

## Response to Referee #1

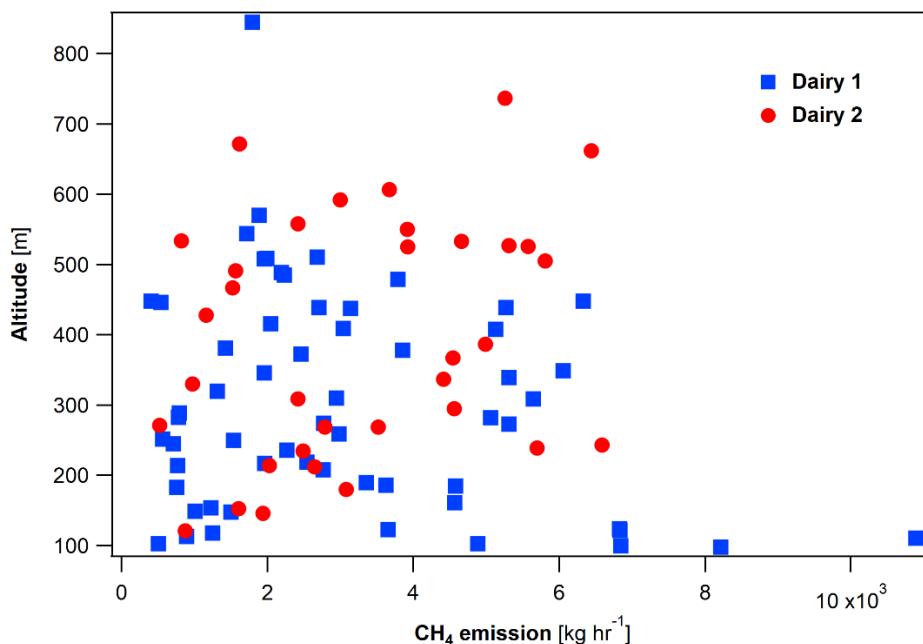
We thank the referee for their insightful comments and questions. We have responded below with referee comments in red and our response in black. We have also included a marked-up version of the revised manuscript to this response.

### General comments

Daube et al. compare three methods of estimating methane emissions from a dairy farm. Two are well-established in the scientific literature: tracer release using a mobile laboratory and mass balance using an aircraft. The third is tracer release using an aircraft. They find generally good agreement between emissions determined using the tracer release/aircraft combination and the other two methods.

This paper extends the work of tracer release using mobile laboratories into the realm of airborne measurements. As such, it is a relatively straightforward analysis comparing the two methods. The only thing I find lacking is a more in-depth discussion of the added complexity of the additional dimension, the vertical, that the aircraft can probe compared to the mobile laboratory. I think a plot of altitude vs. the CH<sub>4</sub>/ethane ratio might be informative. The authors briefly described this in the first paragraph of page 7, but I think a visual presentation might be better.

We agree that a plot of emissions by altitude would enhance discussion of the subject. The plot below uses all the plumes from each dairy to relate aircraft altitude to CH<sub>4</sub> emission rates. Emissions between 0 – 6,500 kg day<sup>-1</sup> appear to be randomly distributed between 100 – 600 m at each site. A couple outliers show higher emission rates at low altitudes and a few others are scattered across a range of emission rates for higher altitude observations. We have added this figure (Fig. 6) and relevant discussion to Section 4.1.



I also question the usage of a linear regression because of the need to align the two plumes. The tracer is presumably a point source, but a manure lagoon might be meters or tens of meters wide. I would expect sources such as this to have much broader plumes than the tracer release. Or worse yet, there could be two separate methane plumes, but only one ethane plume. The only way to properly account for this is by taking the integral. For example, what is the ratio of the integrals for the transect shown in Figure 2?

The reviewer is correct that we failed to discuss this important alternative analysis. We have addressed this concern by performing a reanalysis of the dataset using integration of plumes to determine emission rates. We acknowledge the mismatch of point source release being used for area source representation and recognize the need to seek different release methods for future similar studies of area sources – line source, area source via a grid arrangement, etc. We had employed multiple tracers and relied on greater sampling distances in the original study to obtain better tracer representation as noted in the paper, but obviously those factors don't contribute to this analysis.

In general, we look at discrepancies in line shape as a first indicator of poor tracer representation of a source. In Figure 2, the plumes of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> have similar structure with no unaccounted for CH<sub>4</sub> peaks nearby, signifying adequate co-dispersion and tracer representation for this event. When we perform linear regression, we match offset plumes in time (within a reasonable time range) based on multiple linear regression tests before determining the optimal timing and resulting correlation. The ratio of the integrated areas for the enhancements shown in Figure 2 results in an emission rate of 223 kg hr<sup>-1</sup> for a tracer release rate of 15 slpm. Using the slope of the linear regression, this emission rate is calculated to be 203 kg hr<sup>-1</sup> at the same tracer release rate.

During the original analysis, we had concerns regarding the potential bias in selecting a baseline, especially in cases of low signal-to-noise. For example, in determining a baseline for the C<sub>2</sub>H<sub>6</sub> plume from Figure 2, altering one side of the baseline from the ideal case by 50 ppt resulted in a 20 kg hr<sup>-1</sup> change in emission rate. Using slopes from linear regression for these plumes provided a systematic unbiased approach for relating these enhancements of CH<sub>4</sub> and tracer gas. However, linear relationships can be misleading if taken at face value (i.e. high R<sup>2</sup> value) and modelling of data via linear slopes need to be considered in the context of the entire enhancement event. Analysing the entire dataset again using integration compared quite favourably to our original analysis. This serves as a valuable check on the use of linear regression for this application, as discrepancies between the two methods could imply weak co-dispersion or misleading correlations.

We have replaced values in Table 2 with those determined by integration in the reanalysis. Three plumes were removed from Dairy 2 and two were removed from Dairy 1 observations keeping in accordance with quality control described in Section 2.3.

In Table 2, why do the emissions from Animal Housing and Liquid Manure add up to more than the Whole-site emission?

Emission estimates of Dairy 1 using the tracer release method via a ground-based mobile laboratory are presented in Table 2 for the overall site and two major sources from Arndt, et al., 2018. Whole-site emission rates fall within the uncertainty of the summed source emission rates. In ranges based on the 95% confidence intervals, the whole-site emission rates range 6,349 to 7,611 kg CH<sub>4</sub> hr<sup>-1</sup>. Animal housing emission rates range 1,790 kg to 3,412 kg CH<sub>4</sub> hr<sup>-1</sup>. Liquid manure emission rates range 5,415 to 6,573 kg CH<sub>4</sub> hr<sup>-1</sup>.

### Specific comments

The authors should be consistent with their use of the term “sub-source”. First, they should probably define it, because I’m not sure it’s a word. Second, I would prefer they simply use the word “source”, and remind the readers that a dairy farm is actually a combination of many different sources: cows, manure, settling cells, etc.

We have replaced all mentions of the term “sub-source” with “source”, as suggested.

For Figures 2 and 3, I would clearly label (or title) these as Dairy 1 and Dairy 2, instead of burying this information in the caption.

Corrected.

p. 1, line 25: is that 60% of anthropogenic CH<sub>4</sub> emissions?

Yes, we have added “anthropogenic” to this sentence (Figure 4, page 56 of CARB, et al. 2017).

p. 2, line 27: how “low and close” did the aircraft fly?

We have added average distance altitude values from flights at both sites. Section 3.1 contains details about maxima and minima altitudes.

p. 6, line 8: what altitude was the aircraft at for the transect described here?

The average altitude of the aircraft was 428 m AGL during this 34 s plume.

p. 6, line 28: I think this section needs a little introduction, instead of immediately delving into the ethane background. Which dairy is this? Introduce whatever Figure 4 is before referring us to it.

We have introduced Figure 4 and noted which dairy farm is being shown in the paragraph as well as in Figure 4 (Dairy 2). We have also changed the order of the paragraphs in Section 4.1 to help the flow of this section and add clarity.

Figure 4 and p. 6, line 31: Based on horizontal wind speed, how long would you expect it to take to see the signal from the aircraft after the tracer had been released?

Based on the average wind direction (from the NW) and horizontal speed (4.2 m s<sup>-1</sup>) from 10:39 AM PDT (start of tracer release) to 11:00 AM PDT (first spike of C<sub>2</sub>H<sub>6</sub>), we would expect to begin

seeing tracer gas after ~6 minutes at a distance of 1.6 km (from release point to the intersection between the circular transect and wind direction). We saw the first spike shortly after when would have expected, around 11 minutes after beginning release.

### Technical corrections

p. 2, line 12: should that reference be for “Grainger”?

Yes, thank you for catching that typo.

p. 3, line 1: since ethane isn’t emitted from dairy farms, I suggest saying “. . . emitted from within dairy . . .”

Agreed, we have added the word “within” to the sentence.

p. 4, line 15: add “its” to read “. . . and its long atmospheric lifetime.”

Corrected.

p. 4, line 21: change to “molar enhancement ratio” and add comma after “The molar ratio,”

Corrected.

p. 5, line 16: replace “generally” with “with speeds”

Corrected.

p. 5, line 26: why say “appear to represent”? I would assert that they do represent an entire site.

Agreed, we have changed language in this paragraph to reflect this comment.

p. 5, line 32: “plumes” isn’t really the noun you should use in this sentence. You are talking about emission estimates here.

Agreed, we have changed “plumes” to “emission estimates”.

p. 6, line 4: instead of “recording data”, perhaps say “sampling the same plume”

Corrected.

p. 6, line 6: add comma after “22”

Corrected.

p. 7, line 30: I suggest “sparse number” instead of “sparse amounts”

Corrected.

170 p. 8, line 16: I suggest “circling at a particular radius”

Corrected.

175 p. 9, line 13: I suggest “independent” instead of “stand-alone”

Corrected

p. 9, line 19: remove “of”

180 Corrected.

p. 13, line 3: I suggest “the wind is carrying the plume across the site. . .”

Corrected.

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## Response to Referee #2

215 We would like to thank the referee for their helpful comments and kind words. We have responded below with referee comments in red and our response in black. We have also included a marked-up version of the revised manuscript to this response.

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220 In this paper, the author mix up a well-known technique of estimating emission, the tracer release method with airplane measurement usually used for mass-balance approach. They show that using an aircraft to do the tracer method is feasible even when the original campaign was not exactly designed for it and that the validity and precision of the results could be improved if the airplane flight paths are adapter to the tracer ratio method. This is an interesting and well-written paper and we recommend publication after minor changes.

P1 119-21: The last comment is not really necessary or should be reformulated.

230 We agree, and have removed the last sentence of the abstract.

p3 Instruments for the ground-based transects are described but the one that data are used from (except the TILDAS) are not detailed. please, add some details about instrumentation for CO<sub>2</sub>, wind, ... for the airplane as well.

235 We have added detail about instruments used on the aircraft during this campaign: “Scientific Aviation equipped an aircraft with a Picarro (Santa Clara, CA) G2301-f cavity ring-down spectrometer (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O), Vaisala (Helsinki, Finland) HMP60 Humidity and Temperature probe, and Hemisphere VS330 GPS Compass used for positioning and calculating wind velocity (Conley et al., 2014).”.

240 p6 12 It would be good here already to say why there is no estimate for the manure as we are expecting it from the previous sentences.

245 We agree, and have added a sentence to address the lack of a manure estimate: “Measurements of manure emissions were not compared with established techniques due to uncertainty in representation of the source by the tracer gas.”.

Table 1: there is no \* that relates to the comment below the table. Either erase it or modify the table accordingly

250 We have associated the \* to the “Days Spent” column in Table 1 to indicate that one of the days spent on-site releasing tracer gas was not accompanied by a measurement flight.

Figure 2: Please use SI units and not knots

255 We have changed the unit in the caption of Figure 2 to be in “m s<sup>-1</sup>” and converted the value accordingly.

# Using the tracer flux ratio method with flight measurements to estimate dairy farm CH<sub>4</sub> emissions in central California

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**Abstract.** Tracer flux ratio methodology was applied to airborne measurements to quantify methane (CH<sub>4</sub>) emissions from two dairy farms in central California during the summer. An aircraft flew around the perimeter of each farm measuring downwind enhancements of CH<sub>4</sub> and a tracer species released from the ground at a known rate. Estimates of CH<sub>4</sub> emission rates from this analysis were determined for whole sites and ~~major sub~~-sources within a site (animal housing ~~and~~ liquid manure lagoons). Whole-site CH<sub>4</sub> flux rates for each farm, Dairy 1 (~~6,1085,850~~  $\pm$  ~~821793~~ kg CH<sub>4</sub> day<sup>-1</sup>, 95% confidence interval) and Dairy 2 (~~4,0183,699~~  $\pm$  ~~456685~~ kg CH<sub>4</sub> day<sup>-1</sup>, 95% confidence interval), closely resembled findings by established methods: ground-based tracer flux ratio and mass balance. Individual Sub-ssource emission rates indicate a greater fraction of the whole-site emissions come from liquid manure management than animal housing activity, similar to bottom-up estimates. Despite differences in altitude, we observed that the tracer release method gave consistent results when using ground or air platforms.

## 1 Introduction

Methane (CH<sub>4</sub>) released into the atmosphere as a result of agricultural activity, such as enteric fermentation and anaerobic digestion, significantly contributes to overall greenhouse gas emissions in the United States (USEPA, 2017). The California Air Resources Board (CARB) attributes approximately 60% of recent anthropogenic CH<sub>4</sub> emissions in California to agriculture, with 45% of CH<sub>4</sub> emissions directly related to dairy farm activity for 2013 (CARB, 2017). Reduction strategies proposed by CARB seek to ~~reduce-lower~~ California's CH<sub>4</sub> emissions to 40% below 2013 rates by 2030 (CARB, 2017), thereby emphasizing the need for accurate methods to directly quantify the contribution of different CH<sub>4</sub> sources within agricultural operations. Estimates of CH<sub>4</sub> emissions due to dairy livestock can be calculated using inventory emission factors combined with activity data on animal populations, animal types, and details about feed intake in a particular country (IPCC, 2006). Other methods to estimate CH<sub>4</sub> emissions from ruminants involve direct atmospheric measurements. Emissions from dairy farms have been estimated in the Los Angeles basin, California, using downwind airborne flux measurements (Peischl et al., 2013). Farm-scale measurements of CH<sub>4</sub> have been made using a variety of techniques and instruments, such as open-path infrared spectrometers (Leytem et al., 2017), tunable-infrared direct absorption spectroscopy (Hacker et al., 2016), and column measurements employing solar absorption spectrometers with comparisons to cavity ring-down spectrometers (Viatte et al., 2016). Several studies of various CH<sub>4</sub> sources (e.g. natural gas pipelines, landfills, dairy farms) assert that

inventory-based calculations tend to underestimate emissions compared to atmospheric observations and modelling (Brandt et al., 2014; Miller et al., 2013; Peischl et al., 2013; Trousdell et al., 2016).

Atmospheric studies have often used specific gases as tracers to distinguish a sample of interest from background conditions or interferences. Tracer gases released at known rates have been employed in experiments looking at chemical transport (Ferber et al., 1986), dispersion (Record and Cramer, 1958), source allocation (Lamb et al., 1995; Mønster et al., 2014), and model verification (Sykes et al., 1983) using mobile laboratories (Wang et al., 2009; Yacovitch et al., 2015), radiosondes, sampling towers, and ground-based equipment. Application of tracer gases in agricultural studies have involved insertion of a sulfur hexafluoride ( $\text{SF}_6$ ) permeation tube into the rumen of a cow with subsequent collection of time-integrated breath samples (Graininger et al., 2007). Inverse-dispersion techniques have employed line-source releases of  $\text{SF}_6$  within a dairy farm combined with open-path measurements to understand whole-site emissions (McGinn et al., 2006). Release of a tracer gas directly into the atmosphere, 2-3 m above ground level, can be used to determine and distinguish  $\text{CH}_4$  emissions from various sources within a site (Roscioli et al., 2015). This study quantifies  $\text{CH}_4$  emissions using the well-established tracer flux ratio method at two dairy farms over the course of 8 summer days (Lamb et al., 1995; Roscioli et al., 2015). Controlled releases of tracer gas from various areas on each farm mixed with site-derived emissions and were observed by an instrumented aircraft and mobile laboratory (Arndt et al., 2018). Using this technique provided the flexibility to estimate entire dairy farm emissions and; apportion emissions among sub-sources (animal housing, liquid manure management, etc.); and to determine methane:carbon dioxide ( $\text{CH}_4\text{:CO}_2$ ) ratios on multiple scales.

Uncertainty in measurements from low-flying airborne studies has been attributed to the need to extrapolate results below the minimum safe flight heights (~150 m) as regulated by the Federal Aviation Administration (Conley et al., 2017; Hacker et al., 2016). Prior to this study, Aerodyne Research, Inc. (ARI) performed controlled ground-releases of ethane ( $\text{C}_2\text{H}_6$ ) in Colorado and Arkansas, while Scientific Aviation (SA) made measurements above in a similar aircraft to the one used in this study (Conley et al., 2017). The original release rate of  $\text{C}_2\text{H}_6$  was estimated via a refined mass balance technique, with a +2% difference observed during tests in Colorado (50 laps flown) and +24% difference in Arkansas (19 laps flown) as described in Conley et al. (2017). These releases did not correspond to any  $\text{CH}_4$  source (natural gas site, dairy farm, etc.), but demonstrated the feasibility of using a low-flying aircraft to successfully quantify flow rates from controlled tracer gas releases. Using tracer flux ratio in this study, we again utilized the aircraft to detect emitted tracer gas and then compared with dairy farm emissions to evaluate  $\text{CH}_4$  emission rates.

Theis field study was originally focused on estimating  $\text{CH}_4$  emissions from dairy farms and distinguishing on-site sources using established techniques (Arndt et al., 2018). An intentional effort was made to align measurement time windows of the mobile laboratory and aircraft for the purpose of inter-comparison between the tracer flux ratio and mass balance methods. As a result, the aircraft was exposed to several hours of ground-released tracer gas. Due to this overlap in time, we were able to (1) further assess the viability of observing enhanced concentrations of a ground-released tracer gas from an aircraft at low flow rates, (2) compare  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  enhancements emitted from within dairy farms via tracer flux ratio to determine emission rates, and (3) directly compare the application of tracer flux ratio methodology to simultaneous ground and airborne measurements of the same airmass.



## 2 Project description

### 2.1 Participants

In a collaborative effort, SA and ARI attempted a flight-based tracer release experiment to quantify CH<sub>4</sub> emissions from two dairy farms in central California. This study reanalyzes data collected as part of an Environmental Defense Fund coordinated project that occurred in June 2016 (Arndt et al., 2018). Both groups performed established techniques in the field to estimate dairy farm emissions. ARI employed tracer flux ratio methodology with two tracer gases and a mobile laboratory, while SA conducted a mass balance experiment from a light aircraft.

Aerodyne Research, Inc. (ARI) drove ground-based transects in a mobile laboratory (miniature Aerodyne Mobile Laboratory, “minAML”) equipped with highly precise Aerodyne Tunable Infrared Laser Direct Absorption Spectrometers (TILDAS) measuring a variety of species (CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>6</sub>, ~~and CO<sub>2</sub>, and H<sub>2</sub>O~~). A LI-COR (Lincoln, Nebraska, USA) non-dispersive infrared (NDIR) instrument (Model 6262) measured CO<sub>2</sub> and H<sub>2</sub>O. Meteorological and positional data (wind, temperature, relative humidity, barometric pressure, and GPS) were collected at all tracer release sites and on the vehicle, using multiple AIRMAR (Milford, NH, USA) 200WX WeatherStation® instruments and a Hemisphere (Scottsdale, AZ, USA) V103 GPS Compass. To minimize drift and maintain accurate baseline values on the TILDAS instruments in the minAML, a valve sequence enabled overblowing of the inlet with ultra-zero air every 15 ~~minutes~~ for 45 s (including cell purging). Scientific Aviation equipped an aircraft with a Picarro (Santa Clara, CA) G2301-f cavity ring-down spectrometer (CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O), TILDAS (C<sub>2</sub>H<sub>6</sub>, CH<sub>4</sub>, H<sub>2</sub>O) Vaisala (Helsinki, Finland) HMP60 Humidity and Temperature probe, and Hemisphere VS330 GPS Compass used for positioning and calculating wind velocity (Conley et al., 2014). Since SA also had a TILDAS on-board measuring C<sub>2</sub>H<sub>6</sub> during these times, it was possible to treat these flights as a tracer release experiment similar to that performed with the ground-based equipment. A full description of the equipment used during this project can be found in the Supplementary Information of Arndt et al. (2018).

~~The initial study prioritized overlapping time periods of data collection between all participants for the sake of inter-comparison. Doing so ensured that ARI would be releasing tracer gas during SA flights. Since SA also had a TILDAS on-board measuring C<sub>2</sub>H<sub>6</sub> during these times, it was possible to treat these flights as a tracer release experiment similar to that performed with the ground-based equipment. During each flight, SA recorded multiple species (CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, O<sub>3</sub>, CO<sub>2</sub>, and H<sub>2</sub>O), atmospheric conditions (horizontal winds, temperature, and RH), and positioning information (altitude, latitude, and longitude).~~

During this study, the aircraft flew low and close to the sites, at an average distance of ~900 m and an altitude of ~325 m. Each ~~site~~<sup>which</sup> had a combination of spread-out point source emitters (cows) and large open area sources (anaerobic lagoon and settling cells). SA conducted 11 flights over 6 days, usually flying twice a day, in the late morning and mid-afternoon. Flights typically lasted 1-2 h for a given farm, flying in spirals looping around the perimeter of the farm-animal housing and manure management areas. ARI measured for 3 days at Dairy 1 and 5 days at Dairy 2. The mobile lab drove at several different times of day for each site, trying to capture any diurnal effect, but always overlapping-overlapped with the aircraft at least once a day.

## 2.2 Tracer release

Tracer gases, ethane and acetylene, were released from ground-based tripods (2-3 m high) at a variety of locations on the dairy farms with the intention of co-locating with known emission sources (animal housing, anaerobic lagoons, settling cells, etc.). Tracers were used ~~in an effort~~ to distinguish and ~~to~~ quantify sources, by positioning them within each respective emission area. Often, each tracer was released at a single point from each major ~~sub~~-source, typically the liquid manure management (anaerobic lagoon and settling cells) and animal housing areas (barns and lots). For this study, only the position and release rate of  $C_2H_6$  ~~ethane~~ is relevant (~~Fig. 1~~). Release rates of  $C_2H_6$  ranged from 10–40 slpm throughout the project (averaged 15 slpm). A schematic of tracer release being performed at a dairy farm is shown in Fig. 1. Detailed descriptions of the tracer flux ratio technique used during this work can be found in Arndt et al. (2018) or more generally in Rosicoli et al. (2015). In summary, tracer gas released close to a source produces a plume that experiences the local wind dynamics and meteorological conditions akin to the nearby emission of interest, thereby proving a representation of those emissions. A plume is considered to be a co-located enhancement above ambient concentrations of  $CH_4$  and tracer gas. Active tracer release overlapped with on-site flight transects for approximately 11 h during this week-long project. Exact timing of the overlap between the release of  $C_2H_6$  and sampling periods by the aircraft is shown in Table 1.

Ethane was selected over other gases due to the lack of potential interference with nearby sources and its long atmospheric lifetime. At one of the two sites,  $C_2H_6$  from a small well pad (~2.5 km from closest point of farm) could be observed on the ground at close distances. This interference was characterized and eliminated using its measured  $C_2H_6:CH_4$  ratio (Yacovitch et al., 2014) in combination with wind direction and farm layout.

## 2.3 Data quality assurance

Analysis of tracer flux data involves comparing slopes or areas of enhancements between tracer gas and site  $CH_4$  emissions. Linear regression of the time-aligned  $CH_4$  and  $C_2H_6$  results in a molar enhancement ratio ( $CH_4:C_2H_6$ ). The molar enhancement ratio, scaled by the amount of tracer gas released, determines a  $CH_4$  emission rate for the specific plume encounter. Area analysis compares integrated plumes of  $CH_4$  and  $C_2H_6$ , particularly necessary during close transects when plumes do not temporally or spatially co-align. Both analysis methods were performed on this dataset and are discussed in further detail in Section 3.2. -Due to the speed of the aircraft (typically  $\sim 65 \text{ m s}^{-1}$ ), observations of plume emissions were brief. On average, identified plumes lasted 12 ~~seconds~~ (8 s for Dairy 1; 15 s for Dairy 2), not including a significant amount of time collected before and after enhancements to ensure accuracy of baseline calculations during analysis. ~~On a plume by plume basis, the integrated area method proved inconsistent for many cases, so linear regression was used exclusively (Rosicoli et al., 2015).~~

Prior to analysis, all data had appropriate calibration factors applied, correcting minor deviations in flow ~~control~~ rate by mass flow controllers and instrument performance for specific species. Instrument calibrations occurred in the field at several times during this campaign using mixed-gas standards diluted with ultra-zero air. Distance between tracer release locations and aircraft position was determined using the basic trigonometry. Uncertainties for emission rate estimates are determined as 95% confidence intervals.

Plumes observed by the aircraft were included in the analysis after meeting certain criteria. Requirements included: tracer gas flowing on site for more than 10 minutes prior to observation, correlated plumes of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> based on high coefficient of determination from a least-squares fit ( $R^2 > 0.5$ ), and positive enhancements above baseline for CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>. After meeting these standards, each plume was viewed and additional conditions were manually considered: wind direction and speed (as recorded on the aircraft and on-site), duration of the enhancement, validity of the linear regression fits, quality of calculated baseline for integration purposes, location of the aircraft relative to the sources, and correlation between CH<sub>4</sub> and other species (CO<sub>2</sub>, CO, C<sub>2</sub>H<sub>6</sub>) indicating interferences or source allocation.

### 3 Results

#### 3.1 Flight conditions by site

While flying transects around each site, plumes of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> were observed as frequently as once per minute. Short-lived enhancements ranged hundreds of ppb for CH<sub>4</sub> (typically ~200-300 ppb) and sub-ppb for C<sub>2</sub>H<sub>6</sub> (typically ~0.5-1.5 ppb). Figure 2 depicts an example plume event during a transect at Dairy 1 with correlated enhancements of CH<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> observed as the aircraft passed to the SW of the site.

At each dairy farm, the plane gradually flew a sequence of stacked circles around the facility with an average radius of ~900 m depending on the ratio of the strength of the horizontal wind to the surface heating (Conley et al., 2017.) At Dairy 1, flights went as low as 79 m above ground level (AGL), while achieving a maximum altitude of 1,244 m AGL. Fly-overs at Dairy 2 went even lower, with minima between 33 and 56 m AGL, and consistently reached heights of ~550 m AGL. Flying at low altitudes improved the signal-to-noise ratio for C<sub>2</sub>H<sub>6</sub>, helping to partially compensate for the relatively low release rates. Wind direction varied at Dairy 1 between the morning (NW) and afternoon (SW), generally with speeds building in strength throughout the day (~3 – 4.5 m s<sup>-1</sup>) as is common in the Central Valley due to the diurnal thermal forcing of the vast mountain-valley circulation (Zhong et al., 2004). Dairy 2, situated farther into the San Joaquin Valley, experienced consistent NNW winds that were sampled on days with a slightly greater average speed (~6 m s<sup>-1</sup>).

Dairy 2 consisted of a long rectangular area of animal housing, made up of large free stall barns and open lots. In the northeast of the farm, an open-air manure lagoon was set just north of two long settling cells. Larger than Dairy 2, Dairy 1 had more free stall barns and open lots. Separated from the animal housing, a large lagoon and settling cell extend side-by-side to the north of the barns. Detailed descriptions of meteorological conditions and depictions of each farm layout can be found in Arndt et al. (2018).

#### 3.2 Tracer flux emission estimates via aircraft

Some plumes ~~appear to represent~~ the entire site and all of its ~~sub~~-sources (“whole-site”). Other plumes ~~occur when can represent the wind direction isolates~~ an individual source (e.g. animal housing), when observed for during a given transect from a certain position at a particular wind direction. For ~~these~~ close and fast transects, it can be difficult to have the tracer in a position that ~~appears to adequately represents~~ the site or an individual sub-source. Designating

each observed plume to a source considers many factors but is ultimately up to the discretion of the analyst. Efforts to understand this interpretive bias are described in the Supplementary Information, and use two validation methods, one analyst-driven and one automated.

Plumes from each site were analysed using two different methods: linear regression and integration (Roscioli, 2015). Each method brings benefits and challenges. In the linear regression approach, outliers can deflect a slope off-trend for otherwise consistent data. Highly correlated relationships can be misleading, if not inspected closely. When applying the peak integration, subtle differences when drawing a baseline can have a significant effect on emission rates. Isolating enhancements by area during times of low signal-to-noise can be challenging. Automatically determined baselines were manually readjusted when necessary, requiring consistency and attention to detail. Both methods delivered similar emission rates for each designated source within measurement uncertainties. Emission rates determined by integration analysis were  $6,108 \pm 821 \text{ kg d}^{-1}$  for the whole site and  $2,188 \pm 391 \text{ kg d}^{-1}$  for animal housing at Dairy 1 and  $4,018 \pm 456 \text{ kg d}^{-1}$  for whole site and  $1,675 \pm 747 \text{ kg d}^{-1}$  for animal housing at Dairy 2. Using correlation analysis, emission rates were  $5,854 \pm 841 \text{ kg d}^{-1}$  for the whole site and  $1,867 \pm 299 \text{ kg d}^{-1}$  for animal housing at Dairy 1 and  $3,699 \pm 685 \text{ kg d}^{-1}$  for the whole site and  $1,283 \pm 536 \text{ kg d}^{-1}$  for animal housing at Dairy 2. Given the favourable comparison between methods, we present area analysis only in Table 2. These results indicate that the selected plumes were adequately co-dispersed with the tracer gas, as both analysis methods compare within uncertainty. Differences in emission rates by method would imply that the observed  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  plumes were spatially disparate airmasses.

Whole-site plumes emission estimates averaged for each farm closely resemble agree with the quantification results using estimates by other methods, during the initial Arndt et al. (2018) study (Table 2), and fall within the stated uncertainties of all methods for both farms. Emissions associated with animal housing (based on tracer proximity and wind direction) resemble mobile laboratory findings. Animal housing emission rates cannot be directly compared to the results of the mass balance technique from the original study as there was no apportionment by source (only whole-site estimates). Measurements of manure emissions were not compared with established techniques due to uncertainty in representation of the source by the tracer gas.

### 3.3 Overlapping measurements between platforms

Occasionally, the aircraft flew over the mobile lab while both vehicles were recording data sampling the same plume. One example of this coincidence can be seen in Fig. 3, allowing providing a for direct comparison between these two methods. Around midday of June 22, 2016, the aircraft (11:41:25 – 11:41:50 PDT) and the minAML (11:40:45 – 11:42:00 PDT) encountered the tracer gas and site emission plumes for 25 seconds and 75 seconds respectively. For this section of flight, the aircraft flew at around  $74 \text{ m s}^{-1}$  (165 mph) covering 1.3 km (0.8 mi) at an average altitude of 428 m. Meanwhile, the minAML drove on a paved road at about  $16 \text{ m s}^{-1}$  (35 mph) over 0.8 km (0.5 mi). Both transects occurred in the same direction, from east to west onto the southern side of the site. During the overlapping transects, each platform saw a sharp increase in  $\text{CH}_4$  concentration followed by a broad enhancement at lower concentrations while a similarly rapid rise in  $\text{C}_2\text{H}_6$  concentration was followed by steady decrease. Differences in baseline values of  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  are attributed to different schedules of acquiring backgrounds (inlet overblown with zero air more

frequently on the minAML). Given the similar spatial characteristics of these plumes, it seems likely both platforms were observing the same airmass. As expected, the aircraft-based observations show a lower temporal resolution versus the mobile lab due to speed differences. While these plumes would not be used for emission estimations based on tracer ratio due to poor ~~correlation~~ tracer representation, they show ~~that-how~~ the same airmass ~~can-be~~ appears when sampled on the ground and in the air.

## 4 Discussion

### 4.1 On-site sampling by aircraft

During each flight, identifiable plumes of CH<sub>4</sub> were observed regularly, ~~(approximately every 1-2 min)~~. Figure 4 depicts repeated measurements of CH<sub>4</sub> emissions representative of the whole farm, revealing characteristics about emission sources at each site. Viewed from the south, manure and animal housing areas at Dairy 1 line up together, whereas at Dairy 2, the anaerobic lagoon and settling cells are offset from the housing areas. While these observations largely depend on wind direction and distance from the source, some features gave insight into where emissions came from on-site. Broad emissions can be readily attributed to the large collection of point source emitters milling around barns and open lots (cows of various ages). Sharp peaks and broad plateaus indicate an encounter with outgassing by a large area source (liquid manure ponds). Gaussian shapes appear to be an amalgamation of both major sources mixed downwind.

Temporal and spatial differences exist between the aircraft measurements used in this dataset and the ground-based measurements collected as part of the initial study (Arndt et al., 2018). Measurements by the minAML ~~occured~~ occurred during the day and night at a variety of distances from each site (up to 6 km). The aircraft had good coverage during the middle of the day, with flights in the late morning and early afternoon performing frequently repeated transects ~~above~~ around each site (~1 km radius). The ground-based tracer release experiment ~~struggled at times~~ observed very low plume enhancements in the hot mid-day conditions due to low winds and strong vertical mixing while the aircraft saw good signal, but had no issue collecting night-time measurements when the aircraft could not operate.

Tracer flux ratio methodology thrives with strong winds and downwind road access perpendicular to the dominant wind direction. Close placement of tracer gas to a point source and distant measurements by the mobile lab allow time and space for the tracer to co-disperse with emission gas and merge together in the measured plume. During this field campaign, the aircraft flew close to the site measuring emissions in a calm wind and saw an abundance of signal due to strong surface heating. These conditions proved favorable for the aircraft and mass balance calculations but stretch the possible application of the tracer release method. Even so, the attempt to perform a tracer release experiment observed from an aircraft proved largely successful and provided direct insight as to how these measurements relate to the ground-based observations.

Due to the sensitivity of the C<sub>2</sub>H<sub>6</sub> instrument on the aircraft, it was readily apparent when the tracer gas was present and intermingling with the farm emissions. Figure 5 visualizes the initiation of tracer release at Dairy 2 and the time it takes for tracer gas to disperse on-site. As seen in Fig. 4, pPrior to releasing any tracer gas, the concentration

of C<sub>2</sub>H<sub>6</sub> shows a relatively steady baseline. After initiating the release of tracer gas at 20 slpm, it took approximately 20 minutes before the aircraft begins to detect it initially and another 15 min before the plume characteristics were stabilized. We suspect this was due to the prevailing conditions of weak horizontal winds and strong but varying vertical mixing at the site. The aircraft ascended above the emission plume for 10-20 minutes after the release began, taking it out of plume detection range which may have lengthened the time it took to first detect tracer gas. Based on the average wind direction (from the NW) and horizontal speed (4.2 m s<sup>-1</sup>) from 10:39 AM PDT (start of tracer release) to 11:00 AM PDT (first spike of C<sub>2</sub>H<sub>6</sub>), we could expect to begin seeing tracer gas after ~ 6 min at a distance of 1.6 km (from release point to the intersection between the circular transect and wind direction). We saw the first spike shortly after when would have expected, around 11 min after beginning release. After being observed on board the aircraft, another 15 minutes pass before transects reliably encounter emissions with corresponding tracer gas, which we attribute to a combination of decreasing altitude and evolving dispersion.

For the plumes reported in this dataset, there is no observed does no dependence appear to be of significant correlation between emission rate with and sampling altitude. In Fig. 6, CH<sub>4</sub> emissions are plotted versus aircraft altitude. Emissions between 0 – 6,500 kg d<sup>-1</sup> appear to be randomly distributed between 100 – 600 m at each site (Fig. 5). Two outliers show higher emission rates at low altitudes, unmatched at higher altitudes. Above 650 m are three other points scattered across a wide range of emissions (2,000 – 6,500 kg). Increasing emissions with decreasing height, in some cases, could be attributed to the influence of a strongly lofted lagoon signal at this site. Lower flights could have then caused the aircraft to encounter a larger proportion of the manure-related emissions instead of the ideal case: a well-mixed plume representative of the entire site.

During each flight, identifiable plumes of CH<sub>4</sub> were observed at a steady pace (approximately every 1-2 min). As seen in Fig. 5, repeated plumes also revealed unique characteristics within a site. Viewed from the south, manure and animal housing areas at Dairy 1 line up together, whereas at Dairy 2, the anaerobic lagoon and settling cells are offset from the housing areas. While these observations largely depend on wind direction and distance from the source, some features gave insight into where emissions came from on site. Broad emissions can be readily attributed to the large collection of point source emitters milling around barns and open lots (cows of various ages). Sharp peaks and broad plateaus indicate an encounter with outgassing by a large area source (liquid manure ponds). Gaussian shapes appear to be an amalgamation of both sub-sources mixed downwind.

Temporal and spatial differences exist between the aircraft measurements used in this dataset and the ground-based measurements collected as part of the initial study (Arndt et al., 2018). Measurements by the minAML occurred during the day and night at a variety of distances from each site (up to 6 km). The aircraft had good coverage during the middle of the day, with flights in the late morning and early afternoon performing frequently repeated transects above each site (~ 1 km). The ground-based tracer release experiment struggled at times in the hot mid-day conditions due to low winds and strong vertical mixing while the aircraft saw good signal, but had no issue collecting night-time measurements.



## 4.2 Experimental challenges

Swirling and calm winds shifted emissions around each site at various times over multiple days. When selecting valid plumes, proximity of the aircraft during an enhancement to a single ~~sub~~-source introduces a dilemma. Varying distances between the tracer gas release point and presumed source could affect the determined emission rate, due to imperfect co-dispersion. For example, using a tracer plume located 500 m away to represent a source 300 m away would be problematic. When measuring at greater distances with better resolution (due to sampling in a slower vehicle), it is often trivial to identify when the tracer inadequately represents the emission. Flying several times faster than the driven transect provided notable repeatability but made spatial understanding of the site difficult with respect to emission sources.

Direct estimates of liquid manure emissions proved unrealistic at both dairies due to sparse ~~amounts-number~~ of CH<sub>4</sub> plumes with sufficient tracer representation, despite favorable wind direction and aircraft position. A few plumes of acceptable data quality were identified as being related to liquid manure emissions at Dairy 2 (n = 4), but estimates were significantly higher than reported in Arndt et al. (2018) at  $5,1664.893 \pm 837-1,331$  kg CH<sub>4</sub> d<sup>-1</sup> (~~area analysis~~). Due to concerns that the tracer release location was not close enough to the liquid manure source to be representative, especially due to nonideal transect geometry and limited horizontal wind, this data is not reported in Table 24. Relative apportionment of CH<sub>4</sub> between ~~sub~~-sources (using only whole-site and animal housing values) showed manure-associated plumes leading the fractional contribution at Dairy 1 (~~69:31~~73:27) and Dairy 2 (~~55:45~~71:29). This was an expected finding based on US EPA methodology estimates (Arndt et al., 2018) for this month at Dairy 2 (73:27). Given the temporal natural of manure emissions, as reported by Leytem et al. (2017), it should be reinforced that these results only represent a short period of time (6 measurement days) in a single season. Despite the difficulty of collecting or identifying many distinct manure-associated plumes via measurements taken from this aircraft, the general apportionment of source emissions appears to remain evident.

Clear hot measurement days could have stimulated anaerobic activity in manure lagoons and caused greater release of gases (Safley et al., 1988), while strong thermal convection lofted concentrated and unmixed plumes. Aside from refinements to the method (e.g. moving the tracer gas closer to the source), performing this technique in different seasons, meteorological conditions, and during mixing events (e.g. flushing) would enhance our understanding of the variability in emissions from liquid manure management on dairy farms.

For the mobile laboratory, road access was a challenge at times. Large plots of surrounding cropland typically had a limited number of roads crossing through them, with those available often being private or undeveloped. In order to collect plumes adequately downwind of each site on accessible public roads, the ground-based ARI team required winds to come from certain directions. Being able to fly above the site eliminates these challenges. However, the aircraft flew a set pattern at each site, circling at a particular radius to optimize the established mass balance method, and did not explore downwind like the vehicle. As seen in Fig. 67, plumes used for determining emission rates were clustered in areas above each site that typically agreed with the dominant wind directions along the looping flight path. Wind rose plots for each site represent the wind conditions observed by the aircraft during the midpoint of each plume event (Fig. 76C and 67D). On-site wind measurements during these events provided additional insight as to how the wind evolved between the site and aircraft. Other plume events sometimes occurred inside of the dominant

downwind fetch, especially during calm wind conditions, but lacked the prerequisites to be included in emission estimations.

### 4.3 Future work

Future work towards refining the tracer release method ~~to~~with an aircraft will require several improvements to the current experimental design. Instead of flying around the perimeter of a dairy farm or other emission source in a circle as part of an established mass balance approach (Conley et al., 2017), the aircraft ~~sh~~could mimic the driven transects of the mobile lab via long horizontal transects at varying distances perpendicular to the dominant wind direction (Hacker et al., 2016). Conducting downwind transects at greater distances (e.g. 500 m to 5 km) would allow for better comparisons between platforms but may not be feasible in conditions similar to those experienced in this study (strong surface heating combined with calm horizontal winds), as it could be difficult to encounter the plume.

Rather than relying on only a couple point source releases, tracer gas could be released as a line or grid source along the border of liquid manure management areas or animal housing fence lines (Lamb et al., 1995; McGinn et al., 2006). Increasing the flow rate of tracer rate from 15 slpm by several factors would improve signal-to-noise ratios of tracer enhancements. Furthermore, an aircraft carrying a second instrument on-board that quickly (1 Hz) and precisely (ppt sensitivity) monitors a second tracer gas (e.g. C<sub>2</sub>H<sub>2</sub>) would provide a check on the observed tracer concentrations or could aid source identification. With two tracer gases, the initial ratio of release rates ought to persist throughout the migration of the plumes and be reflected in the ratio of downwind enhancements (“dual tracer ratio”; Roscioli et al., 2015). Deviations from the expected value indicate loss of tracer gas and inadequate representation of a source. It should be noted that the two tracers used in this original study were employed as independent tracers for better coverage over large multisource areas, while the scenario described above applies to overlapping use of tracer gases (two tracers for a single source). Benefits of adding a second tracer (dual-tracer flux ratio methodology) are described further in Roscioli et al. (2015).

Overall, combining these measurement techniques through aircraft-observed tracer release promotes positive aspects of each method. Low-flying aircraft measurements occur rapidly on a versatile platform with no road access restrictions. Tracer gases can indicate sources, identify interferences, and enable quantification without relying on modelling or highly accurate wind measurements. Using this method, an aircraft can have greater confidence identifying sources and can confirm ground-based observations.

## 5 Conclusion

By quantifying CH<sub>4</sub> emissions to within the uncertainties of ~~stand-alone~~independent ground-based tracer and aircraft mass balance measurements, this study demonstrates the viability of performing a tracer release experiment from the ground observed by an aircraft flying overhead. Other than intentionally overlapping measurement times, we were able to demonstrate a third method of monitoring dairy emissions using data collected for previously established techniques, without prior coordination or making any procedural changes in the field. In this case, an aircraft flying transects prioritized for a mass balance methodology successfully collected data viable for single tracer flux ratio analysis. Simultaneous observations by the aircraft and mobile laboratory on a similar spatial scale provide a brief



look into ~~of~~ how each technique experiences single tracer flux ratio methodology. Considering the success in applying this method, a refined approach could greatly improve and further demonstrate the feasibility of this technique.

*Data availability.* A table containing Site IDs, source designations, measurement durations, CH<sub>4</sub> emission rates, plane altitude, measurement distance from the point of tracer release, wind direction and speed, and coefficients of determination for CH<sub>4</sub>/C<sub>2</sub>H<sub>6</sub> can be found in Supplementary Materials (SM Dataset).

*Author contribution.* CA coordinated the field campaign. SC and ICF collected the aircraft data. CD, TIY, and JRR participated in the mobile laboratory measurements. CD, TIY, JRR, SC, ICF, and SCH performed data interpretation and analysis. CD prepared the manuscript with contributions from ICF. All authors critically reviewed the manuscript. All authors approved the submitted version for publication.

*Competing interests.* The authors declare that they have no conflict of interest.

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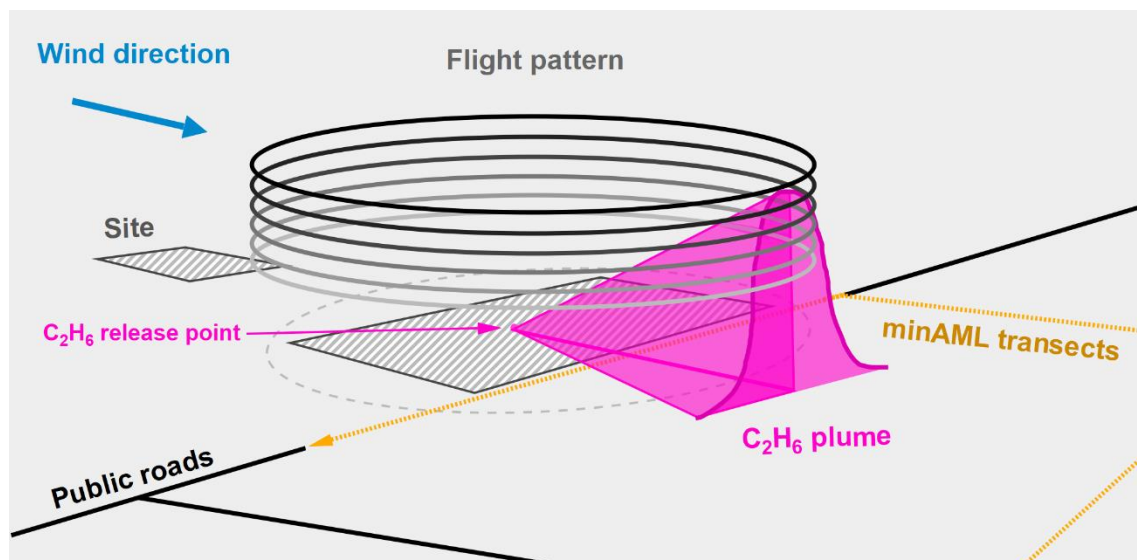
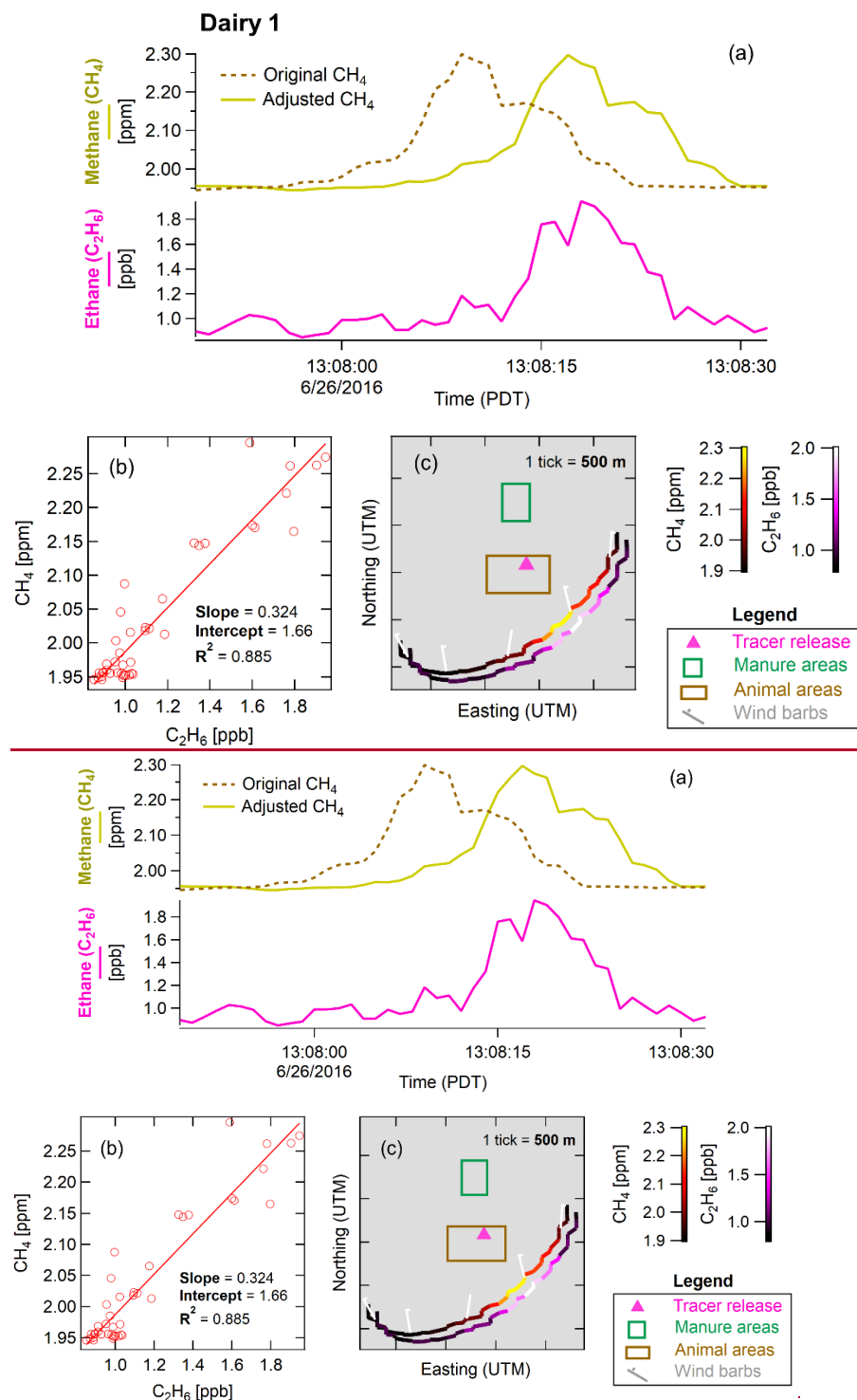
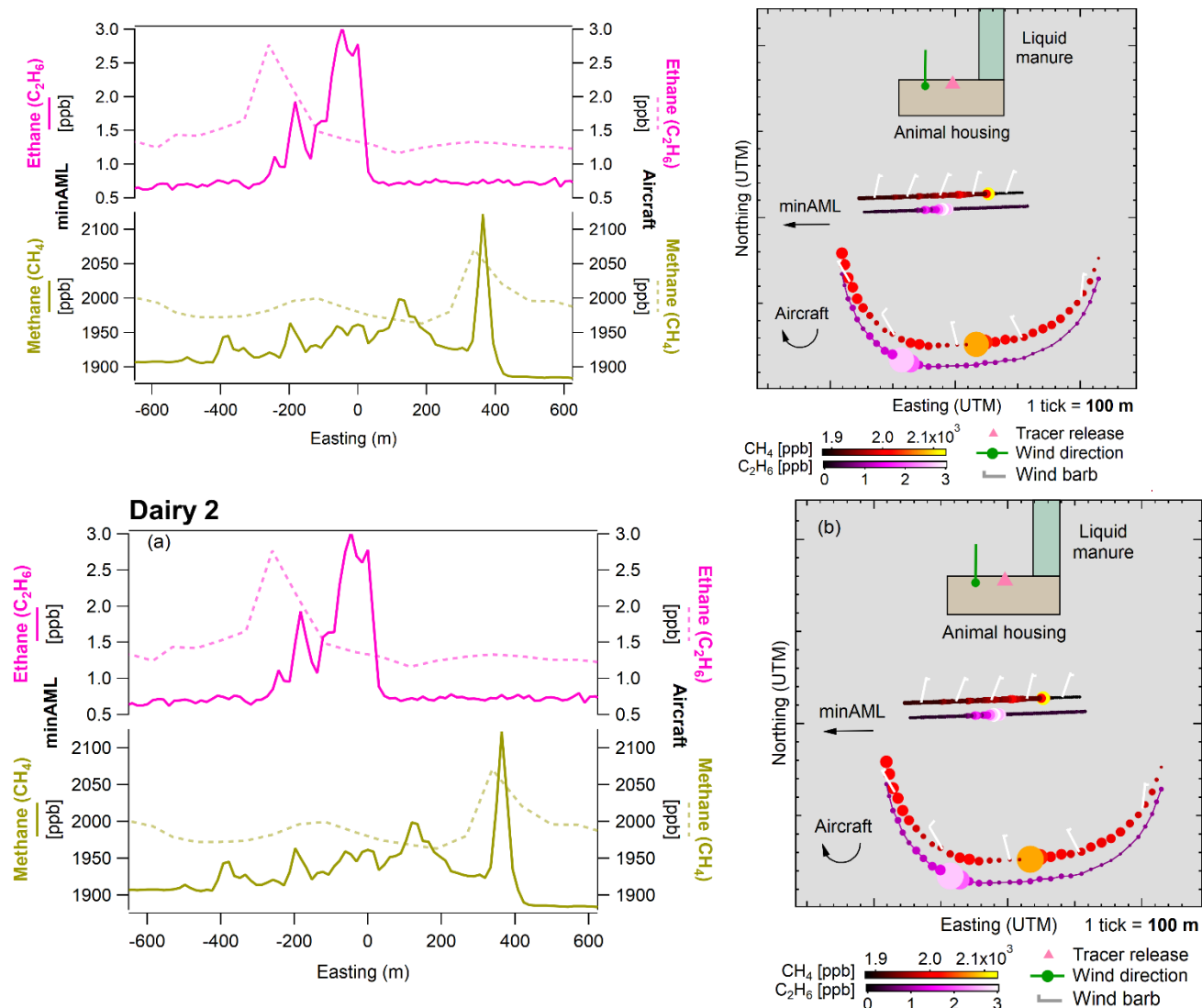


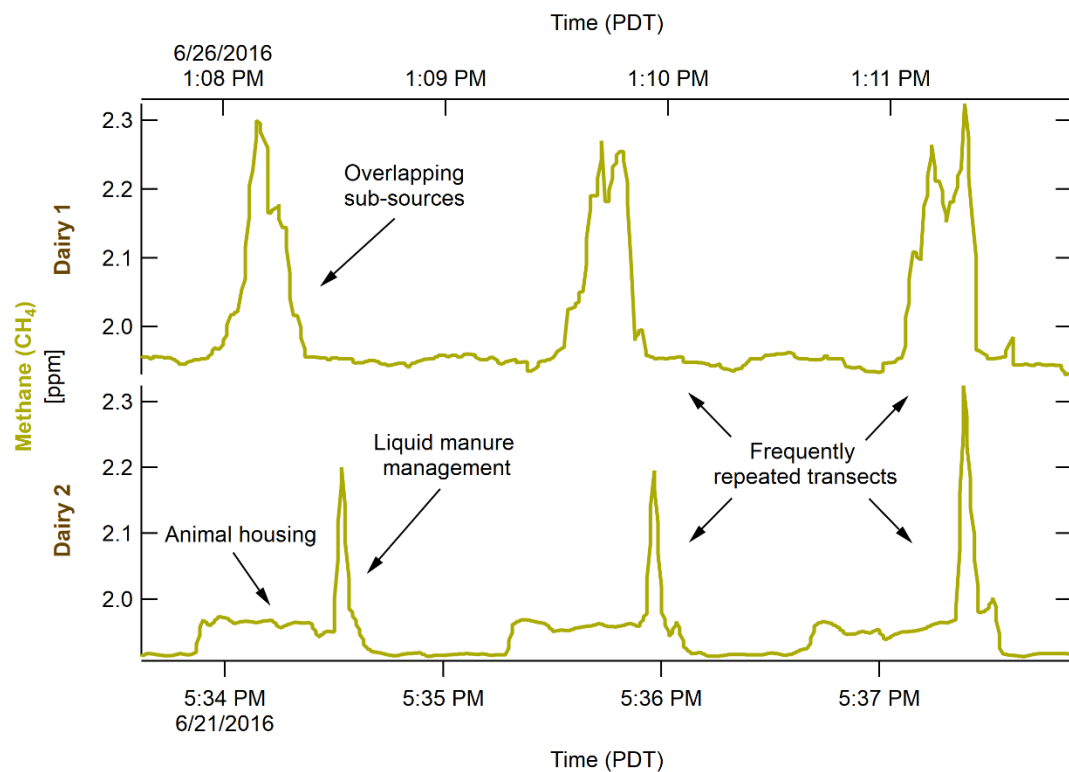
Figure 1. Experimental schematic of tracer release (ethane;  $C_2H_6$ ) at a dairy farm, as observed by a small aircraft and miniature Aerodyne Mobile Laboratory (minAML). In this ideal scenario, the wind is carrying the plume across the site perpendicular to accessible public roads.



**Figure 2. Time traces of methane ( $\text{CH}_4$ ) and ethane ( $\text{C}_2\text{H}_6$ ) during a flight around Dairy 1 (panel a). A correlation plot with a best-fit line (panel b) compares enhancements above baseline of  $\text{CH}_4$  and  $\text{C}_2\text{H}_6$  (panel a) after accounting for differences in instrument response times and tracer position relative to site emissions. See text for discussion of alternate analysis by area ratios. A map of Dairy 1 overlaid with the flight path is colored by  $\text{CH}_4$  concentration (panel c). An identical transect colored by  $\text{C}_2\text{H}_6$  is offset slightly for clarity. Wind barbs depict the wind velocity (averaging  $2.4\text{ m s}^{-1}$  from NNW) at several points during the transect.**



**Figure 3. Plumes observed by the miniature Aerodyne Mobile Laboratory (minAML) and aircraft. Plots of methane ( $CH_4$ ) and ethane ( $C_2H_6$ ) are overlaid for each platform (panel a). Observations occurred during transects by each vehicle to the south of Dairy 2, during a release of  $C_2H_6$  into a southerly wind (panel b). Potential emission sources on the farm have been identified as colored sections, though not as an exact scaled representation.**



**Figure 4.** Selected sampling periods (approximately 5 minutes) at each dairy farm showing characteristics of emitted methane plumes as observed by the aircraft downwind to the south. Each time trace depicts the high rate of repetition in the flown transects around each site.

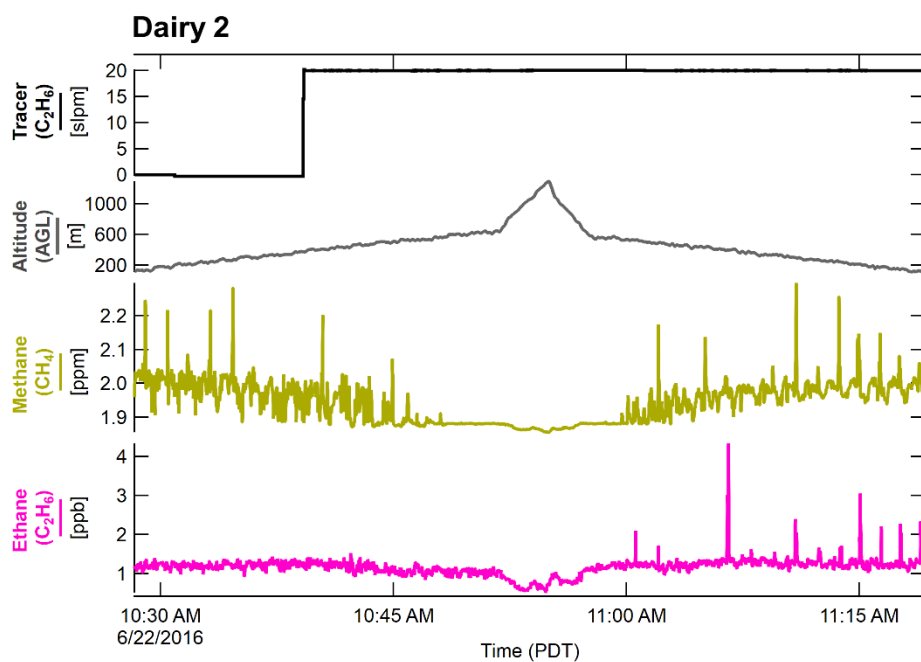
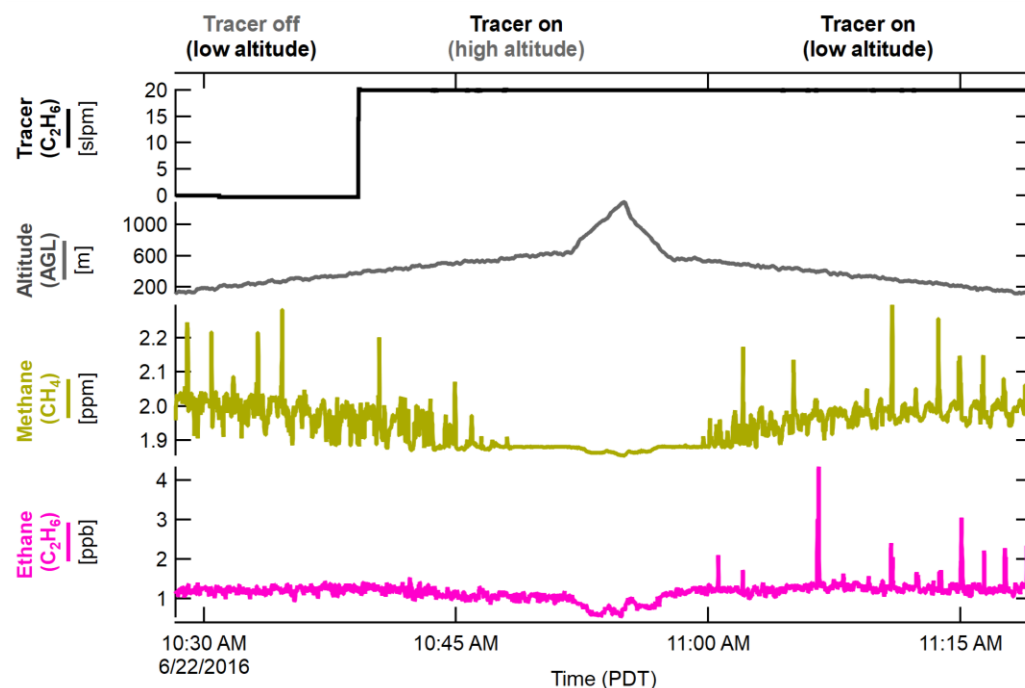
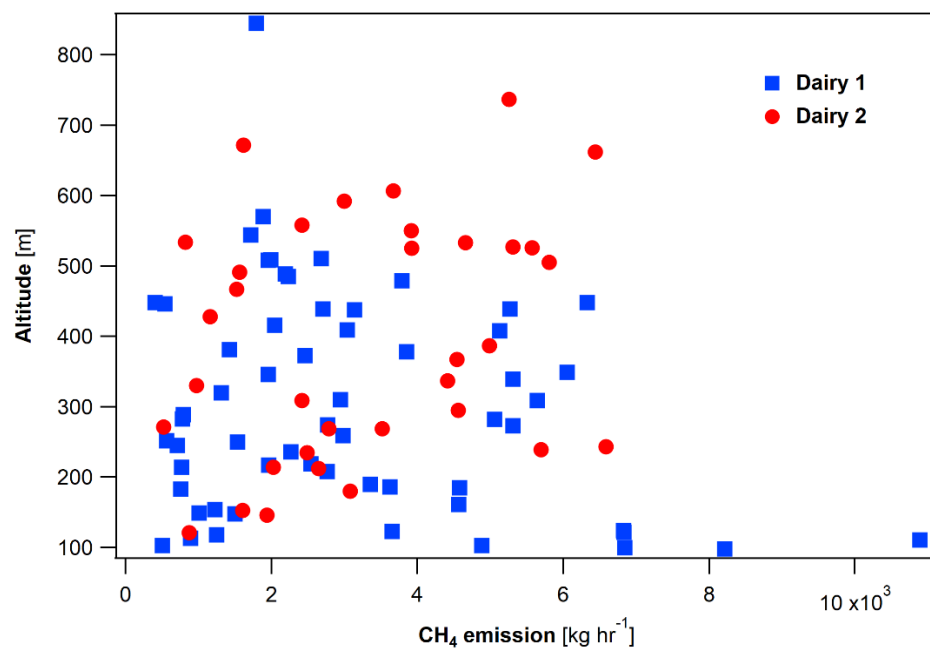


Figure 5. Comparison of flight sampling periods prior to and during release of tracer gas (ethane,  $C_2H_6$ ), showing enhancements of methane above Dairy 2 with and without corresponding peaks of  $C_2H_6$  depending on release rate, altitude (AGL), and dispersion.

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**Figure 6. Observed methane emissions (CH<sub>4</sub>; kg d<sup>-1</sup>) plotted by aircraft altitude at both dairy farms (Dairy 1 and Dairy 2). Emission rates are distributed randomly across hundreds of meters in altitude with a handful of outliers at lower and higher altitudes.**

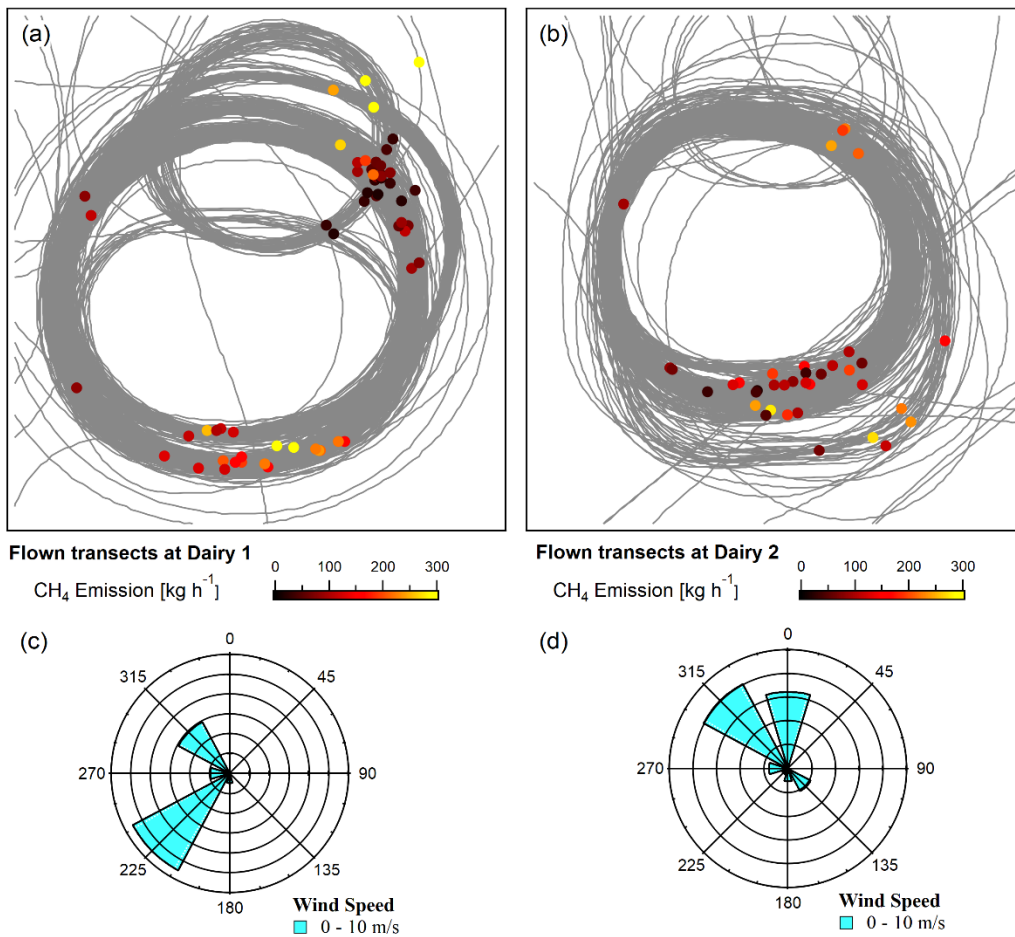
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**Figure 7. Methane (CH<sub>4</sub>) emission rates displayed on every flight track as dots, positioned at the mid-point of each enhancement event (panel a and b). Corresponding wind roses average the originating direction and magnitude of the wind from the mid-point of each plume event (panel c and d).**

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**Table 1. Overlap between flight times and release of tracer gas (ethane) over the course of the field campaign.**

	Days spent*	Total release	Overlap	Overlap by flight
	[n]		[Elapsed time - hh:mm]	
Dairy 1	5	13:00	3:55	0:47
Dairy 2	6	27:05	7:25	1:14
Both Sites	11	40:05	11:20	-

\*Release on June 25th but no flights.

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**Table 2. Comparison of methane emission estimates ( $\text{kg d}^{-1} \pm 95\% \text{ C.I.}$ ) for two dairy farms between this paper (“tracer-plane”) and established tracer release (“ARI”) and mass balance (“SA”) methods.**

Source	Tracer-Plane	ARI*	SA*
[ $\text{kg CH}_4 \text{d}^{-1} \pm 95\% \text{ C.I.}$ ]			
Whole-site	-	-	-
Dairy 1	5,850 $\pm$ 793	6,985 $\pm$ 626	7,249 $\pm$ 2,153
Dairy 2	3,699 $\pm$ 685	3,046 $\pm$ 814	3,274 $\pm$ 745
Animal housing	-	-	-
Dairy 1	1,835 $\pm$ 295	2,601 $\pm$ 811	-
Dairy 2	1,648 $\pm$ 563	1,636 $\pm$ 513	-
Liquid manure	-	-	-
Dairy 1	-	5,994 $\pm$ 579	-
Dairy 2	-	2,141 $\pm$ 637**	-
*Arndt et al., 2018.	-	-	-
**Settling basin value only, from Arndt et al., 2018.	-	-	-

Source	Tracer-Plane	ARI*	SA*
[ $\text{kg CH}_4 \text{d}^{-1} \pm 95\% \text{ C.I.}$ ]			
Whole-site			
Dairy 1	6,108 $\pm$ 821	6,985 $\pm$ 626	7,249 $\pm$ 2,153
Dairy 2	4,018 $\pm$ 456	3,046 $\pm$ 814	3,274 $\pm$ 745
Animal housing			
Dairy 1	2,188 $\pm$ 391	2,601 $\pm$ 811	-
Dairy 2	1,675 $\pm$ 747	1,636 $\pm$ 513	-
Liquid manure			
Dairy 1	-	5,994 $\pm$ 579	-
Dairy 2	-	2,141 $\pm$ 637**	-

\*Arndt et al., 2018.

\*\*Settling basin value only, from Arndt et al., 2018.