanes) and VOCs co-emitted with methane. The model also generates uncertainty estimates, using a Monte Carlo approach, and the source code is publicly available. This work describes MEET's capabilities for individual well sites. Additional work describes MEET's capabilities for gathering and compression stations and MEET's capabilities for estimating emission compositions (Cardoso-Saldaña et al., 2021).

2. Methods

MEET is a modular model developed in Python and its basic structure is shown in Fig. 1. The process begins with the user providing activity data for the site(s) to be simulated. For well sites, activity data include information such as well locations, gas production, oil/condensate production, water production and equipment counts at each site. Compositions of produced gas are also input by the user or selected from the Emission Composition Tool (ECT), a searchable library of emission compositions, available with MEET (Cardoso-Saldaña et al., 2021). Emission rates are estimated for each major equipment type at well sites. In each source category, emissions are estimated by multiplying activity data by emission factors selected from a library of measurements or user defined data. Emission time series are generated by a discrete event simulator at user specified time resolutions. Monte Carlo simulations are then performed to generate uncertainty estimates. Each of these steps is described in more detail below and in Supporting Information (SI).

2.1. Import activity data

The emission estimation process begins with the user importing activity data such as location, oil/condensate/gas/water production, equipment counts, and episodic event data for each site. Pre-formatted Excel spreadsheets of data are completed for each site. The spreadsheets and the associated data requirements are described in Supporting Information (SI).

2.2. Import molecular composition of emission sources

MEET categorizes emission sources into five major composition types. These compositions are labeled as separator feed, separator overhead (produced gas), condensate tank flash, water tank flash, and pipeline gas. The locations of these source compositions in a typical supply chain are shown in Fig. 2 (Allen et al., 2017). All of the compositions except pipeline gas are related. Separator feed, water tank flash, condensate tank flash and separator overhead (produced gas) all feed to or emerge from a well site separator. The separator allows sufficient retention time for the hydrocarbon and water mixture to reach thermodynamic equilibrium. Changing the operating conditions of the separator will change the compositions of the streams; changing the specifications for any one of the streams (e.g., produced gas composition) will lead to changes in the specifications

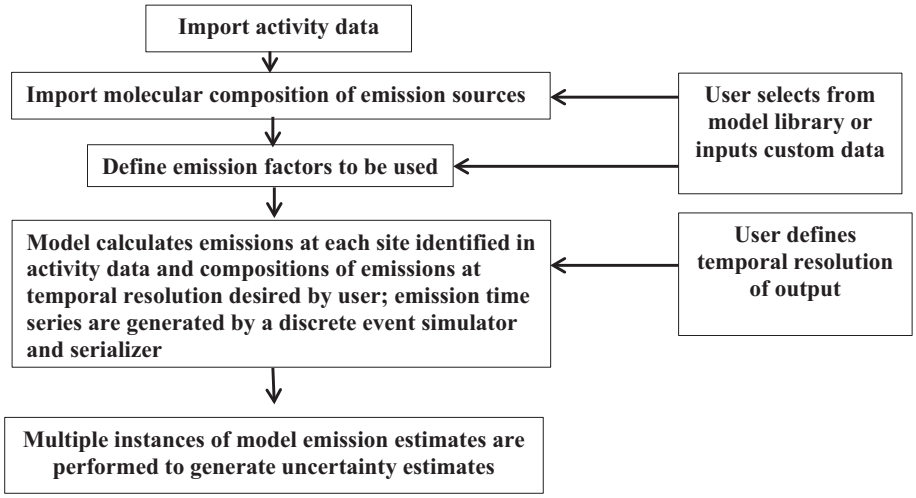

Fig. 1. Information flow for the modular Methane Emission Estimation Tool (MEET).

for all of the other streams. These changes have been simulated with thermodynamic models based on the Peng-Robinson Equation of State and Henry's Law and assembled into the ECT's searchable library, which includes approximately 10^6 self-consistent sets of compositions. The MEET model can import compositions at each well site from the ECT's searchable library by matching data commonly available for natural gas and oil well sites using the ECT's look up feature (Cardoso-Saldaña et al., 2021).

2.3. Define emission factors to be used

As documented recent reviews (National Academies of Science, 2018), multiple sources of data are available for oil and gas sector emission factors. In many recently collected data sets, total emissions are dominated by a small subset of the sampled population. For example, for pneumatic controllers, 19% of devices in a national population of controllers at well sites accounted for 95% of emissions (Allen et al., 2015a). In Oklahoma, 3.5% of the pneumatic controllers at well sites accounted for 73% of controller emissions (Gibbs, 2015). Other data sets show similar characteristics for many other sources (e.g., well completions, compressors, wells with liquid unloadings that result in venting). These highly skewed distributions of emissions from individual sources have been utilized in multiple ways in developing emission inventories. Some investigators have fit the distributions to mathematical functions (e.g., log-normal distributions). Others have used Monte Carlo type approaches and have randomly sampled

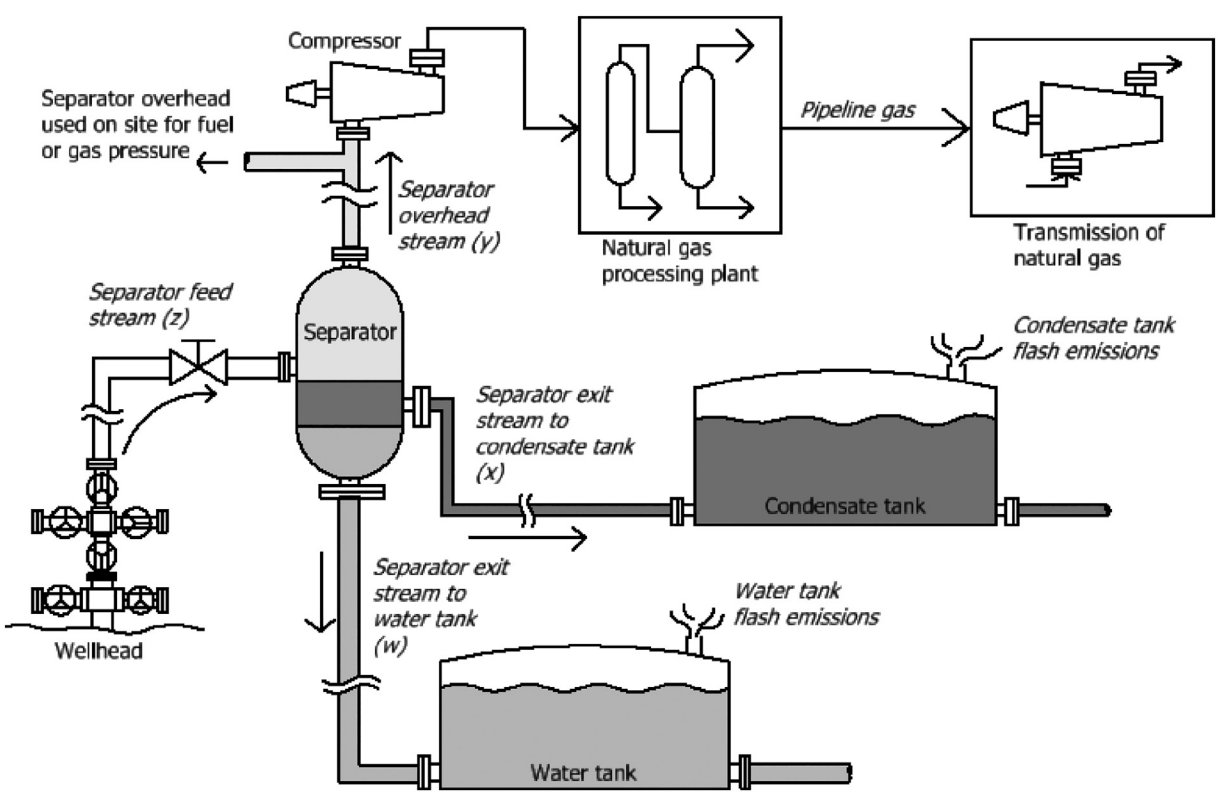

Fig. 2. Categories of emissions in a representative oil and gas production region. Reprinted with permission from (Allen et al., 2017) David T. Allen, Felipe J. Cardoso-Saldaña, and Yosuke Kimura Environmental Science & Technology 2017 51 (20), 12,016–12,026 DOI:https://doi.org/10.1021/acs.est.7b02202. Copyright 2017 American Chemical Society

individual data from distributions in developing inventories. Other investigators and national inventories like the EPA GHGI use average values from the distributions. MEET supports multiple approaches to using emission factor data sets including the use of constant emission factors, sampling from an ensemble of individual measurements, and allowing a user to input their own emission factors. The choice of approach is selected by the user. The emission source categories supported by the model for well sites are listed in Table 1. Emission calculations for each source type are described in the SI. The emission estimation processes for pneumatic controllers, tank flashing, leaks and liquid unloadings are briefly described below. More details on the calculation methods for these sources, descriptions of procedures for other emission source categories, and the sensitivity of emission predictions to different emission factor selections are described in the SI.

For pneumatic controllers, MEET offers the user the opportunity to use EPA average emission factors, emissions selected from a user defined or library data set, or emissions for normal and abnormal emissions selected from user defined or library data sets with transition probabilities between normal and abnormal operation defined by user or MEET default values. The library data set is a data set of 377 measurements of emissions from pneumatic controllers distributed at oil and gas production sites throughout the United States (Allen et al., 2015a). If the library data set is chosen along with constant emissions, an emission factor for each controller is randomly sampled from the distribution of measured emission rates. Emissions are assumed to be continuous. If the user selects transitions between normal and abnormal emissions, the controller will be assigned emissions from normal and abnormal emissions selected from the library or a user specified data set. The controller will be set in an initial mode (either normal or abnormal emissions) and a time in mode. At the end of the time in mode, a discrete event simulator, described below, will transition the controller into the other mode and will select a new time in mode. This process will continue throughout the simulation. Details are described in the SI.

Another category of emissions, that is intermittent in character, is tank flashing. Upstream production sites that produce water, condensate or crude oil send liquids from a pressurized separator (~100–1500 psi) to an atmospheric pressure tank. Light hydrocarbons dissolved in the liquids at separator conditions will "flash" into the gas phase when they reach the atmospheric pressure storage tanks. Typically, the liquids transfer from the separator to the atmospheric pressure tanks is done in batches (or dumps), rather than continuously. The separator accumulates liquid until a maximum liquid volume is reached and at that point a controller opens a valve, allowing discharge to the atmospheric pressure tank. The MEET well site tank module predicts the volume of flashed gas associated with each dump, the frequency of the dumps, the duration of the flash, and the temporal emission pattern of flashed gas. The volume of the flash gas associated with the separator dump is calculated based on the composition from the ECT emission composition database (mass of emissions per barrel of liquid produced) and the liquid volume in each dump from MEET (either user specified or a default value). The frequency of separator dumps is calculated by dividing the liquids production rate by the dump volume. The duration of the flash is either user specified or given a default value. The gas evolved during the flash is assumed to be a triangular wave. Details are described in the SI.

While each tank flash event has a characteristic temporal pattern, the amount of material flashed has a longer time scale pattern caused by declines in production. As production declines over time, less liquid is generated, and therefore separator dumps are less frequent. Since the volume of a dump is assumed to be determined by the design of the separator, the characteristics of each individual dump are assumed to remain constant, however, the frequency of dumps decreases over time. Well production decline models used by MEET are described in the SI.

Liquid unloadings have the potential to dominate emissions from a well site, when they occur. Unloading may be necessary when a well, that produces gas along with liquids, accumulates liquids in the well bore. The liquids accumulation may be due to a variety of causes, including decreases in gas velocity in the well, decreases in reservoir pressure, or changing gas to liquid ratios. As liquids accumulate, well production can decline and an operator may choose to unload the liquids from the well to restore production. Liquids can be unloaded in a variety of ways. For example, the well tubing can be modified to increase gas velocity or a pump may be installed to remove downhole liquids. Neither of these methods result in venting. Other unloading methods, such as temporarily diverting the flow from the well to an atmospheric vent, do lead to venting. MEET estimates emissions from unloadings that vent. The venting typically lasts minutes to hours and occurs with a frequency that can range from several times per year to thousands of times per year. MEET models three types of liquid unloadings: manual unloadings of wells without plunger lifts, manual unloading of wells with plunger lifts, and automated unloadings of wells with plunger lifts. The activity data and emissions used by MEET are drawn from a national data set of liquid unloading measurements (Allen et al., 2015b),

Table 1
Well site emission source categories in MEET.

Source category[a]	Emission factor type	Emission factor description	Temporal pattern
Chemical injection pump	Constant	EPA emission factor or emissions selected from a user defined or library data set	Constant
Pneumatic controller	Constant	EPA emission factors, or emissions selected from a user defined or library data set	Constant
	Transitions between normal and abnormal emissions	Emissions for normal and abnormal emissions selected from user defined or library data sets; transition probabilities between normal and abnormal operation defined by user or MEET default values	Time series with emissions transitioning between normal and abnormal operation for individual controllers
Condensate or water tank flash	Intermittent separator dump	Emissions calculated from thermodynamic model of separator and volume of intermittent separator dumps to atmospheric pressure tanks	Triangular wave form
Leaks	Transitions between leaking and non-leaking mode	Emissions for leaking components selected from user defined or library data sets; transition probabilities between normal and abnormal operation defined by user or MEET default values	Time series with emissions transitioning between leaking and non-leaking operation for individual components
Liquid unloadings	Automated plunger lift	EPA emission factor or emissions per event and unloading frequency selected from a user defined or library data set	Duration of unloading event selected from a user defined or library data set; emissions during event constant
	Manual plunger lift	EPA emission factor or emissions per event and unloading frequency selected from a user defined or library data set	Duration of unloading event selected from a user defined or library data set; emissions during event constant or selected from a user or library variability pattern
	Manual without plunger lift	EPA emission factor or emissions per event and unloading frequency selected from a user defined or library data set	Duration of unloading event selected from a user defined or library data set; emissions during event constant or selected from a user or library variability pattern
Completions	Constant	EPA emission factor or emissions selected from a user defined or library data set	Constant
Additional fugitive emissions	Transition between emitting and non-emitting	User input distributions or selected from libraries of field observations	Constant while in emitting mode

[a] Modules for compressors and dehydrators, sometimes found on well sites are described in the companion paper on gathering and compression sites.

with emissions capped at a rate dependent on gas production. Details are provided in the SI.

In addition to emissions associated with normal well operations, MEET includes estimation tools variously referred to as fugitive, abnormal or unintended emissions. These emissions will be referred to as fugitive emissions in this work and include leaks, abnormal operation of pneumatic devices and other unintended releases. All of these fugitive releases are treated stochastically in MEET.

Leaks are modeled through leak/no-leak transitions. Components on major equipment (well-head, separator, tanks, piping) are assigned to be either in leaking or non-leaking mode and a time in mode. At the end of the time in mode, a discrete event simulator, described below, transitions the component into the other mode and selects a new time in mode. This process continues throughout the simulation. Details are described in the SI.

As noted in the description of pneumatic controllers, if the user chooses to model controllers as transitioning between normal and abnormal emissions, the controller will be assigned emissions from normal and abnormal emissions selected from the library or a user specified data set. The controller will be set in an initial mode (either normal or abnormal emissions) and a time in mode. At the end of the time in mode, a discrete event simulator, described below, will transition the controller into the other mode and will select a new time in mode.

Finally, there are a variety of fugitive emissions that have been observed from well sites, from disparate sources including, but not limited to unlit flares, flares with low combustion efficiencies, improperly operating separator dumps, and emissions from open tank hatches. MEET models these emissions by assuming a frequency distribution of these events and cycling modes between emitting and non-emitting. These frequency distributions are based on field observations but it is expected that individual production basins and operators will have very different emission frequencies. Because of the uncertainties associated with these emissions, in this work, these estimates will be used in sensitivity analyses.

2.4. Discrete event simulator

Developing time series of emission estimates requires MEET to march over time. A Discrete Event Simulator (DES) was used to implement time marching. The DES is implemented in Python. Various sources have emissions that vary over time due to cycling through different states of behavior, such as a pneumatic controller cycling between normal and abnormal emission modes or a well going through a periodic liquid unloading. The DES models these states of behavior on a time axis. Initiating a new state results in the assignment of a time in that state and the next state transition is scheduled. For example, when a tank flashes, MEET predicts when the next flash will occur based on the dump volume and the liquid production rate. This scheduling can be deterministic, or stochastic, or dependent on other events, such as starting and shutting down a compressor. DES orchestrates events from various entities and saves a sequence of event logs identifying timing of events. Time is recorded as seconds from the beginning of the simulation. The sequence of events that DES recorded is post-processed by a serializer by interpreting each event into an emission time series at desired temporal resolution (second, minute, hourly, daily, monthly, annual).

2.5. Uncertainty and variability

Uncertainty and variability in well site emissions are both characterized by the MEET model. For this work, variability in emissions is defined as the distribution of emissions that a short time resolution measurement might record at a single well site over the course of a long sampling period. In this work, variability will be expressed as (i) the distribution of emissions, aggregated over a one day period, which would be observed over one to four years; and (ii) the distribution of emissions, aggregated over a one minute period, that would be observed over a one day period. Variability could also be assessed over different averaging times. Uncertainty is driven by the use of distributions, rather than single values as emission factors. In characterizing uncertainty for emissions from single well sites, activity data (e.g., numbers of controllers) and emission compositions, are assumed to have no uncertainty. Emission factors (e.g., emission rate assigned to each controller) and operations data, however, can be selected from distributions, and when emission distributions are used, Monte Carlo simulations are performed to characterize uncertainty. This work will express uncertainty as the variation in annual emission rates over 30 simulations.

3. Results and discussion

The use and utility of MEET is demonstrated through three types of applications. These are (i) analysis of site total emissions from single facilities; (ii) analysis of the impact on methane emission intensities, over time, of emission mitigation measures, and (iii) analysis of total emissions from a large ensemble of sites, representing a collection of sites that may be measured by aircraft flying transects upwind and downwind of a production region.

3.1. Case studies of applying MEET to estimate emissions at individual well sites

The purpose of this case study is to illustrate (i) that a typical well site will have average emission rates that change over time, (ii) that at any given time a well site will exhibit a range of emission rates that exhibit complex patterns, and (iii) that the time resolution at which the site total emission rates are measured will impact the patterns observed in the emission rates.

Emission time series of five prototypical well sites were generated using MEET. Three sets of well parameters are used as inputs to the searchable emission composition (ECT) database, and the key parameters are shown in Table 2. Each set of parameters represents a typical production profile and will return a set of 5 emission composition profiles, as described in the Methods section. The first parameter set (Composition profile ID labeled as Wet gas 1) represents a wet gas rich in gas (high gas to oil ratio) with a low separator pressure; the profile labeled Wet gas 2 describes a wet gas rich in condensate with a moderate separator pressure; The profile labeled Oil represents an oil production profile with a high separator pressure. MEET is also able to simulate wells with only dry gas production (no liquids produced) and wells with 2 separators.

Each of the five simulated wells takes an emission composition profile from Table 2. The five wells are assumed to be located in the Eagle Ford Shale, LaSalle County, where gas-to-oil ratios (GORs) of the wells range from 100 to over 50,000, and the API gravity of the liquid produced ranges from 30 to over 63 (Gherabati et al., 2016). Water and oil production of these wells use hyperbolic decline curve parameters, in LaSalle County, reported by the U.S. EIA National Energy Modeling System (US Energy Information Administration, 2013). The initial oil gas and water production rates are shown in Table 3. Table 4 shows the emission estimation methods used in the simulations. The first two wells and the fifth well are simulated for one year without liquid unloading events. The third well, with a relatively large amount of both oil and water production but a low separator pressure, is assumed to accumulate liquids, and to require liquid unloading

Table 2
Key input parameters of the searchable thermodynamic model database.

Composition profile ID	Number of separator stages	API gravity	Gas-to-oil ratio (scf/bbl)	Temperature (°F)	1st stage pressure (psia)	Produced gas methane (molar fraction)
Wet gas 1	1	58	9100	80	150	0.78
Wet gas 2	1	49.8	2000	80	650	0.80
Oil	1	36.5	630	100	1400	0.89

Table 3
Composition profile, production, and liquid unloading information for five prototypical wells.

Well ID	Composition profile	Initial oil production (bbl/day)	Initial gas production (mcf/day)	Initial water production (bbl/day)	Liquid unloading technology	Liquid unloading frequency (events/year)
1	Wet gas 1	807	7343.7	1500	NA	NA
2	Wet gas 2	807	1614	1500	NA	NA
3	Wet gas 1	807	7343.7	8070	Automated Plunger	1000
4	Wet gas 1	807	7343.7	1500	Manual	4
5	Oil	807	508.41	1500	NA	NA

Table 4
Summary of emission factors and activity data for the five prototypical well sites.

Emission source	Emission factors	Activity factors
Well completion	Allen et al. (2013) distribution of measured emissions	Duration of completion sampled from Allen, et al. (Allen et al., 2013), fixed emission rate
Liquid unloadings	Allen et al. (2015b) distribution of measured emissions	Event frequency for case studies specified in Table 3
Pneumatic controllers	Allen et al. (2015a) distribution of measured emissions; definitions of normal and abnormal emissions defined in SI	2 controllers per well; emissions transitioning between normal and abnormal emissions factors
Condensate/water tank flash	Emissions per barrel of liquid determined based on thermodynamic analysis; vapor recovery units with a capture efficiency of 98% assumed in base case analysis; sensitivity analyses conducted with uncontrolled tanks	1 condensate tank and 1 water tank per well; Dump volume: 15 gals of liquid per dump; triangular emission wave form
Chemical injection pumps	Allen et al. (2013) distribution of measured emissions	1 pump per well
Leaks	Allen et al. (2013) distribution of measured emissions	One well head; one separator, 2 tanks, yard piping

operations (specified as 1000 automated plunger lifts per year) to maintain production. In comparison, the fourth well, with a lower water production, may only require occasional liquid unloading operations (specified as 4 manual unloadings per year). The start time of liquid unloading events are randomly selected. A four year simulation is performed for the wells with liquid unloadings.

Figs. 3 and 4 show a 1-year methane emission time series of the wet gas wells 1 and 2 with emissions aggregated over one day periods. As shown in these figures, typical well site will have average emission rates that change over time. During the first month of simulation, as the well is completed and brought into production, emissions are dominated by emissions from well completion operations. Depending on the program-returned or user-specified activity and emission factor data, emissions from well completion may or may not exceed the initial emission levels from other sources, which dominate emissions during the rest of the simulation period.

Figs. 5 and 6 show 4-year methane emission time series of the wet gas wells 3 and 4 with emissions aggregated over one day periods. Emissions from wells 3 and 4 are dominated by emissions from tank flash in the first 6 months of production. The emissions from tank flash decline as oil and water production of the wells decline. Once liquid unloadings begin, they can dominate emissions, either for frequent but relatively

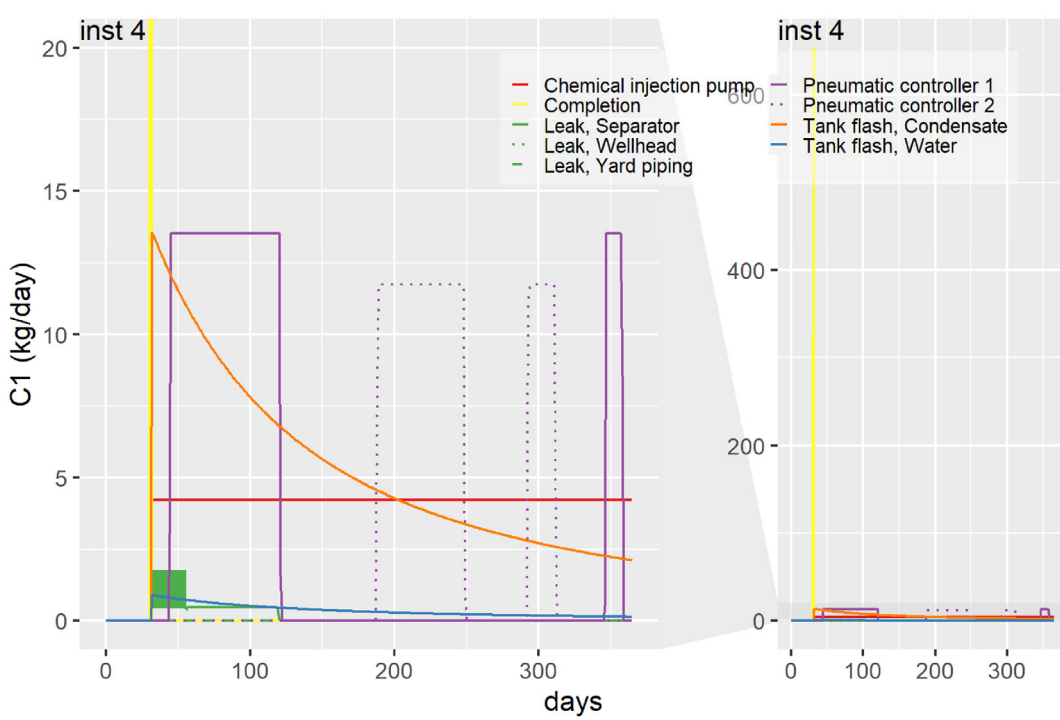

Fig. 3. 1-year methane emissions (kg/day) time series of well 1 by emission source with daily time resolution; the simulation shown is the fourth instance (inst 4) of 30 used in Monte Carlo simulations to estimate uncertainty; note different vertical scales; all 30 instances are summarized in the SI.

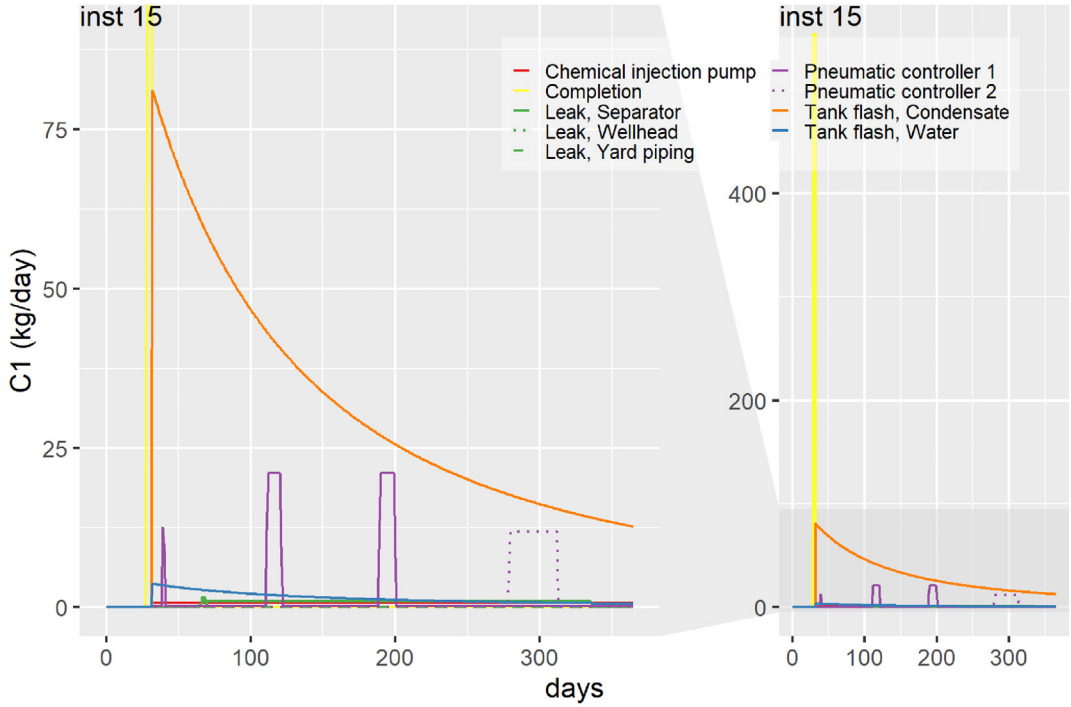

Fig. 4. 1-year methane emissions (kg/day) time series of well 2 by emission source with emissions aggregated on a daily basis; the simulation shown is the 15th instance (inst 15) of 30 used in Monte Carlo simulations to estimate uncertainty; note different vertical scales; all 30 instances are summarized in the SI.

small liquid unloading events, or for infrequent but large unloading events (Figs. 3 and 4).

Fig. 7 shows a 1-year methane emission time series of the oil well 5 with daily time resolution. With a higher separator pressure, more methane is dissolved in the condensate and water phases in the separator and flash as emissions from condensate and water tanks under atmospheric pressure.

The time series presented in Figs. 3–7 would change, depending on the values selected for the emission factors for the controllers (each controller has an emission rate selected from a data set; Allen et al., 2015a), for the completions, for chemical injection pumps (each pump and completion has an emission rate selected from another data set; Allen et al., 2013), and for the unloadings (each well's unloadings has an emission rate selected from an unloading data set;

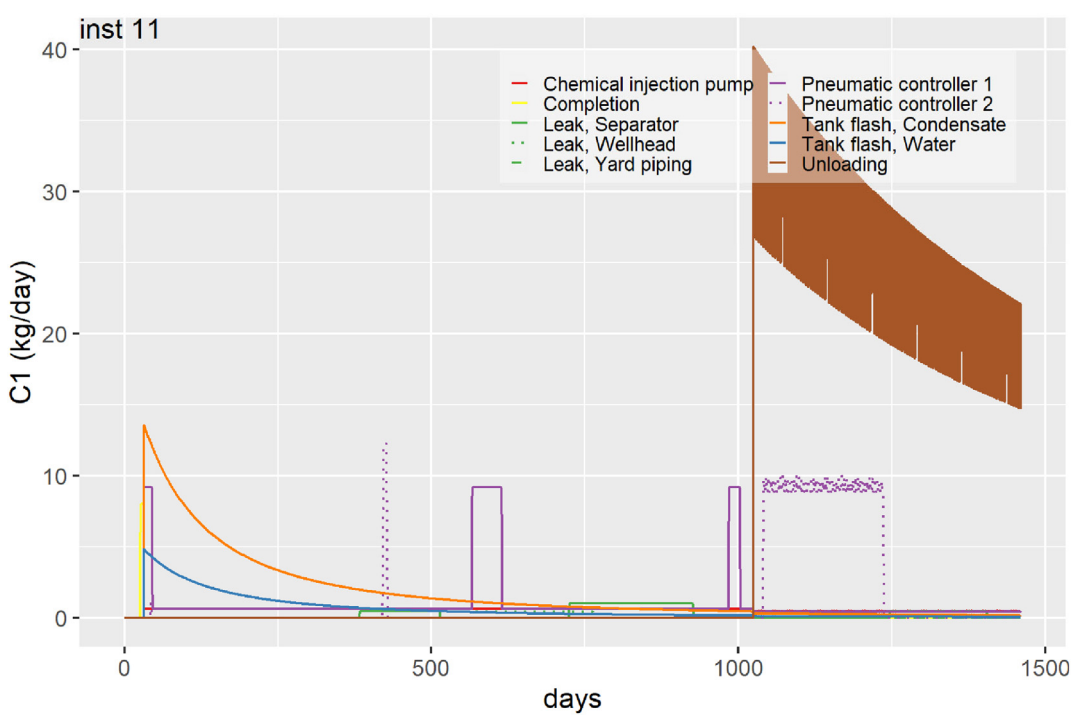

Fig. 5. 4-year methane emissions (kg/day) time series of well 3, with frequent automated liquid unloadings after 3 years of production, by emission source with emissions aggregated on a daily basis; the simulation shown is the 11th instance (inst 11) of 30 used in Monte Carlo simulations to estimate uncertainty; all 30 instances are summarized in the SI.

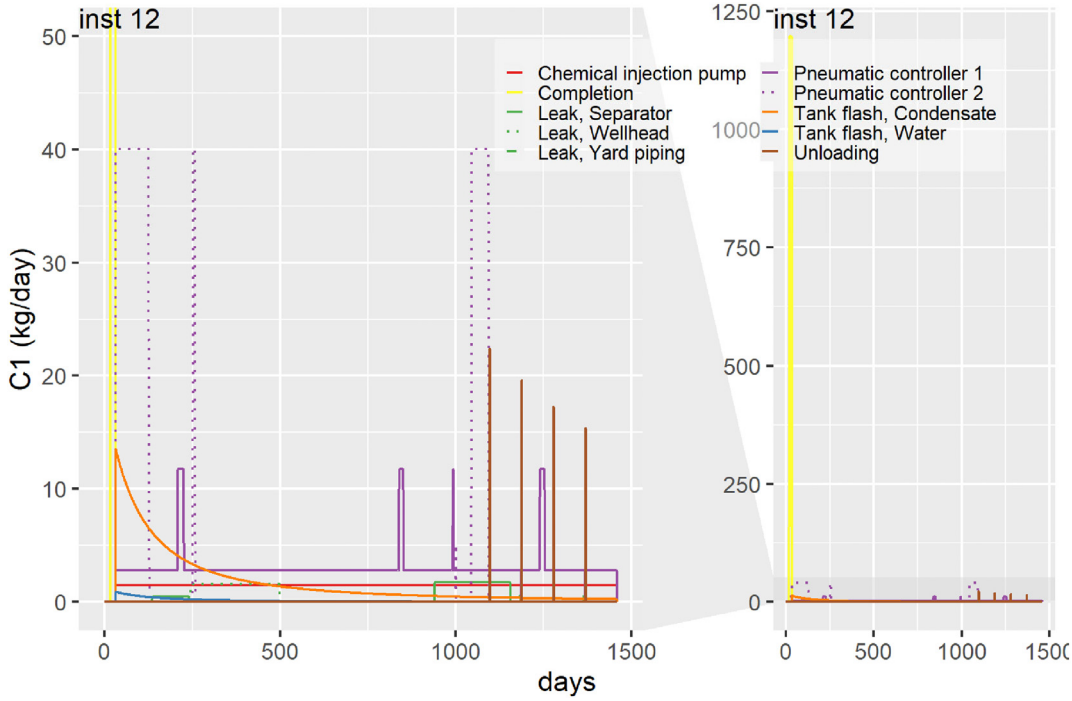

Fig. 6. 4 year methane emissions (kg/day) time series of well 4, with infrequent manual liquid unloadings after 3 years of production, by emission source with emissions aggregated on a daily basis; the simulation shown is the 12th instance (inst 12) of 30 used in Monte Carlo simulations to estimate uncertainty; note different vertical scales; all 30 instances are summarized in the SI.

Allen et al., 2015b). To characterize the uncertainty associated with the distribution of emission factors, Monte Carlo simulations were performed for each well. Results for 30 instances of the simulation for well 3 are presented in Fig. 8. Unloadings exhibit the largest uncertainty in estimated emissions, depending on the event selected. Statistics of the Monte Carlo simulations are described in Supporting information (SI).

MEET can also be used to assess emission variability. Fig. 9 illustrates that at any given time a well site will exhibit a range of emission rates that exhibits complex patterns. The figure shows the

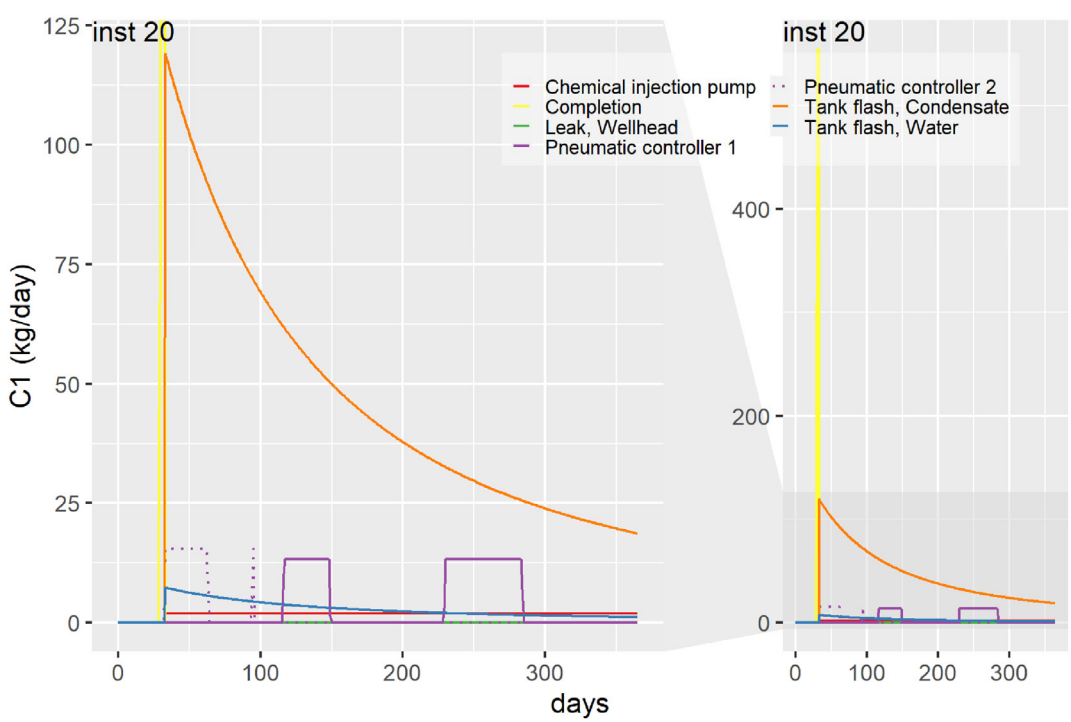

Fig. 7. 1 year methane emissions (kg/day) time series of well 5 by emission source with emissions aggregated on a daily basis; the simulation shown is the 20th instance (inst 20) of 30 used in Monte Carlo simulations to estimate uncertainty; note different vertical scales; all 30 instances are summarized in the SI.

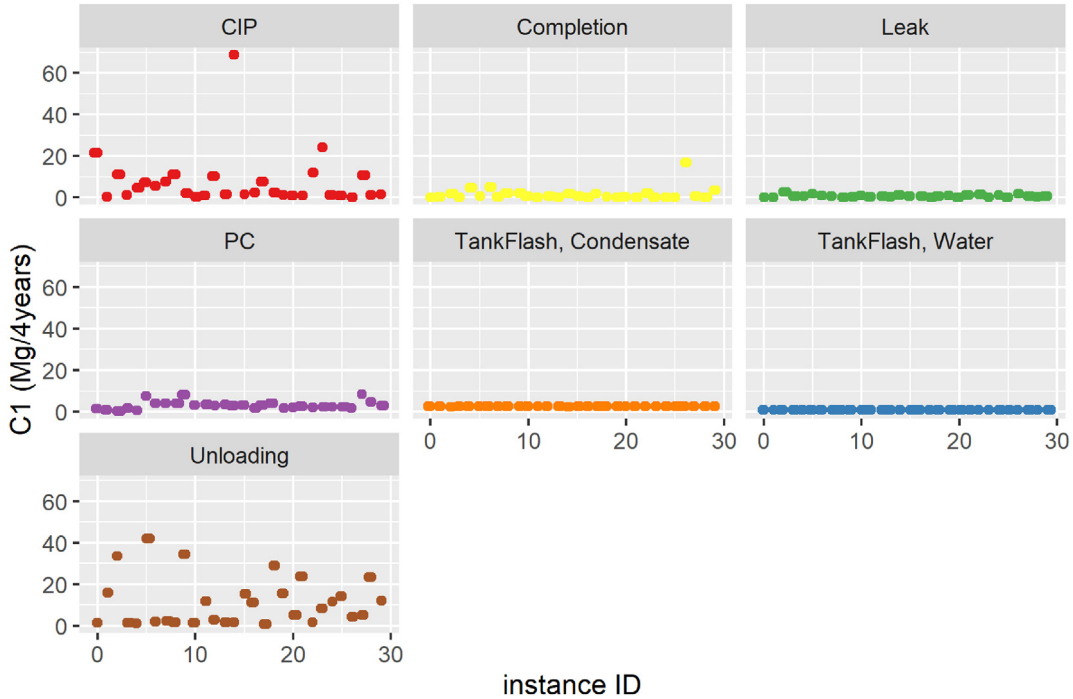

Fig. 8. Thirty instances of emissions, aggregated over 4 years of operation, for well 3. CIP represents chemical injection pumps and PC represents pneumatic controllers.

predicted emission variability for well 3, when emissions are aggregated on a daily basis for two instances drawn from the 30 simulations summarized in Fig. 8. Fig. 9a shows the distribution of predicted daily emissions for an instance with large unloading emissions (140 kg/unloading event). The unloadings appear with emission rates of 20–50 kg/day, larger than any of the other routine emissions.

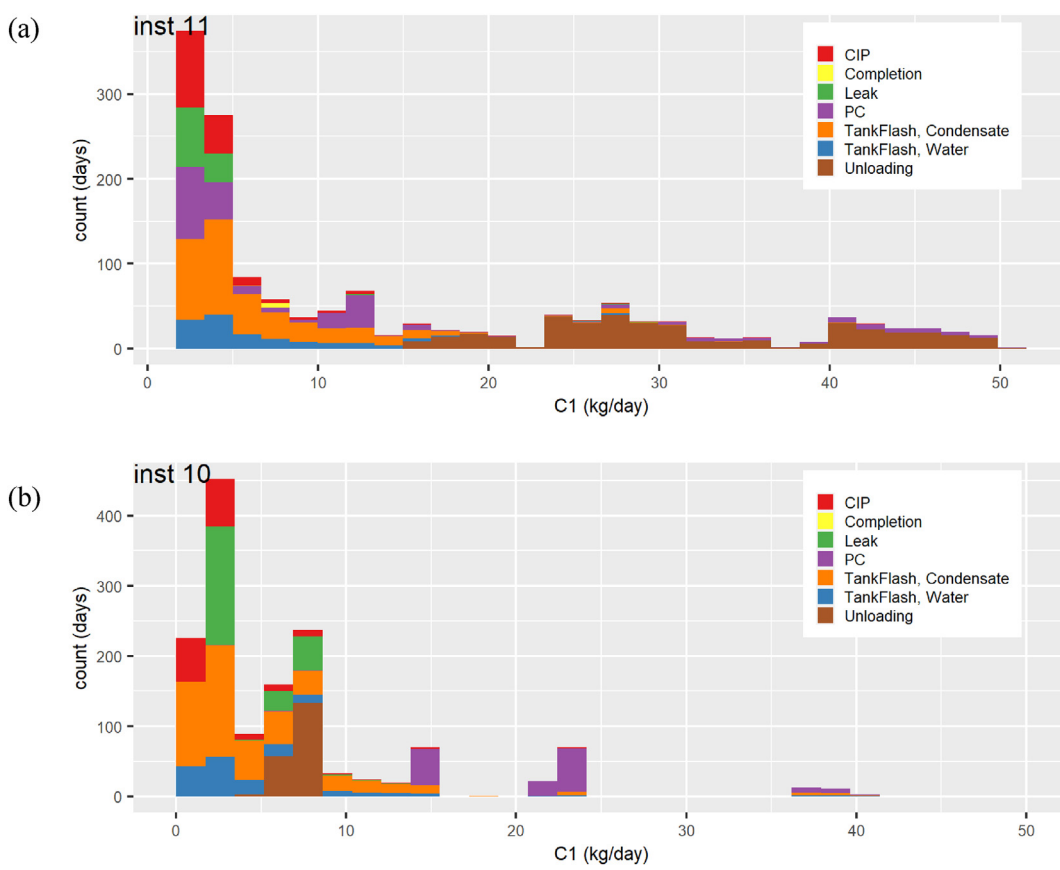

Fig. 9. Distribution of emissions over 4 years of operation, aggregated on a daily basis, from well 3. (a) Scenario with high emissions per unloading event (140 kg/event); (b) scenario with low emissions per unloading event (23 kg/event).

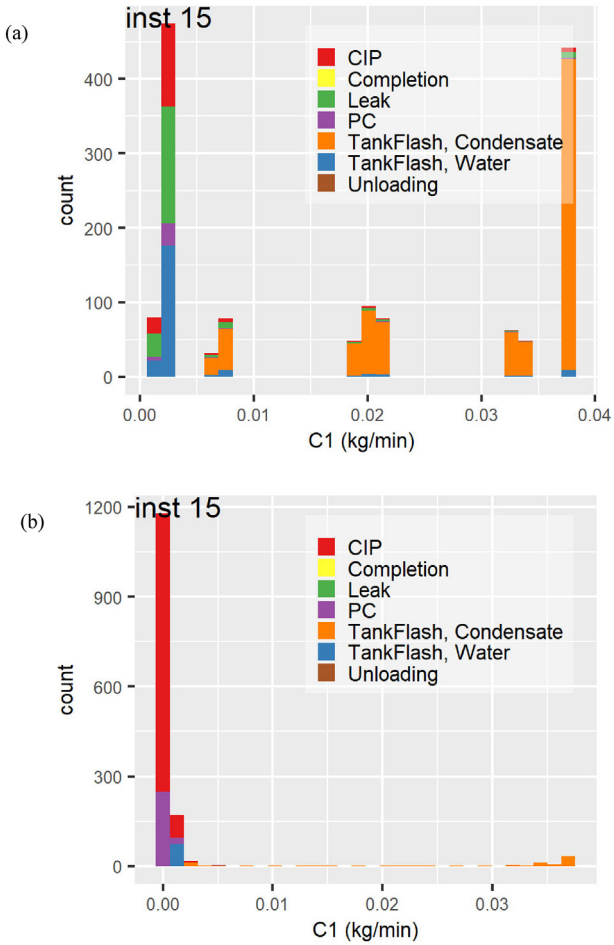

Fig. 10. Distribution of emissions for well 2, aggregated at 1 min intervals for a one day simulation, after 6 months of production (a) and after 3 years of production (b).

With these emission rates, the site total emission event when unloadings are occurring is readily distinguishable from other modes of operation for the site. In contrast, however, if the emissions per unloading event are significantly lower (23 kg/event), the unloadings are difficult to separate in the distribution on the basis of emissions per day.

Finally, the time resolution at which the site total emission rates are measured will impact the patterns observed in the emission rates. For example, tank flashes occur periodically and have durations of just a few minutes. If emissions are aggregated at a 1 min resolution, as shown in Fig. 10 for one day of simulation for well 3, a bimodal to multi-modal distribution can result, depending on whether or not a tank flash is occurring during a particular minute. This distribution can evolve over time (Fig. 10), as the frequency of tank flashing decreases with declines in production.

3.2. Case study of applying MEET to evaluate the impact of mitigation measures on methane emission intensity

A frequently reported metric in studies of methane emissions from oil and gas production sites is the methane emission intensity. The precise definition of methane emission intensity in this work is methane emissions, divided by methane in the gas produced at the site, expressed as a percentage. The purpose of this case study is to illustrate that emission intensity varies over long time periods, and that different emission mitigation measures will have different impacts on the evolution of methane emission intensity.

Emission intensity varies over time because some emissions, such as tank flashes, scale with production rate, while other emissions, such as leaks, do not. Cardoso-Saldaña and Allen (2020) examined this issue in detail and Fig. 11a illustrates the behavior of emission intensities over time for well 3, described in the previous section, assuming no emission controls on any of the sources. As described in more detail in the SI, although overall emissions go down over time as tank flashes decrease with decreases in production, overall methane emission intensity goes up, particularly once liquid unloadings are initiated. Overall, as shown in Fig. 11, the evolution of emission intensity over time is complex. Fig. 11 also illustrates three emission mitigation scenarios: 98% emission controls on tanks (Fig. 11b), 100% emission controls on pneumatic controllers (Fig. 11c), and 100% emission controls on liquid unloading events (Fig. 11d). Different mitigation measures have different impacts on emission intensity over time. Control measures that reduce emissions that are independent of production rate (e.g., liquid unloadings) have a different impact on emission intensity over time than control measures focused on emissions that scale with production (e.g., controls on tanks). More details of this case study are provided in the SI.

3.3. Case study of applying MEET to an ensemble of production sites

A large number of studies have been conducted in which aircraft, flying transects upwind and downwind of production regions, have estimated total emissions from a production basin. These basin total emissions are frequently compared to annual average emission inventories. If the sites being sampled have large episodic emissions, such as liquid unloadings, the episodic emissions can influence hourly or daily emission rates. This was the case in the Fayetteville Shale production region study reported by Vaughn et al. (2018). In this production region, unloading events are relatively frequent, and a single unloading event can influence basin total emissions. As a case study, MEET was applied to 261 sites with 746 well sites in the Fayetteville Shale, using site data assembled by Vaughn et al. (2018). Twenty four hourly emission estimates from MEET were grouped by source aggregated for the 746 sites. Fig. 12 shows that the aggregate emissions can vary over a factor of two, depending on the unloading events that are occurring in any particular hour. More details of this case study are available in the SI.

Many more case studies could be shown, including emissions due to abnormal operation modes for a variety of devices. The SI describes details of the three case studies presented here as well as additional analyses. Overall, the results emphasize the importance of developing detailed emission inventories at appropriate time scales, when comparing measurements to routine emissions. The model described in this work supports such comparisons and is freely available.

CRediT authorship contribution statement

Felipe J. Cardoso-Saldaña, Yosuke Kimura, Qining Chen, Zhanhong Xiang, Clay Bell, Chris Lute, Jerry Duggan: Formal analysis.

Daniel Zimmerle, Matthew Harrison: Conceptualization.

David T. Allen: Conceptualization; Funding acquisition; Methodology; Writing – initial draft, review & editing.

Disclosure

The authors declare the following competing financial interest(s): This work was supported by the Collaboratory to Advance Methane Science. One of the authors (DTA) has current research support from Exxon Mobil Upstream Research Company, and the National Institute for Clean and low Carbon Energy (NICE). Over the past five years, DTA has also worked on methane emission measurement projects that have been supported by multiple natural gas producers and Environmental Defense Fund. DTA has done work as a consultant for multiple companies, including British Petroleum, Cheniere, Eastern Research Group, ExxonMobil, KeyLogic,

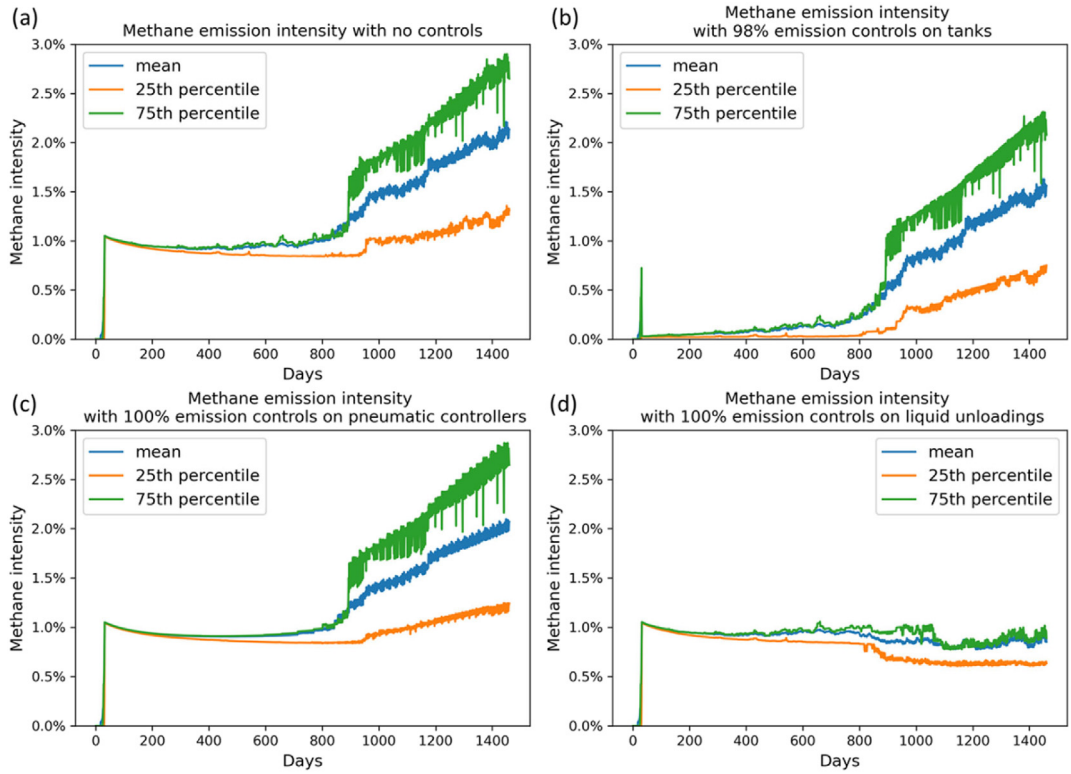

Fig. 11. 4-year methane emission intensity time series for well 3 with (a) no emission controls, (b) 98% emission controls on tanks, (c) 100% emission controls on pneumatic controllers, and (d) 100% emission controls on liquid unloadings; emissions aggregated on a daily basis; the plots show the mean, 25th percentile, and 75th percentile of 30 Monte Carlo instances for each emission mitigation scenario.

NICE and SLR International. Another co-author (F.J.C.S.) did an internship at ExxonMobil Upstream Research Company during the period of work on this project.

Declaration of competing interest

The authors declare the following competing financial interest(s): This work was supported by the Collaboratory to Advance Methane Science (CAMS), a group of companies operating facilities in the natural gas supply chain.

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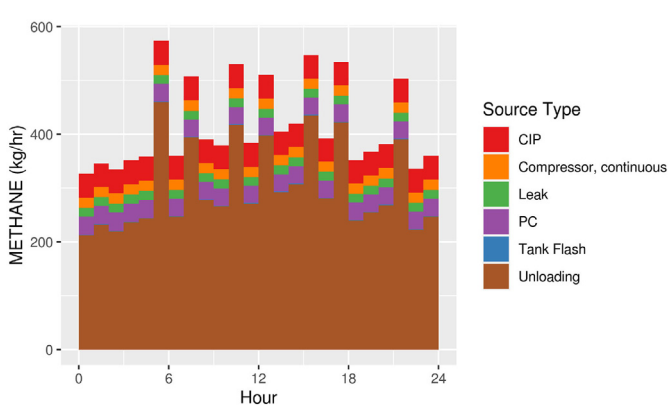

Fig. 12. Hourly emission estimates for an ensemble of 746 wells in the Fayetteville Shale.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.154277.

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