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Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods

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Abstract

Increased natural gas production in recent years has spurred intense interest in methane (CH_4) emissions associated with its production, gathering, processing, transmission and distribution. Gathering and processing facilities (G&P facilities) are unique in that the wide range of gas sources (shale, coal-bed, tight gas, conventional, etc.) results in a wide range of gas compositions, which in turn requires an array of technologies to prepare the gas for pipeline transmission and distribution. We present an overview and detailed description of the measurement method and analysis approach used during a 20-week field campaign studying CH_4 emissions from the natural gas G&P facilities between October 2013 and April 2014. Dual tracer flux measurements and onsite observations were used to address the magnitude and origins of CH_4 emissions from these facilities. The use of a second tracer as an internal standard revealed plume-specific uncertainties in the measured emission rates of 20–47 %, depending upon plume classification. Combining downwind methane, ethane (C_2H_6), carbon monoxide (CO), carbon dioxide (CO_2), and tracer gas measurements with onsite tracer gas release allows for quantification of facility emissions, and in some cases a more detailed picture of source locations.

1 Introduction

The natural gas industry has undergone a transformation in recent years, in large part due to technological advancements such as hydraulic fracturing and horizontal drilling. These advances have led to increases in domestic natural gas production (EPA, 2014b), although concomitant with this increase has been a rising concern over methane emissions from the entire natural gas system, from the perspective of both environmental impact and a loss of resources or product. Over the past decade, many studies have aimed at quantifying these emissions using a variety of methods, yielding a wide range of emissions assessments (Pétron et al., 2012; Allen et al., 2013; Kar-

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sions from facilities associated with a variety of types of gas, such as gas with low- and high-C₂₊ hydrocarbon content (here referred to as dry and wet gas, respectively), as well as sour (high sulfur and/or CO₂ content) and sweet gas sources (low sulfur and/or CO₂ content). More detailed information about site selection is presented by Mitchell et al. in the associated Measurements paper (Mitchell et al., 2014). These facilities handled natural gas derived from a variety of origins, including shale, coal-bed, and conventional wells. In many cases, the emission profiles associated with these facilities reflect the equipment used to prepare the natural gas (EIA, 2006; Kidnay et al., 2011). For example, the first step during gathering is often passage through gathering lines and a compressor (gathering) station. One of the primary purposes of gathering facilities is to collect and compress the input stream of gas to pipeline pressures, usually ~ 800 psi. This requires the use of compressors and associated equipment, for which there are multiple possible emission sources such as compressor seals, natural gas-driven pneumatic devices and engine exhaust. Frequently gathering facilities will also remove water from the gas stream using dehydration trains, which provide more possible emissions points. Following gathering, sweet, dry gas can typically be easily conditioned and sent to the distribution network. However, gas that is sour, wet, or with a high water content requires significant subsequent processing, such as the removal of natural gas liquids using forced extraction, and sometimes a dehydration step to further remove water (Kidnay et al., 2011; Jumonville, 2010). These relatively complex structures can involve distillation columns, turboexpanders, separators, compressors, pneumatic devices and heat exchangers, all of which can emit CH₄ either through minor fugitive components or venting. Finally, extracted natural gas can have high CO₂ and/or H₂S content (i.e. sour, especially in coal-bed methane and some shale gas regions), which requires amine treating (frequently collocated with other gas processing or compression facilities) to make it distribution-ready (Kidnay et al., 2011). Again, this equipment and additional processing adds to the number of possible emission sources.

Presented in the second half of this paper are examples of the unique chemical profiles associated with the gathering, treatment and processing systems utilized by the

natural gas industry. In the process of measuring CH₄ emission rates, these signatures can provide important information about contributions from specific methane sources on site.

2 Challenges in measuring emissions from natural gas facilities

5 The necessity for emissions measurements at natural gas facilities is two-fold: (i) as an assessment of the impact of facility operation upon regional and national air quality and climate (EPA, 2014a) and (ii) to quantify losses due to normal operation or identify large emission sources. In the case of (i), measured emissions provide an opportunity to compare to national estimates, and assess the overall impact of the natural gas
10 supply chain on CH₄ emissions in the US (Marchese et al., 2014; Subramanian et al., 2014). In the case of (ii), these measurements aid the natural gas industry in minimizing product losses.

2.1 Bottom-up approaches

Several approaches have been utilized to observe emissions at industrial facilities. In
15 some cases, a bottom-up approach is employed, wherein the magnitudes of emissions from individual components are directly measured and then added together to estimate the facility-level emission rate (FLER) (Subramanian et al., 2014; Harrison et al., 2011). This makes use of stack test data, manufacturer data, emission factors, engineering estimates, activity factors and onsite measurements. These onsite measurements can
20 take many forms, such as acoustic emission detection, which quantifies leaks through suspected leak points such as valves, and Hi-Flow[®] sampling, which can accurately determine emission rates from a variety of fixtures. While these methods are widely used and are capable of many measurements in a short time, they are not applicable to all possible emission sources due to the number and accessibility of fixtures within
25 facilities (Subramanian et al., 2014). This issue is particularly relevant at large process-

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ing and treating plants, where the inability to measure emissions from a large number of components could lead to an asymmetric bias in the reported FLER. In addition, in order to accurately scale bottom-up studies to nationwide (or even regional) estimates, care must be taken to ensure that the sampled population, which is typically small, accurately represents the national or regional inventory of facilities.

Optical gas imaging (e.g. infrared cameras such as FLIR[®]) is a method by which leaks can be identified by using real-time infrared imaging. This method provides a high duty cycle – dozens of fixtures within a facility can be investigated per hour – and large emitters can be readily identified. It is often used in conjunction with the above methods to locate possible leak sources. However, because the method does not measure CH₄ concentrations or flow rates, it does not quantify the emission magnitudes. It nonetheless serves as a powerful qualitative tool in leak detection, and is therefore leveraged in this study to identify suspected emission points at each G&P facility.

2.2 Top-down approaches

Top-down estimates aim to quantify methane emissions from a particular geographic region. These results can then be compared to inventories constructed from bottom-up measurements. Two top-down approaches are commonly used for determining regional methane emissions: mass-balance flights and fixed sensors fields (Zavala-Araiza et al., 2014). The mass-balance flight method, exemplified in several recent oil and gas basin studies (Karion et al., 2013; Pétron et al., 2013, 2012), uses upwind and downwind transects to capture emissions from a bounded region. This area can be as small as an individual facility, or as large as an entire basin. Under favorable meteorological conditions, such measurements can potentially estimate emissions from a large area with a single flight, but these techniques are costly and provide little to no source-specificity. This lack of source-specificity makes it especially difficult for top-down studies to determine the relative emissions from various activities within the industry (i.e. from gathering, processing, transmission, or production), or even differentiate between emissions from different industries, such as natural gas vs. feedlots vs. farming op-

is applied to quantify total facility-level methane emission rates (fugitive, venting and combustion) at natural gas processing plants, treatment facilities and midstream compressor stations.

Conceptually, the tracer release method is based upon the simple relation that the downwind concentration enhancement of gas X above ambient background, $\Delta[X]$, is directly related to the flow rate at its source, F_X :

$$\Delta[X] = \alpha \cdot F_X \quad (1)$$

The relation between these two quantities is determined by α . The coefficient α is a complicated function of meteorological information, such as wind speed, wind history, turbulence, solar irradiance, temperature, boundary layer height, local topography and downwind distance. In principle this information can be estimated using, for example, a Gaussian dispersion model (Beychok, 2005). Such models have had success in qualitatively reproducing measured plume data, but frequently lack the precision and accuracy required for this study, especially in areas with complex terrain and meteorology.

The tracer release method provides an empirical means to bypass the need for determining α (Lamb et al., 1986, 1995). By deploying a known flow of tracer gas located physically near a CH_4 emission source, the downwind tracer concentration enhancement (above background), $\Delta[T]$, downwind CH_4 concentration enhancement (above background), $\Delta[\text{CH}_4]$, and tracer flow rate, F_T , become measurable quantities. The ratio of the two downwind concentrations is then equal to the ratio of flow rates:

$$\frac{\Delta[\text{CH}_4]}{\Delta[T]} = \frac{\alpha F_{\text{CH}_4}}{\alpha F_T} = \frac{F_{\text{CH}_4}}{F_T} \quad (2)$$

where F_{CH_4} refers to the flow of CH_4 from the facility. Because concentrations $\Delta[\text{CH}_4]$ and $\Delta[T]$ are measured, and F_T is known, F_{CH_4} can be determined without the need for detailed information about α .

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conditions (stability classes A, B, or C) and downwind distances (100–3000 m), the horizontal width of a plume that is propagating according to Gaussian dispersion is ~ 20–50 % of the distance that it has traveled from its source. That is, the ratio of plume width to downwind distance is 0.2–0.5, where low wind conditions yield wider plumes (~ 0.5), and high wind conditions yield narrower plumes (~ 0.2). A plume observed 1000 m downwind of its origin, for example, is typically 200–500 m wide.

If the plume widths of two gases being measured downwind (e.g. CH₄ and N₂O) are much larger than the separation of their sources, the plumes will generally be co-dispersed, or spatially overlapping. Therefore the ratio of the distance between emission sources to the downwind transect distance must be less than 0.2–0.5 in order to achieve co-dispersion. If, for example, the separation between an N₂O tracer and a CH₄ source is 100 m, the downwind distance required to observe the onset of co-dispersion is > 500 m in high winds, and > 200 m in low winds. Alternatively, if local road access limits the downwind distance to 200–500 m, the N₂O tracer must be placed within 100 m of the suspected CH₄ emission source.

This same rule-of-thumb approach can be applied to cases where a nearby CH₄ source, such as a wellhead, may interfere with the FLER measurement at a G&P facility. In these cases, the downwind transect must be *close* enough that the interfering plume width is smaller than its separation from the G&P facility. For example, if the distance between a wellhead and facility is 50 m, downwind transects must be less than 100–250 m in order to isolate and exclude the wellhead plume from the FLER estimate.

When the second tracer is used as an internal standard, it can serve to quantify the uncertainty of the measurement. As will be shown below and in the Supplement, this uncertainty decreases when the two tracer plumes are spatially overlapping, as compared to cases where the plumes are separated. Because this precision reflects the uncertainty in the FLER, efforts are made by the study team to maximize the co-dispersion of methane and tracer plumes. In light of the above discussion, this can be achieved by attempting to place one or both tracers near the dominant suspected emission source at a facility, if one exists. When these conditions are met, the down-

wind distance required to observe co-dispersion is reduced, thereby increasing the instrumental signal-to-noise and further separating any possible interfering sources.

Initial placement of the tracers at opposite ends of the facility allows for early transects to identify suspected methane emission locations. In some cases, the observed methane plume will appear covariant with one of the two tracers, indicating that the dominant methane emitter is in the vicinity of that tracer. In many cases, however the methane plume is observed between the two tracer plumes. In this scenario, one (or both) of the tracers is typically moved such that its plume is spatially overlapping the methane plume. This process is iterated multiple times over the course of the measurement in order to yield plumes that exhibit high degrees of CH₄-to-tracer correlation.

While two tracers act as an internal standard in the horizontal plane, a complicating factor unique to some large facilities (e.g. processing plants and larger gathering facilities) studied here is the presence of flares and/or engine exhaust stacks, some of which can be over 20 m tall. Presented in the Supplement (SI) is a Gaussian plume and Brigg's equation analysis of the effect of a possible elevated CH₄ source on the measured emission rate (Beychok, 2005). A simple rule-of-thumb approach as used above is hampered here by both buoyant plume rise effects and plume reflection off of the ground. These calculations indicate that in strong wind conditions (i.e. high atmospheric stability classes, such as in winds above 5 m s⁻¹), the measured emission rate determined from close transects can be biased considerably low, depending upon the fraction of the emission coming from elevated positions. In wind conditions below 5 m s⁻¹, the dispersion is large enough that the bias is lessened to 0–50 %. To minimize this bias, plumes were obtained as far downwind as possible, and at several processing plants a tracer was emitted at an elevated position such as the side of a demethanizer column or stack. The impact of the bias upon the overall data set and resulting conclusions is discussed in more detail in the accompanying Measurements paper (Mitchell et al., 2014).

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tracer flow rates are controlled by Alicat[®] MC-series mass flow controllers. The mass flow rates are recorded via RS232 to an onboard computer in the vehicle.

In addition to the tracer gas flow systems, three portable meteorological stations (Airmar[®] 200WX) are deployed on tripods, sometimes serving as physical supports for the tracer release tubing. They are capable of recording GPS, true wind direction and wind speed with 1 s resolution. Each unit broadcasts that information wirelessly or via an RS232 cable at 1 Hz to a computer onboard the tracer release vehicle, where it is recorded and displayed for observation by the tracer release personnel to advise the mobile laboratory as needed. When considered in the context of tracer placement, the wind data can immediately inform mobile laboratory personnel whether a tracer is being deployed in an area onsite that is not well-ventilated. If this is the case (frequently due to the local wind currents near buildings) the tracer can then be moved to allow it to be carried downwind by the larger regional wind mass. This information also provides a crude wind field for later analysis to better understand the sources of error and uncertainty in tracer release methods.

Calibrations and ranges

In both laboratories, the inlet was periodically overblown (injected with a flow larger than the intake flow) with ultrazero air (AirGas[®] or Praxair[®]) to zero the instruments, typically every 15 min for 30 s. Because CH₄ and N₂O are present in background ambient air (1900 and 325 ppbv, respectively), zeroing events also serve as an approximate check of those instrument calibrations. Full instrument calibrations were performed several (4–5) times over the course of the measurement campaign using calibration standards. For these dilution calibrations, a controlled mass flow of calibration gas is released into a known zero-air flow, and the resulting mixture is overblown into the inlet. The mixture is changed by varying the calibration gas flow using either a series of critical orifices or mass flow controllers (Alicat[®] MC Series). The results of these calibrations changed less than 5% over the course of the campaign. The mass flow controllers onboard the

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and the locations of nearby facilities. Because of their size and scale, a full day was reserved to sample emissions from processing facilities.

5 Plume types and analysis methods

There are multiple ways in which downwind tracer plumes can be analyzed, depending upon the plume intensity and spatial overlap between the tracer and CH₄ plumes (Subramanian et al., 2014). Figures 2–5 show the four possible plume types observed during the G&P campaign.

5.1 Dual correlation

The ideal scenario occurs when the measurement transect is far enough downwind of the facility that the CH₄, N₂O, and C₂H₂ plumes are spatially overlapping. The resulting measurements of concentration vs. time exhibit a high degree of covariance between species, as shown in the top panel of Fig. 2. Analysis of these “dual correlation” plumes consists of plotting the concentration of one species vs. another, and performing a linear orthogonal distance regression (ODR) fit as shown in the bottom panels of Fig. 2. This regression analysis is performed for CH₄ vs. N₂O, CH₄ vs. C₂H₂, N₂O vs. C₂H₂, and C₂H₆ vs. CH₄. From these linear regressions, the slope indicates the ratio of concentrations of the two gas species (for use in Eq. 2), and R^2 indicates the degree of correlation. These values are recorded for use in determining whether the plume meets the acceptance criteria for the CH₄ emission rate to be considered valid. If the R^2 values derived from fits of CH₄ vs. N₂O, CH₄ vs. C₂H₂, and N₂O vs. C₂H₂ are all greater than 0.75, and the tracer ratio ($[\text{C}_2\text{H}_2] / [\text{N}_2\text{O}]$) is within a factor of 1.5 of the known tracer flow rate, the plume is a candidate for dual correlation analysis. The choice of acceptable R^2 and tracer ratio were based upon values at which further relaxation of the criteria would alter the uncertainty and accuracy of the FLER mea-

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surement (Mitchell et al., 2014). A discussion of the use of a factor for the tracer ratio criterion, as opposed to a deviation such as $\pm 50\%$, is presented in the Supplement.

5.2 Dual area

In certain circumstances, wind conditions along with local road access and intervening CH_4 sources prevent the ability to get far enough downwind for the tracer gas and CH_4 plumes to become spatially overlapped. However, transects may still be performed closer to the facility ($\sim 50\text{--}500\text{ m}$) such that all three species will be observed. As illustrated in the example shown in Fig. 3, under these circumstances correlation diagrams do not provide useful information about the ratio of species (bottom panels). In these cases a “dual area” technique is used, where the analysis must rely on the integrated area of each species’ plume over the time of the transect. Here, the deviation of the species’ mixing ratios from ambient conditions must be considered, rather than the raw integrated intensity. This point is particularly relevant for CH_4 and N_2O , whose ambient concentrations are ~ 1900 and ~ 325 ppb, respectively. In the analysis of the data, the baseline (non-plume) mixing ratio was determined by fitting a line through the average of several data points immediately before the plume transect began and the average immediately after the transect ended. The fit line was then subtracted from the data to yield a baseline-corrected plume. This accounted not only for background concentrations (e.g. 1900 or 325 ppb), but also any minor baseline drift that may have occurred over the course of the transect. The quality of the baseline fit was visually confirmed and corrected if it did not accurately represent the true baseline. For the plume to be considered a candidate for dual area analysis, the ratio of areas of the C_2H_2 and N_2O plumes must be within a factor of 2 of the known tracer flow rates.

5.3 Single correlation

In scenarios where the CH_4 mixing ratio was highly correlated with only one of the two tracers, a “single correlation” analysis was performed, as shown in Fig. 4. This

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approach corresponds to that originally used by Lamb et al. in early demonstrations of the tracer release method (Lamb et al., 1995). The need to use the single correlation technique can be the consequence of several possible measurement conditions: (i) one of the tracers is placed geographically close to the dominant emitter within the facility (e.g. a compressor or large fugitive source), (ii) the site is emitting a tracer species (i.e. C₂H₂ during certain combustion processes), forcing the measurement to become single-tracer only, or (iii) the plume transect is far enough downwind (frequently > 2 km) that one of the tracer species' mixing ratio is at or below the instrumental detection limit. In single correlation cases, correlation analysis is performed for both tracers, but only the well-correlated tracer serves to provide the true CH₄ emission rate. For a plume to be a candidate for single-correlation analysis, the R^2 value derived from the linear regression fit of CH₄ to one of the two tracers must be greater than 0.75.

5.4 Linear combination of tracer plumes

In certain circumstances, unique tracer placement, road access and wind conditions allow for intermediate-distance transects where the CH₄ plume profile is not well correlated with either individual tracer, but is well correlated with a linear combination of the tracer plumes, i.e.

$$\Delta[\text{CH}_4] = a \cdot \Delta[\text{N}_2\text{O}] + b \cdot \Delta[\text{C}_2\text{H}_2] \quad (3)$$

where a and b are multiplicative coefficients of the N₂O and C₂H₂ plumes, respectively. Such an example is shown in Fig. 5. This scenario is equivalent to performing two independent single-tracer measurement, where the plumes are overlapping in time. In these cases facility emission rates can be determined by performing a correlation analysis of CH₄ vs. ($a \cdot \Delta[\text{N}_2\text{O}] + b \cdot \Delta[\text{C}_2\text{H}_2]$) while adjusting the values of a and b in Eq. (3). The a and b values that provide the largest possible R^2 value in the fit are used to determine the CH₄ emission rate associated with each tracer. While the sum of these values serves as the facility level emission rate (FLER), the individual emission rates

when the mobile laboratory was onsite were compared to the GC data to confirm (or disprove) that the emission composition was in agreement with the GC data.

Both mobile laboratories measured ethane and methane at a 1 Hz sampling rate or faster, allowing for an accurate determination of the E / M ratio of individual plumes.

5 The E / M ratio for every downwind plume obtained in the campaign (determined using correlation analysis) was measured and compared to the known ratio from GC analysis (or measured onsite E / M ratio in cases where the GC data was unreliable). A detailed comparison between the observed E / M ratio and that from the inlet GC analysis is presented in the results section. A plume was only accepted for further analysis if
10 the observed ratio was within a factor of 1.5 of the known value. This criterion was suspended in cases where the facility itself was actively changing the ethane content (e.g. from a demethanizer), where the E / M ratio was varying across the facility, or when the downwind C₂H₆ mixing ratio was below the detection sensitivity limit.

15 Finally, under certain scenarios, a small number of plumes that would be rejected as described above are manually accepted during analysis. These exceptions are possible for one of several reasons. One is that the plume transect is far enough downwind that the tracer or CH₄ plume concentrations are near the detection limit of the onboard instruments. Under such a scenario the correlation analysis may reveal $R^2 < 0.75$, despite the plume being legitimate. Another possible reason for manually accepting
20 a plume is when the E / M ratio is variable across the facility, frequently due to the presence of a high emission point source such as a venting condensate tank. Because condensate tank emissions may exhibit an E / M ratio larger than that of the remainder of the facility, the observed downwind ratio may be variable, even on the timescale of a single plume.

25 6 Results

In this section, we present results from a number of case studies that illustrate the capabilities of the dual tracer release method.

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6.1 Gathering facilities

A gathering station serves as a point where multiple natural gas sources (wells) are combined to produce a high pressure stream of gas. These facilities typically include equipment such as inlet separators to remove liquid phase water and condensate (C_{5+}), if present, and systems for pipeline maintenance activities (e.g. “pigging”). Compression at these facilities is accomplished by a series of 1 to 20 individual compressors powered by electric motors, reciprocating engines or gas turbines with total engine powers ranging from 500 to 25 000 HP, depending on the inlet gas pressure and total gas throughput (Mitchell et al., 2014). Gathering stations also typically contain condensate storage tanks, produced water storage tanks, and other gas handling equipment including pneumatic valves (often powered by natural gas) and gas metering systems. If the gas has a high water content, glycol dehydration systems are also frequently present to dry the gas (Goetz et al., 2014; Kidnay et al., 2011).

There are three main sources of continuous emissions from these facilities. First, compressors can serve as significant sources of CH_4 via both fugitive leaks as well as through seals in the compressor housing. In the case of wet compressor seals, it should be noted that the primary emission route is due to absorption of methane into the seal fluid at high pressure, followed by exposure of the fluid to ambient pressure, where the methane is routed through a vent to atmosphere (EPA, 2006). Second, because the natural gas is typically under high pressure, fugitive and vented emissions may occur at the facility, including from continuous-bleed natural gas pneumatic devices, dehydration units, and a variety of flanges and valves. Third, methane slip (i.e. unburned methane in engine exhaust gases) through onsite combustion sources such as engines and turbines can be a source of CH_4 , depending upon a wide variety of combustion characteristics. The relative importance of this emission source to the FLER is discussed in the associated Measurements report (Mitchell et al., 2014) and in previous studies of combustion emissions in natural gas transmission and storage (Subramanian et al., 2014). Similarly, methane and other unburned hydrocarbons are

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present in flare emissions, and may vary greatly depending upon the flare combustion efficiency (Torres et al., 2012).

Some intermittent methane emission sources may also be found at gathering facilities, such as intermittent-bleed natural gas-driven pneumatic controllers, produced water tanks, and condensate tanks. Of particular importance to the associated Measurements paper (Mitchell et al., 2014), produced water and condensate tanks may *transiently* emit CH₄, C₂H₆ and higher hydrocarbons from thief hatches or other pressure relief valves attached to the tank. Because of the nature of the liquids stored in them, i.e. long-chain hydrocarbons, the ethane to methane ratio observed from a condensate tank can be much higher than the natural gas composition entering or exiting the facility. However, these units may sometimes also serve as venting release points for equipment onsite, in which case the E / M ratio will be very similar to that of the inlet stream.

An example of an emission rate measurement from a compressor station (C station) is shown in Fig. 6a. Similar to the example plume shown in Fig. 2, this plume as accepted as dual-correlation ($R^2 = 0.998$, tracer ratio error = 1.05, E / M ratio error = 1.4). In this case, the methane and ethane signals are strongly correlated with both tracers at a distance of 1600 m downwind of the facility. Note that inclusion of the CO and CO₂ in the analysis indicates that both of these gases are also being emitted from the facility, likely due to combustion. While this plume alone can provide an accurate determination of the FLER from the facility, even more information can be extracted by also investigating transects from only 100 m away, shown in Fig. 6b (a dual-area plume, with tracer ratio error = 0.7, E / M ratio error = 1.5). While such a close transect may not provide as precise of a FLER, we see from the figure that the CO and CO₂ signatures are coincident with only a fraction of the methane being emitted, and are not well-correlated with it. This indicates that some, but not all, CH₄ emitted at the facility may be associated with combustion. In this case, the remaining CH₄ emission is likely from other non-combustion sources onsite. At some facilities, such as that shown in Fig. 6c, CO and CO₂ are correlated with a distinct part of the CH₄ plume, indicating the

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directly from gathering facilities into transmission pipelines (and sometimes directly into distribution networks).

The initial process that is typically found at a gas-processing plant involves a continuation of the treatment types found in the gathering system of the region. At some facilities, the initial product will be a first cut at collecting natural-gas condensate, which is typically comprised of functionalized hydrocarbons above C_5 , using an inlet separator (if they have not been collected further upstream in the gathering network). Water may also be removed using glycol dehydration. Other trace contaminants are often filtered using a series of molecular sieve apparatus that are staggered for effective continuous regeneration. As discussed below, natural gas liquids (NGLs) are removed from the gas stream using either a cryogenic separation or separation based on solubility in lean oil (Kidnay et al., 2011). Additional details of this class of compounds and specific equipment used are discussed in the next section.

Due to the nature of the various processing steps and types of equipment found at processing plants, as well as the somewhat larger geographic scale they typically occupy, there are typically multiple methane emission points, with various co-emitted compounds. On the surface, this type of source is a direct challenge to the tracer release methodology given the constraint for the controlled tracer release to be as close to the emission source as possible. The following examples and discussion describe how these types of facility are quantified using the dual tracer methodology as well as using the nature of the co-emitted compounds to deduce the dominant emission sources.

The geographic scale of processing plants presents a challenge to the dual tracer flux ratio quantification given the constraints of wind direction and roadway access. Figure 9 depicts a pair of transects from a processing plant. Each transect was collected with the mobile lab maneuvering north to south. This is depicted by the rainbow bar in each of the two split time series (a) and (b) in the left hand panel and portrayed on the right hand panel with the relative distance (north vs. east). In the case where the transect was captured at the facility fence line (a), we see relatively high spikes in

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was 14.58, i.e. nearly entirely ethane. Therefore, this transect indicates that the pipeline is not a significant source of CH₄ emissions.

6.5 Comparison of C₂ content with operator data

In this study, the E / M ratio serves several purposes: (i) confirmation that a plume is from a target facility, (ii) elimination of plumes from neighboring facilities or biogenic sources, and (iii) distinguishing between different emission sources within a given facility. The quantification of a facility's methane emissions leverages (i) and (ii) above. Figure 11 shows a comparison between the measured E / M ratios at each facility and the operator-provided data on gas composition. Agreement is good overall, with a few outliers. Also shown in the figure are 95 % confidence limits on the measured E / M ratios. Large error bars in the facility average for E / M ratios are usually due to variations in the emission composition, since the error for any individual ratio measurement is low. The operator gas composition information was not always measured on the same day as the field testing. For gathering facilities, gas composition is periodically measured by gas sampling and subsequent third party analysis. For processing plants, gas composition data is typically acquired in real time at multiple locations at the facility. In either case, the gas composition exiting the gathering facility or processing plant may not always reflect the gas composition of the emission sources. This can be due to the E / M ratio changing as the gas moves through the facility, or from emissions from condensate/produced water tanks. This variety of equipment and processes at gathering facilities and processing plants explains much of the discrepancy between measured and operator E / M ratios, as compared to the transmission and storage study, where the composition of the gas does not vary during handling (Subramanian et al., 2014; Yacovitch et al., 2014). Table 3 outlines the minimum, median and maximum facility average E / M ratios divided by primary gas type. It should be noted that the classification by gas type is not rigid. That is, there may be multiple gas types other than the primary present at these facilities. The points in Fig. 11 are colored based on this gas classification. As noted above, coal bed methane facilities typically have the lowest E / M ratios.

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Conventional facilities sit somewhere in the middle, with the shale gas facilities split into several clusters. The shale gas is scattered about the plot, with some clustering associated with various geographic basins. The three main shale clusters observed in Fig. 11 (green points) correspond loosely to: the Denver (Denver-Julesburg), Permian (Eagle Ford and Delaware), and Appalachian basins (~ 12–23 %); the Anadarko (Mississippian Lime Gas play), Uinta (Natural Buttes) and Piceance basins (~ 4–6 %); and the Arkoma basin (~ 1 %). Other shale basins were also visited but the number of facilities for each of these basins is low.

7 Conclusions

Reported here is a detailed description of the measurement and analysis methods used during a field campaign to quantify methane emissions and emission sources from natural gas gathering and processing facilities. The campaign covered a wide range of geographic regions, basins, gas types and facilities. The measurement method used, dual tracer ratio, yielded facility-specific methane emission rates for 130 facilities. The field measurements were complemented by onsite infrared imaging and equipment surveys. The analysis technique applied to the data allowed for accurate determination of the emission rates using multiple downwind plume categories. Overall emission profiles, quantified by measuring CH₄, C₂H₆, CO, CO₂, C₂H₂, and N₂O frequently afforded an understanding of the unique chemical signatures associated with various natural gas gathering and processing equipment onsite. This paper provides a background and method description for additional work aimed at compiling the dataset (Mitchell et al., 2014) and developing an estimate (with uncertainty) for the total methane emissions from gathering and processing in the US (Marchese et al., 2014).

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Table 1. Instruments and sensitivities for measured species on Aerodyne and CMU Mobile Laboratories.

Instrument	Species Detected	Sensitivity
Aerodyne Mobile Laboratory		
Aerodyne Dual QCL	CH ₄	1 ppb
	C ₂ H ₂	200 ppt
Aerodyne Mini QCL	C ₂ H ₆	100 ppt
Aerodyne Mini QCL	N ₂ O	100 ppt
	CO	100 ppt
Li-Cor NDIR	CO ₂	500 ppb
Carnegie Mellon Mobile Laboratory		
Picarro CRDS	CH ₄	3 ppb
	C ₂ H ₂	600 ppt
Aerodyne Dual QCL	C ₂ H ₆	100 ppt
	N ₂ O	100 ppt
	CO	100 ppt

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Table 2. Plume analysis types, preference, criteria, prevalence, and variance.

Analysis Type	Preference	Criteria	# of plumes	Variance ($\sqrt{\text{Variance}}$)
Dual Correlation	1	<ul style="list-style-type: none"> – $R^2 > 0.75$: N₂O vs. C₂H₂, N₂O vs. CH₄, C₂H₂ vs. CH₄, C₂H₆ vs. CH₄ – Tracer ratio error < 1.5 – E / M ratio error < 1.5 	250	0.04 (0.2)
Dual Area	2/3	<ul style="list-style-type: none"> – $R^2 > 0.75$: C₂H₆ vs. CH₄ – Tracer ratio error < 2 – E / M ratio error < 1.5 	441	0.14 (0.37)
Single Correlation	3/2	<ul style="list-style-type: none"> – $R^2 > 0.75$: C₂H₆ vs. CH₄, Tracer vs. CH₄ – E / M ratio error < 1.5 	728	0.09/0.22 (0.3/0.47)
Linear Combination	4	<ul style="list-style-type: none"> – $R^2 > 0.75$: C₂H₆ vs. CH₄ 	16	–

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Table 3. Measured E / M ratios as a function of gas type at gathering and processing facilities. Minimum, median and maximum average measured ratios are noted. Offshore gas is not included here due to the small number of offshore facilities measured.

Gas Type	Measured E / M ratio			Count
	min	median	max	
Coal Bed Methane	0.00	0.014	0.045	8
Coal Bed Methane and Conventional	0.0057	0.018	0.031	4
Shale	0.0055	0.051	0.24	64
Conventional	0.012	0.068	0.22	37

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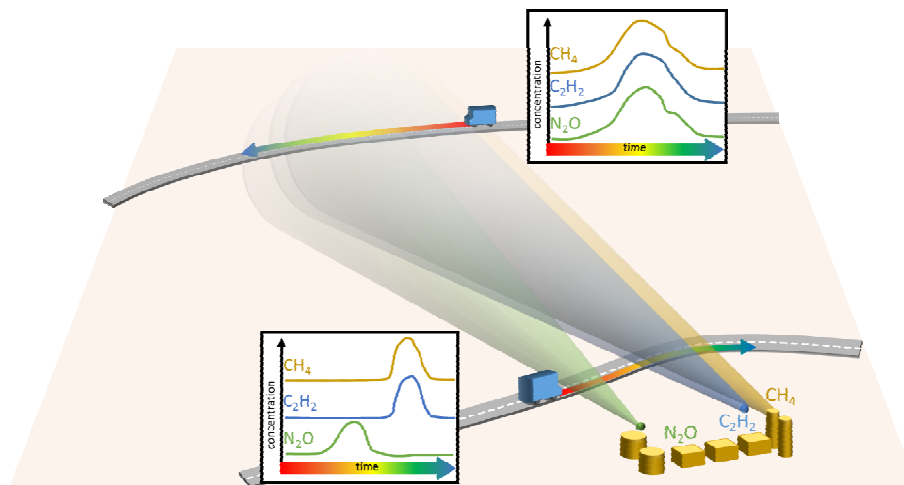


Figure 1. Schematic of dual tracer release technique. At distances far downwind (top), both tracers and CH_4 are spatiotemporally overlapped. At distances closer to the facility, the spatial position of the CH_4 plume relative to the two tracer plumes can indicate the location of an emission vector onsite with sub-facility resolution.

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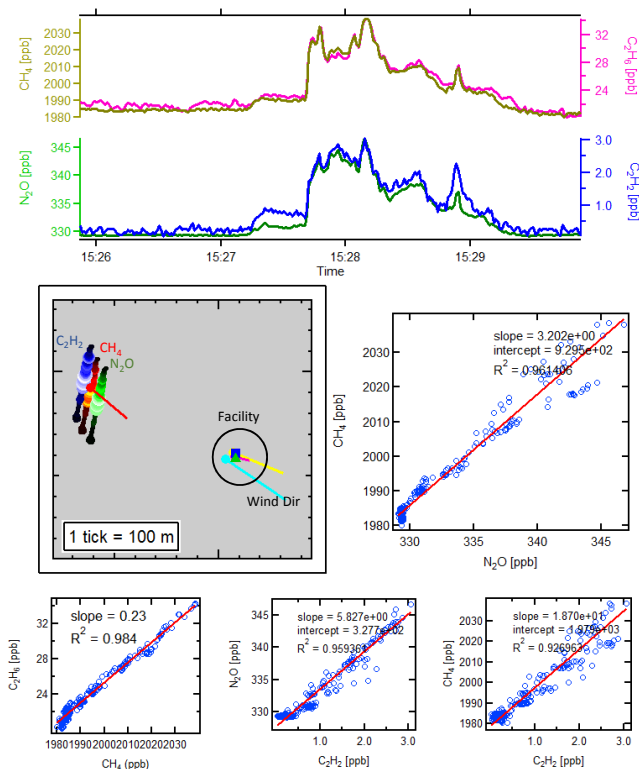


Figure 2. Example dual-correlation plume from a natural gas facility. Top panel: time trace of CH₄, C₂H₆, N₂O, and C₂H₂ concentrations, showing high temporal correlation. Center left panel: map of tracer location (right side) and transect location (left side) during the course of the plume. Red, blue, and green weighted lines correspond to CH₄, C₂H₂, and N₂O intensities, respectively, during the transect, spatially offset for clarity. Thin lines point into the wind at the mobile laboratory (red) and at the facility (light blue, pink, and yellow). Blue square and green triangle indicate C₂H₂ and N₂O release locations, respectively. Lower panels: correlation analysis of C₂H₆ vs. CH₄, N₂O vs. C₂H₂, CH₄ vs. C₂H₂ and CH₄ vs. N₂O.

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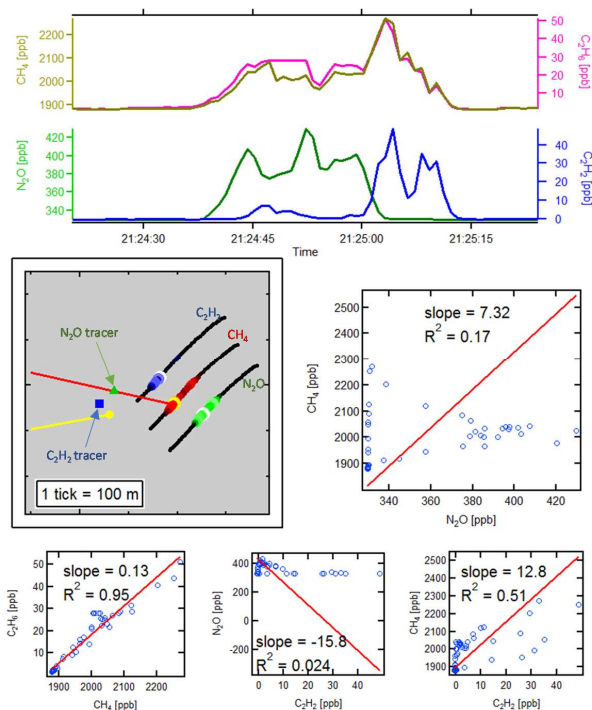


Figure 3. Similar to Fig. 2., illustrating dual area-type plumes. Top panel: time trace of CH₄, C₂H₆, N₂O, and C₂H₂ concentrations, showing high temporal correlation. Center left panel: map of tracer location (right side) and transect location (left side) during the course of the plume. Red, blue, and green weighted lines correspond to CH₄, C₂H₂, and N₂O intensities, respectively, during the transect, spatially offset for clarity. Thin lines point into the wind at the mobile laboratory (red) and at the facility (light blue, pink, and yellow). Blue square and green triangle indicate C₂H₂ and N₂O release locations, respectively. Lower panels: correlation analysis of C₂H₆ vs. CH₄, N₂O vs. C₂H₂, CH₄ vs. C₂H₂ and CH₄ vs. N₂O. Note the lack of correlation in lower left and center panels, indicating that the analysis must rely on an area method.

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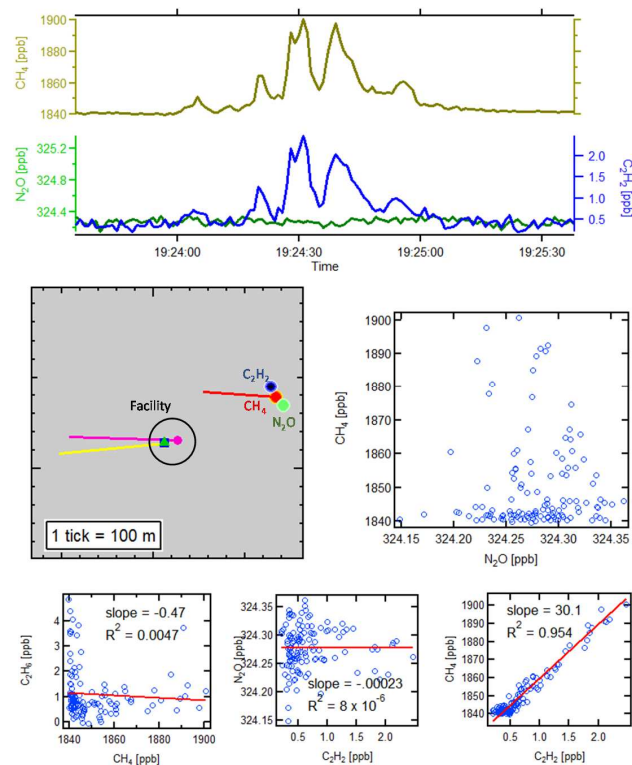


Figure 4. Example of a single-correlation plume (CH₄ correlation with C₂H₂). Top panel: time trace of CH₄, C₂H₆, N₂O, and C₂H₂ concentrations, showing high temporal correlation. Center left panel: map of tracer location (right side) and transect location (left side) during the course of the plume. Red, blue, and green weighted lines correspond to CH₄, C₂H₂, and N₂O intensities, respectively, during the transect, spatially offset for clarity. Thin lines point into the wind at the mobile laboratory (red) and at the facility (light blue, pink, and yellow). Blue square and green triangle indicate C₂H₂ and N₂O release locations, respectively. Lower panels: correlation analysis of C₂H₆ vs. CH₄, N₂O vs. C₂H₂, CH₄ vs. C₂H₂ and CH₄ vs. N₂O.

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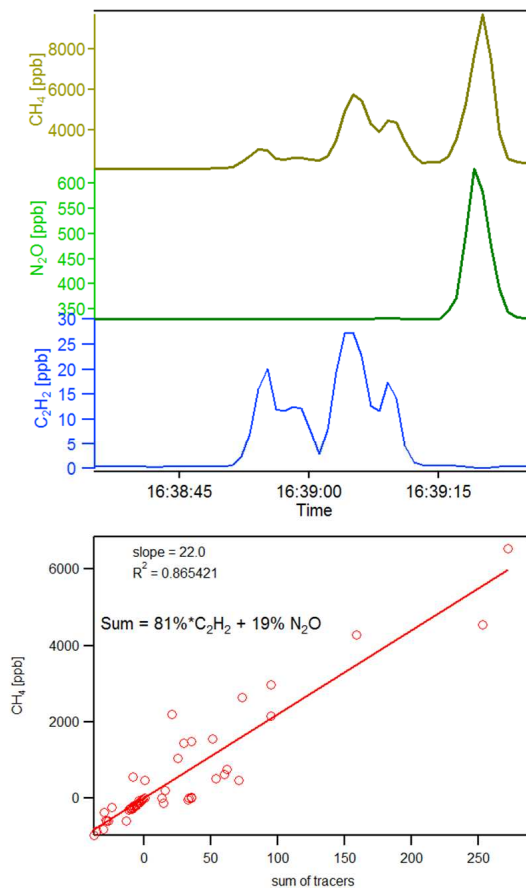


Figure 5. Example of analysis using a linear combination of tracer plumes. Note that N_2O and C_2H_2 are associated with different sections of the CH_4 plume (top). Adding the two tracer plumes in an 81%/19% combination yields a correlation diagram (below) with high R^2 value (0.87).

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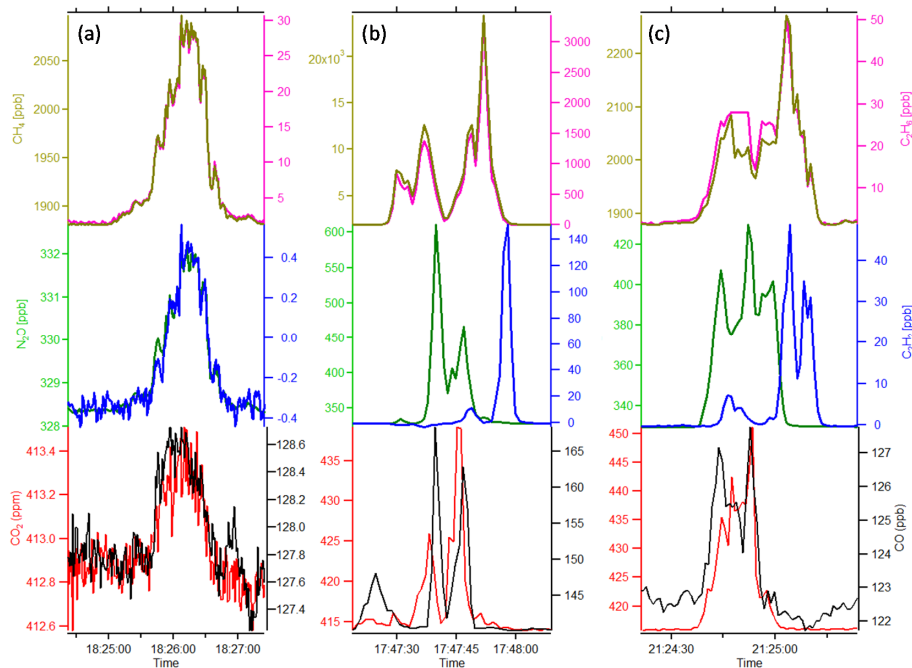


Figure 6. Three exemplary plumes from a gathering station. **(a)** Far-field plume (1.6 km) showing strong correlation between CH₄, C₂H₆, N₂O, C₂H₂, CO₂, and CO. **(b)** Close plume transect (100 m away) of same facility, showing loss of correlation and isolation of CO₂ and CO combustion products to a section of the facility. **(c)** Example of a close plume transect (200 m away) showing CO and CO₂ correlation with a component of the CH₄ trace.

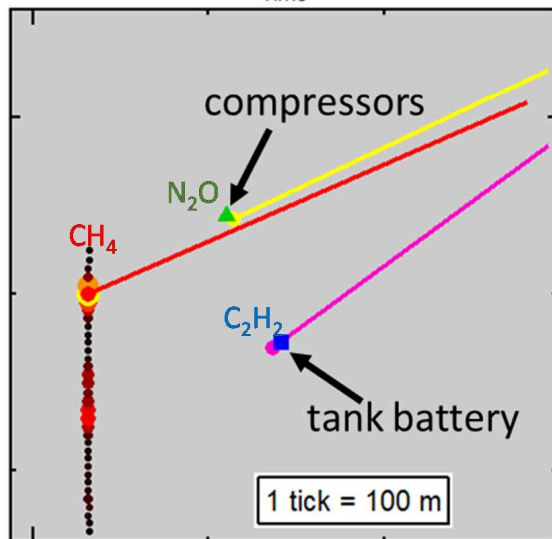
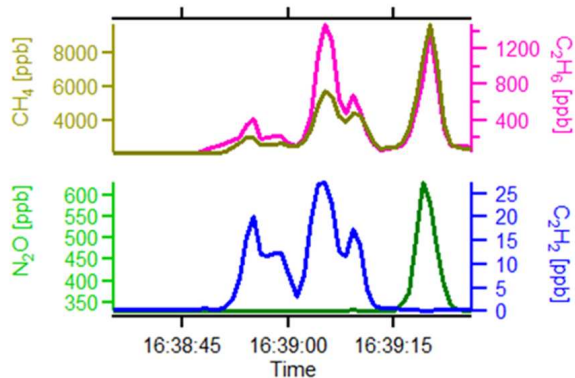


Figure 7. Example of varying E / M ratio during a close transect due to the presence of a condensate tank battery onsite. Note the $\sim 2\times$ decrease in the E / M ratio toward the end of the plume.

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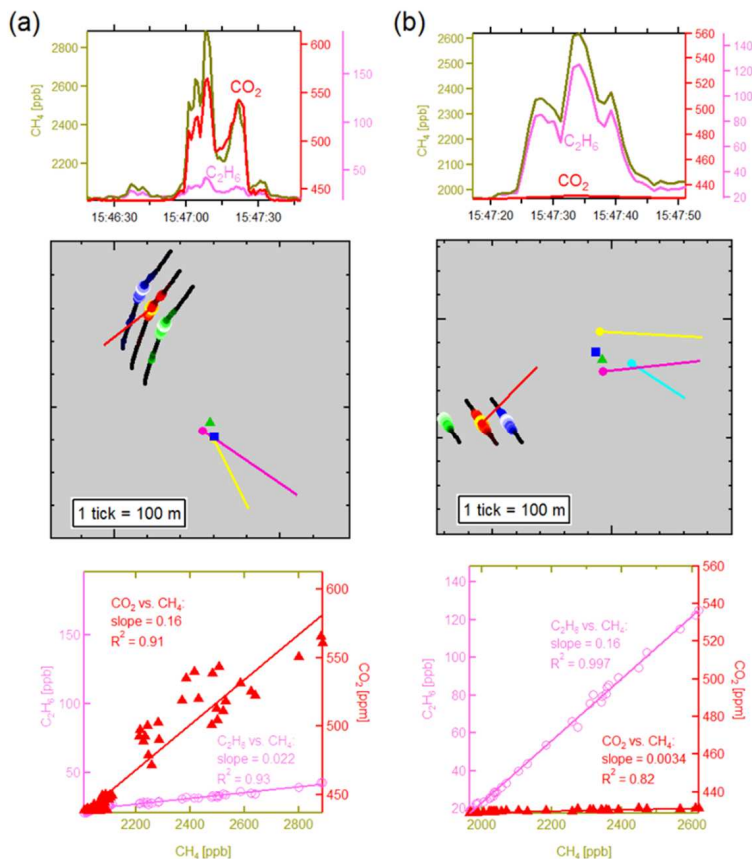


Figure 8. Example of differing CO₂ plume profiles as a function of gas play. **(a)** Emissions from a plant in a coal-gas region, with an amine scrubbing unit, showing significant CO₂ emissions, and **(b)** emissions from a gathering facility with no treatment in a shale gas region.

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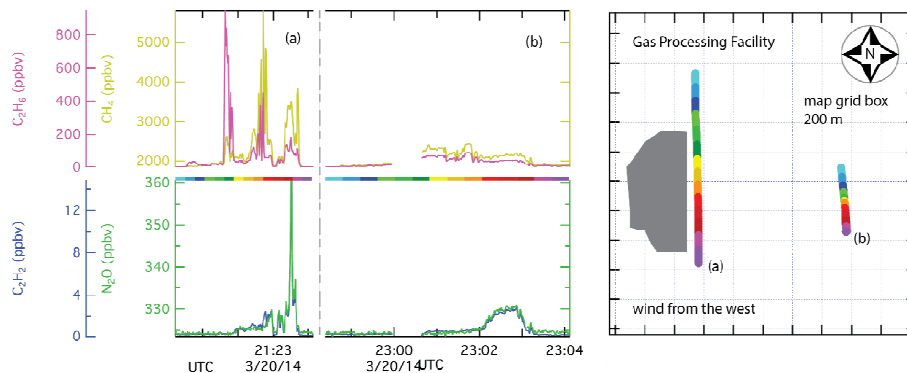


Figure 9. In the left hand panel, the time series for methane, ethane, nitrous oxide and acetylene are depicted for two transects, **(a)** and **(b)**. In the right hand panel, the geographic location is portrayed for the processing plant (grey) and the two transects **(a)** and **(b)**. See text for additional discussion.

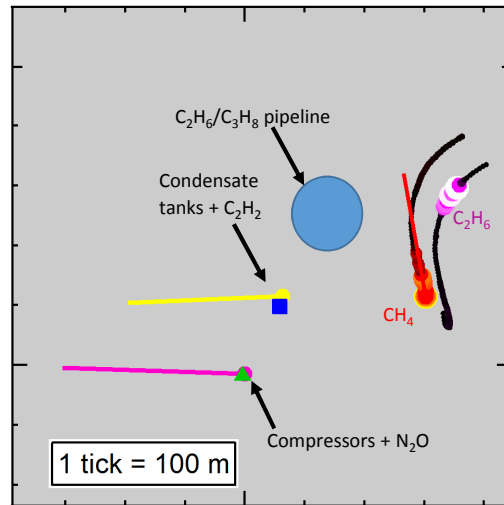
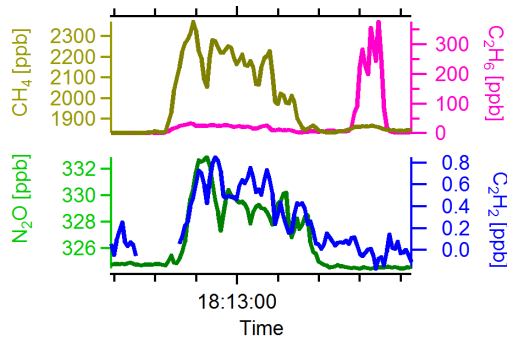


Figure 10. Downwind plume transect showing mixing ratio as a function of time (top) and a map (bottom). Tracer release locations are shown as a green triangle (nitrous oxide) and a blue square (acetylene). The plume transect is colored by methane mixing ratio (black to yellow). Ethane mixing ratio is also shown with a geographic offset. Wind vectors (pink, red and yellow) point into the wind.

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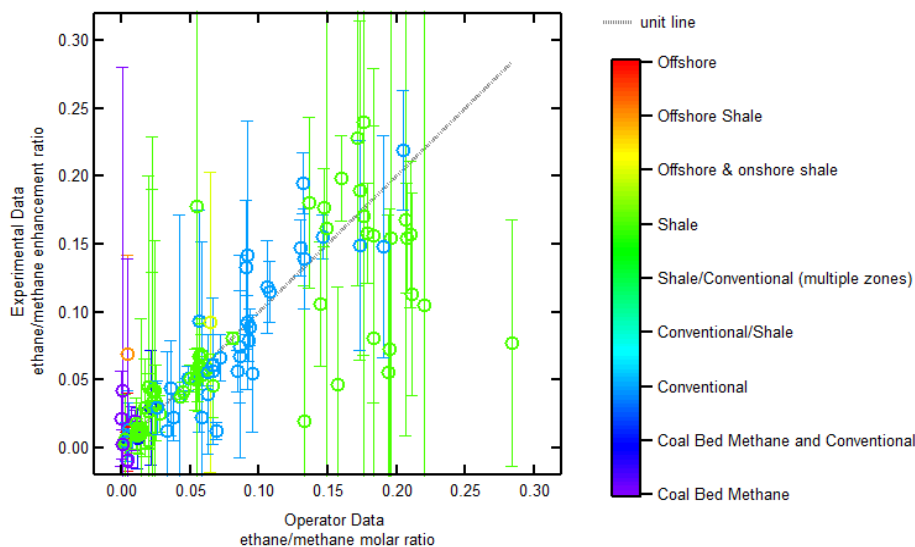


Figure 11. Comparison between measured ethane/methane ratio and operator data on gas composition. Error bars correspond to the 95 % confidence limits from the replicate experimental plumes. Points are also colored by the type of gas at each site. A line to guide the eye is drawn at a 1 : 1 correspondence between measured and operator data.

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