

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/328213733>

# Short-term methane emissions from 2 dairy farms in California estimated by different measurement techniques and US Environmental Protection Agency inventory methodology: A case stu...

Article in *Journal of Dairy Science* · October 2018

DOI: 10.3168/jds.2017-13881

CITATIONS

2

READS

75

9 authors, including:



**Claudia Arndt**

CATIE - Centro Agronómico Tropical de Investigación y Enseñanza

24 PUBLICATIONS 170 CITATIONS

SEE PROFILE



**Daniel Zavala-Araiza**

Environmental Defense Fund

39 PUBLICATIONS 567 CITATIONS

SEE PROFILE



**Ian Faloona**

University of California, Davis

107 PUBLICATIONS 3,096 CITATIONS

SEE PROFILE

Some of the authors of this publication are also working on these related projects:



Oil and Gas Sector Methane Emissions Quantification and Mitigation [View project](#)



On Farm Evaluation of Methodologies for Estimating Methane Emissions from Dairies in California [View project](#)



J. Dairy Sci. 101:1–20  
<https://doi.org/10.3168/jds.2017-13881>

© 2018, The Authors. Published by FASS Inc. and Elsevier Inc. on behalf of the American Dairy Science Association®.  
 This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

## Short-term methane emissions from 2 dairy farms in California estimated by different measurement techniques and US Environmental Protection Agency inventory methodology: A case study

C. Arndt,<sup>\*1</sup> A. B. Leytem,<sup>†</sup> A. N. Hristov,<sup>‡</sup> D. Zavala-Araiza,<sup>§</sup> J. P. Catiavela,<sup>#</sup> S. Conley,<sup>||</sup> C. Daube,<sup>¶</sup> I. Faloona, and S. C. Herndon<sup>¶</sup>

<sup>\*</sup>Environmental Defense Fund, San Francisco, CA 94105

<sup>†</sup>USDA-Agricultural Research Service, Northwest Irrigation and Soils Research Laboratory, Kimberly, ID 83341

<sup>‡</sup>Department of Animal Science, The Pennsylvania State University, University Park 16802

<sup>§</sup>Environmental Defense Fund, Austin, TX 78701

<sup>#</sup>Dairy Cares, Sacramento, CA 95814

<sup>||</sup>Department of Land, Air, and Water Resources, University of California, Davis 95616

<sup>¶</sup>Aerodyne Research Inc., Billerica, MA, 01821

### ABSTRACT

Reported estimates of CH<sub>4</sub> emissions from ruminants and manure management are up to 2 times higher in atmospheric top-down calculations than in bottom-up (BU) inventories. We explored this discrepancy by estimating CH<sub>4</sub> emissions of 2 dairy facilities in California with US Environmental Protection Agency (US EPA) methodology, which is used for BU inventories, and 3 independent measurement techniques: (1) open-path measurements with inverse dispersion modeling (hereafter open-path), (2) vehicle measurements with tracer flux ratio method, and (3) aircraft measurements with the closed-path method. All 3 techniques were used to estimate whole-facility CH<sub>4</sub> emissions during 3 to 6 d per farm in the summer of 2016. In addition, open-path was used to estimate whole-facility CH<sub>4</sub> emissions over 13 to 14 d per farm in the winter of 2017. Our objectives were to (1) compare whole-facility CH<sub>4</sub> measurements utilizing the different measurement techniques, (2) compare whole-facility CH<sub>4</sub> measurements to US EPA inventory methodology estimates, and (3) compare CH<sub>4</sub> emissions between 2 dairies. Whole-facility CH<sub>4</sub> estimates were similar among measurement techniques. No seasonality was detected for CH<sub>4</sub> emissions from animal housing, but CH<sub>4</sub> emissions from liquid manure storage were 3 to 6 times greater during the summer than during the winter measurement periods. The findings confirm previous studies showing that whole-facility CH<sub>4</sub> emissions need to be measured throughout the year to estimate and evaluate annual inventories. Open-path measurements for liquid manure storage emissions were similar to monthly US EPA estimates during the summer, but

not during the winter measurement periods. However, the numerical difference was relatively small considering yearly emission estimates. Manure CH<sub>4</sub> emissions contributed 69 to 79% and 26 to 47% of whole-facility CH<sub>4</sub> emissions during the summer and winter measurement periods, respectively. Methane yields from animal housing were similar between farms (on average 20.9 g of CH<sub>4</sub>/kg of dry matter intake), but CH<sub>4</sub> emissions normalized by volatile solids (VS) loading from liquid manure storage (g of CH<sub>4</sub> per day/kg of VS produced by all cattle per day) at 1 dairy were 1.7 and 3.5 times greater than at the other during the summer (234 vs. 137 g of CH<sub>4</sub>/kg of VS) and winter measurement periods (78 vs. 22 g of CH<sub>4</sub>/kg of VS), respectively. We attributed much of this difference to the proportion of manure stored in liquid (anaerobic) form, and suggest that manure management practices that reduce the amount of manure solids stored in liquid form could significantly reduce dairy CH<sub>4</sub> emissions.

**Key words:** methane, measurement technique, emission, manure

### INTRODUCTION

Reducing short-lived climate pollutants is key to limiting global warming to 2°C above preindustrial levels (Shindell et al., 2012). Methane is a short-lived climate pollutant with 3 times the global warming potential (84 vs. 28) in the short term (20 yr) than the long term (100 yr; IPCC, 2013). Consequently, the contribution of CH<sub>4</sub> to anthropogenic greenhouse gas is greater in the short term than in the long term (28 vs. 11%; US EPA, 2017b). Livestock alone contribute 35% [9.3 million metric tonnes (MMT) of CH<sub>4</sub>, with 72% from enteric fermentation and 28% from manure management] of the anthropogenic CH<sub>4</sub> in the United States (US EPA, 2017b). In California, the largest dairy-producing state

Received September 22, 2017.

Accepted August 11, 2018.

<sup>1</sup>Corresponding author: [claudia.arndt@catie.ac.cr](mailto:claudia.arndt@catie.ac.cr)

with 19% of the total US milk production (USDA-ERS, 2017), the California Air Resource Board (CARB, 2017) estimates that dairy livestock alone contributes 55% (0.7 MMT of CH<sub>4</sub>, with 45% from enteric fermentation and 55% from manure management) of the anthropogenic CH<sub>4</sub> emissions. The CARB (2017) estimates are bottom-up estimates and based on US EPA (2017a) methodology (Deshpande et al., 2014), which is derived from the Intergovernmental Panel on Climate Change (IPCC, 2006b) guidelines for emissions from livestock and manure management.

Regional top-down (TD) measurements, which used inverse modeling to attribute CH<sub>4</sub> emissions to different emission sources, have suggested that the national and California inventories underestimate CH<sub>4</sub> emissions from livestock (Miller et al., 2013; Wecht et al., 2014a; Bergamaschi et al., 2015; Jeong et al., 2016; Cui et al., 2017). Based on long-term measurements, Miller et al. (2013) estimated that national CH<sub>4</sub> emissions from ruminants and manure management are up to 2 times greater than the US Environmental Protection Agency (EPA) inventory estimates. Similarly, long-term measurements by Jeong et al. (2016) estimated that CH<sub>4</sub> emissions from dairy livestock in the San Joaquin Valley are 2 times greater than the California Air Resource Board (CARB) estimates. Short-term measurements by Trousdell et al. (2016) and Cui et al. (2017) estimated that CH<sub>4</sub> emissions from dairy livestock in the San Joaquin Valley are up to 2.4 times greater than CARB estimates. Other regional short-term TD measurements have suggested that dairy livestock emissions are similar to CARB estimates (Peischl et al., 2013; Wecht et al., 2014b). Top-down inventory estimates are based on inverse modeling and use gridded bottom-up (BU) inventories as prior estimates. Thus, errors associated with gridded BU inventories can lead to errors in the TD inventory (Hristov et al., 2017); this makes TD estimates not ideal to evaluate US EPA methodology. Measurements of facilities that provide activity and management data avoid this potential error, and thus are more suited to evaluate US EPA methodology. Open-path has been used to estimate emissions from dairy facilities for many years, and more recently vehicle- and aircraft-based techniques have become available. It is important to evaluate these different techniques because CH<sub>4</sub> measurements are needed to not only evaluate US EPA methodology because of the discrepancy in TD and US EPA inventories, but also to estimate reduction potentials of mitigation strategies. This is even more important in California, after the California Legislature passed Senate Bill 1383 (State of California, 2016) in September 2016, which requires CARB to adopt regulations to reduce CH<sub>4</sub> emissions

from dairy manure management by up to 40% below the dairy sector's 2013 levels by 2030.

As CH<sub>4</sub> emissions from dairy farms come from 2 main sources, enteric fermentation and manure storage [primarily liquid (anaerobic) manure storage], it is important to distinguish between the 2 sources to validate US EPA methodology and to estimate their contributions to the current CH<sub>4</sub> inventories. The objectives of our study were to (1) compare whole-facility CH<sub>4</sub> measurements utilizing different measurement techniques, (2) compare whole-facility CH<sub>4</sub> measurements to US EPA inventory methodology estimates, and (3) compare CH<sub>4</sub> emissions between 2 dairies.

## MATERIALS AND METHODS

Whole-facility CH<sub>4</sub> emissions were measured at 2 commercial dairy farms (dairy 1 and 2) in California. Each farm was visited once in summer 2016 and once in winter 2017. On dairy 1, emissions were measured from June 24 to 30 (summer 2016) and from January 17 to February 1 (winter 2017). On dairy 2, emissions were measured from June 20 to 23 (summer 2016) and from February 2 to 16 (winter 2017). During the summer measurement period, 3 independent techniques were used to measure emissions: (1) open-path measurements with inverse dispersion modeling (hereafter open-path); (2) vehicle measurements with tracer flux ratio method (vehicle); and (3) aircraft measurements with a closed-path (elliptical) method (aircraft). During the winter measurement period, only open-path measurements were conducted.

### Farm Selection

The dairy farms were selected based on facility layout (separation of animal housing and liquid manure storage system), layout relative to downwind roads, distance to other CH<sub>4</sub> sources, and owner willingness to participate in the study. On both farms, the animal housing was in the southern part of the facility and the manure storage system (settling basins and anaerobic lagoon) was in the northern part of the facility (Figures 1 and 2). Both dairies were surrounded by irrigated cropland on 4 sides and located in areas with no known close CH<sub>4</sub> sources; the nearest expected CH<sub>4</sub> source, such as another dairy or oil and gas facility, was more than 2.5 km away from each dairy.

### Farm Management

Both farms had Jersey cattle as the primary breed. The California Department of Food and Agriculture (CDFA) estimated that in California, in 2016, Jersey

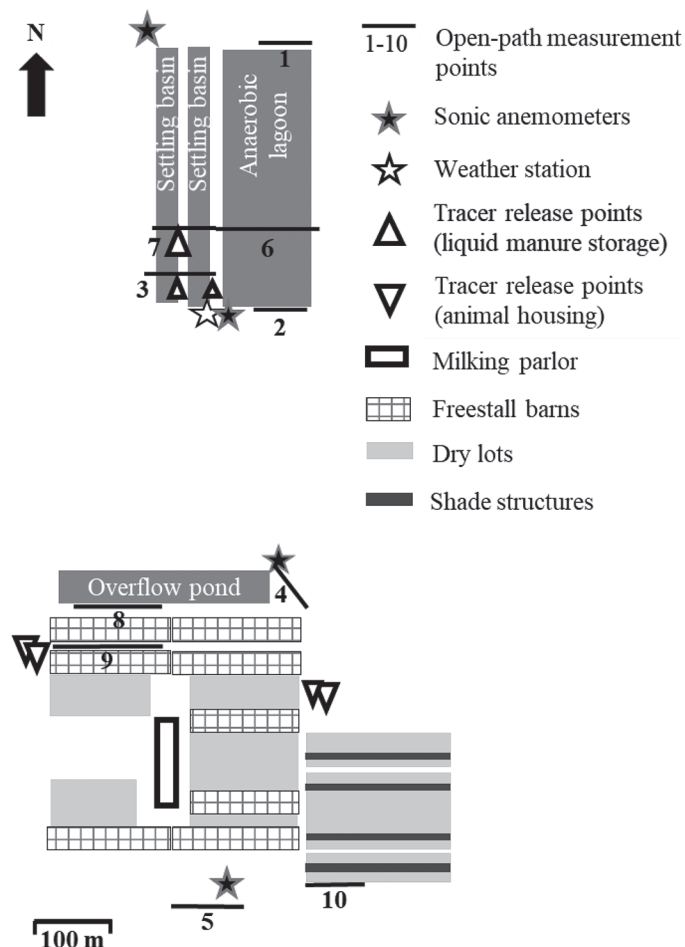
and mixed herds produced 10.2 and 11.6% of California's milk, respectively (CDFA, 2017).

Dairy 1 had 6,036 cattle (54% dairy cows, 37% replacement heifers, and 10% calves; Table 1) and cows were milked twice daily. During the measurement periods (summer 2016 and winter 2017), average DMI was  $20.0 \pm 0.1$  and average milk production was  $27.4 \pm 0.2$  kg/lactating cow per day (mean  $\pm$  SD). Replacement heifers and dairy cows were fed TMR starting at 0500 h and continued until 1600 h; replacement heifers and nonlactating cows were fed once daily (morning) and lactating cows were fed twice daily (morning and afternoon).

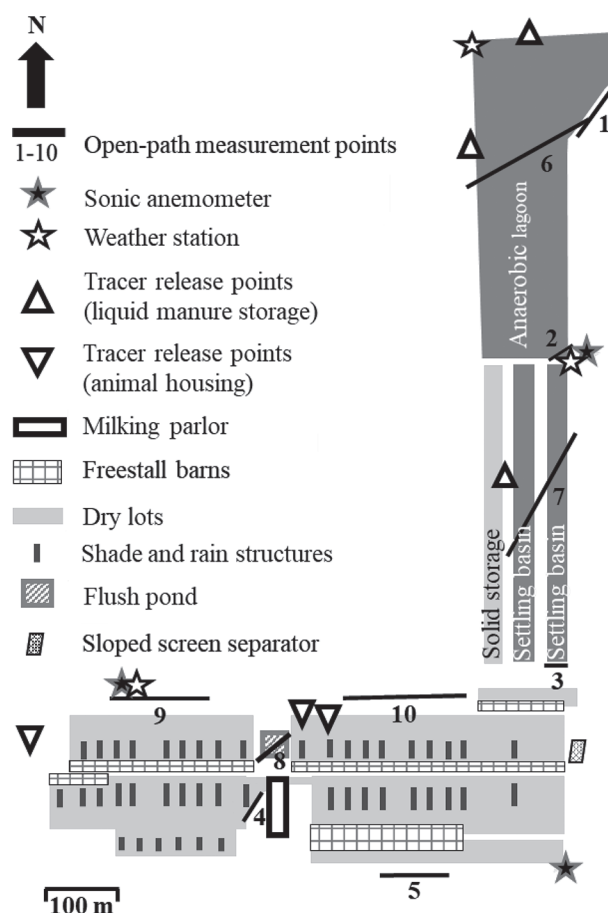
The dairy cows (lactating and dry cows) and late-pregnant heifers were housed in freestall barns with and without dry lot access on the western part of the facility, whereas replacement heifers and older calves were housed in dry lots on the eastern part of the facility. Dairy cows had no access to adjacent dry lots when the lots were muddy due to high precipitation during

the winter months. The stalls in the barns were bedded with dried manure solids [2.5 to 3.0 kg of volatile solids (VS)/dairy cow per day], which is a common practice for dairies in California. During winter, when precipitation was high, approximately twice the amount of bedding was used to keep the stalls dry. The farm had run out of manure solids to use for bedding by January 17, 2017; thus, rice hulls (8.2 t/d on a DM basis) were used for bedding instead of manure solids during the winter open-path measurement period (between January 17 and February 1, 2017). Manure solids for bedding were recovered from scraping the dry lots (every 2 wk when the lots were dry) and evacuating the crust on the settling basins (every May and October, except for October 2016). Recovered manure solids were exclusively used for bedding.

Manure and bedding were removed from the freestall barns and the feed alleys of the dry lots by flushing twice a day. Recycled anaerobic lagoon water was used for flushing. The wash water from the milking parlor went straight into the liquid manure storage. Two sequential parallel settling basins (each 9,464 m<sup>2</sup> surface



**Figure 1.** Facility layout, open-path measurement, and tracer release points for vehicle measurements on dairy 1.



**Figure 2.** Facility layout, open-path measurement, and tracer release points for vehicle measurements on dairy 2.

**Table 1.** Characteristics of dairy 1 and dairy 2

Item <sup>1</sup>	Dairy 1	Dairy 2
Total cattle, animal unit <sup>2</sup>	4,590	3,088
Total cattle, head	6,036	3,241
Dairy cows, <sup>3</sup> head	3,244	2,416
Replacement heifers, <sup>4</sup> head	2,217	825
Calves, <sup>5</sup> head	576	0
Dairy cow housing		
Freestall barns without dry lot, % of cows	59	0
Freestall barns with dry lot, % of cows	41	73
Dry lot with feed alleyway flush, % of cows	0	27
Replacement heifer housing		
Freestall barns without dry lot, % of heifers	17	0
Dry lots with feed alleyway flush, % of heifers	83	100
Calf housing		
Dry lots with feed alleyway flush, % of calves	100	0
Manure separator (% of VS removal)	No	Yes (40) <sup>6</sup>
Bedding material	Manure solids and rice hulls	Manure solids
Source of manure solids for bedding	Dry lot scrapings and settling basin crust <sup>7</sup>	Separated solids
% of VS from manure stored in the liquid manure storage system <sup>8</sup>	91	49
% of VS from manure that is hauled off the farm <sup>8</sup>	0	38
% of VS from manure that leaves liquid storage through irrigation water <sup>8</sup>	9	13
Alternate use of settling basins	No	Yes <sup>9</sup>
Last time settling basin was emptied/switched	October 2013	October 2014 and 2016
Last time anaerobic lagoon was emptied	Has not been emptied since commissioning	Has not been emptied since commissioning

<sup>1</sup>VS = volatile solids.

<sup>2</sup>Animal units (AU) = 454 kg of live weight. We used 1.00 AU for each Jersey dairy cow (mature animals), 0.57 AU for each Jersey heifer (between 5 and 24 mo of age), 0.81 AU for each Holstein heifer (between 6 and 24 mo of age), and 0.13 AU for each Jersey calf (4 mo of age). All weights were based on average monthly BW published by Penn State College of Agricultural Sciences (2017a, b).

<sup>3</sup>Dairy cows = lactating and dry cows.

<sup>4</sup>Replacement heifers = animals between 5 and 24 mo of age. On dairy 1 and 2, replacement heifers were Jersey and Holstein, respectively.

<sup>5</sup>Calves = animals 4 mo of age consuming a starter ration.

<sup>6</sup>Separator efficiencies according to US Environmental Protection Agency (Cortney Itle, Eastern Research Group Inc., Chantilly, VA, personal communication).

<sup>7</sup>Natural crust of settling basin was evacuated in May and October each year, except for October 2016.

<sup>8</sup>Volatile solid flows were estimated based on farm manure management.

<sup>9</sup>The settling basins were switched every 2 yr. After switching, it takes 1 yr for a settling basin to dry before manure solids can be recovered.

area) and 1 anaerobic lagoon (40,560 m<sup>2</sup> surface area) were used for liquid manure storage. The anaerobic lagoon water was used to irrigate the surrounding fields when needed. Thus, the lagoon water level was higher during the winter (rainy season) and lower during the summer (dry season) due to water demand of the crops on the surrounding fields. However, despite the use of lagoon water for irrigation, the liquid manure storage system had not been completely emptied since commissioning (>5 yr). Supplemental File S1 (<https://doi.org/10.3168/jds.2017-13881>) provides further details on facility layouts and management practices.

Dairy 2 had 3,241 cattle (75% dairy cows, 25% replacement heifers, and 0% calves; Table 1). Holstein heifers were reared on-site and moved to a different facility before calving, whereas Jersey replacement heifers and calves were reared offsite and moved back to dairy 2 before calving. Cows were milked twice daily. During the measurement periods (summer 2016 and winter 2017), average DMI was  $20.8 \pm 0.7$  and aver-

age milk production was  $26.5 \pm 0.3$  kg/lactating cow per day (mean  $\pm$  SD). Similar to dairy 1, replacement heifers and dairy cows were fed TMR starting at 0500 h and continuing until 1600 h; replacement heifers and nonlactating cows were fed once daily (morning) and lactating cows were fed twice daily (morning and afternoon).

The dairy cows were housed in freestall barns with dry lot access and in dry lots. The replacement heifers were housed in dry lots. Stalls in the barns were bedded with dried manure solids (2.5 to 3.0 kg of VS/dairy cow per day). During winter, approximately 1.5 to 2 times the amount of bedding was used to keep the stalls dry. Similar to dairy 1, cows housed in barns had no access to adjacent dry lots when the lots were muddy due to high precipitation during the winter months. Manure solids were recovered by scraping the dry lots, emptying dried settling basins, and the use of a sloped-screen manure separator that removed solids from the flush water from the barns and dry lot alleys. The farm used

90% of the manure solids recovered by the sloped-screen manure separator for bedding, whereas the remaining manure solids were hauled off the farm to be used as fertilizer elsewhere.

Manure and bedding were removed from the freestall barns and the feed alleys of the dry lots by flushing twice a day. The wash water from the milking parlor was collected in a flush pond north of the milking parlor (1,823 m<sup>2</sup> surface area; Figure 2) and later used for flushing the barns and feed alleys of the dry lots. Two parallel settling basins (9,068 m<sup>2</sup> surface area to the east and 10,256 m<sup>2</sup> surface area to the west) and an anaerobic lagoon (65,424 m<sup>2</sup> surface area) were used for liquid manure storage (Figure 2); only 1 of the settling basins was used at a time. After one had been used for approximately 2 yr, it was left to dry and accumulated manure solids were removed while the other settling basin was used. The settling basins were alternated in October 2014 and 2016. The anaerobic lagoon water was used to irrigate the surrounding fields when needed. Thus, similar to dairy 1, lagoon water level was higher during the winter (rainy season) and lower during the summer (dry season) due to the water demand of the crops on the surrounding fields. Similar to dairy 1, the anaerobic lagoon had not been completely emptied since commissioning (>5 yr). Supplemental File S1 (<https://doi.org/10.3168/jds.2017-13881>) provides further details on facility layouts and management practices and Table 1 summarizes the characteristics of dairy 1 and 2.

### **Sampling of Feed, Manure, and Bedding**

The amount of TMR offered (on an as-fed basis) and diet DM used by the farm to formulate rations were recorded during each measurement period. In addition, all TMR loads were sampled according to Robinson and Meyer (2010), and samples were stored at  $-20^{\circ}\text{C}$  until analysis. During the summer measurement period, daily samples of each TMR load were collected from June 25 to 27 and June 21 to 23 on dairy 1 and 2, respectively. During the winter measurement period, samples of each TMR load were collected on January 18 and 26 and on February 3 and 9 on dairy 1 and 2, respectively. On dairy 1, refused feed was also collected, weighed, and sampled during the summer (on June 25 and 29) but not during the winter measurement period. On dairy 2, all cattle were fed at 0% refusals during the summer and winter measurement periods.

Lagoon samples were taken during the summer measurement periods and used to determine the VS removed in lagoon water used for irrigation. Approximately 7 to 8 samples (500 mL) were collected around the perimeter of the anaerobic lagoons at a depth of

approximately 0.6 m using a pole with retractable lid. Samples were frozen at  $-20^{\circ}\text{C}$  until analysis. Bedding was sampled from stockpiled material during the winter measurement period. Approximately 8 subsamples were composited and frozen ( $-20^{\circ}\text{C}$ ); frozen lagoon and bedding samples were shipped to the USDA-ARS laboratory in Kimberly, Idaho, for analysis.

### **Sample Analysis**

Samples from individual TMR loads and refused feed were composited across days by farm on an equal-weight basis (as fed). Composited TMR, refused feed, and bedding samples were dried at  $55^{\circ}\text{C}$  (forced-air oven) for 72 h and ground to pass through a 2-mm Wiley mill screen (Thomas Scientific, Swedesboro, NJ). Samples of ground TMR and refused feed were shipped to Dairyland Laboratories Inc. (Acadia, WI), where they were analyzed for DM content, ash, CP, ash-free neutral detergent fiber (aNDF), and ether extract. Details on the individual analysis procedures can be found in the Supplemental File S2 (<https://doi.org/10.3168/jds.2017-13881>). An average value from samples that have been run at Dairyland Laboratories was used to estimate neutral detergent insoluble CP of the TMR. Nonfiber carbohydrates were calculated according to (NRC, 2001) using CP, aNDF, neutral detergent insoluble CP, ether extract, and ash. Gross energy of the TMR was calculated using heat of combustion of 4.2 Mcal/kg for carbohydrates (aNDF and NFC), 5.6 Mcal/kg for CP, and 9.4 Mcal/kg for fatty acids (estimated by ether extract - 1; Maynard et al., 1979; Allen, 2000). Bedding and anaerobic lagoon samples were analyzed for VS according to standard method 2540E (Eaton et al., 2005).

### **Volatile Solids Production and Flow Rates**

The VS excreted by the different cattle types [dairy calves (4 mo of age), replacement heifers (>4 mo of age), and dairy cows (lactating and dry cows)] were estimated based on US EPA (2017a) methodology using the measured DMI and the calculated GE content of the diet. Details of the calculation can be found in Supplemental File S3 (<https://doi.org/10.3168/jds.2017-13881>).

Volatile solids were stored dry (dry lot and solid storage) and in liquid (liquid manure storage system). On dairy 1, the liquid storage system consisted of 2 settling basins, an anaerobic lagoon, and an overflow pond. The overflow pond was only used during winter 2017 due to record-high precipitation in California during the winter of 2016 and 2017. On dairy 2, the liquid storage system consisted of a flush pond, manure separator,

2 alternately used settling basins, and an anaerobic lagoon. The proportion of VS stored in the dry and liquid system was calculated based on US EPA (2017a) assumptions, which included the removal of 40% VS from the flush water by the manure separator before entering the anaerobic lagoon on dairy 2 (Cortney Itle, Eastern Research Group Inc., Chantilly, VA, personal communication), and on-farm practices (Table 2). The on-farm VS flows were estimated because US EPA (2017a) assumptions did not reflect on-farm manure management practices of the measured dairies. On dairy 1, all VS from manure entered the liquid manure storage system either as manure or bedding (manure solid) because dairy 1 did not haul manure off the farm. The only VS that left the liquid manure storage on dairy 1 were the VS that were pumped out of the anaerobic lagoon as part of the irrigation water. On dairy 2, the on-farm VS flow of manure and bedding to the liquid manure system was determined from the amount and composition of manure solids that were used for bedding, the percentage of recovered VS used for bedding (90% of the manure solids recovered by the manure separator were used for bedding), and the efficiency of the manure separator (40%). In addition, similar to dairy 1, VS left the liquid manure storage as part of the irrigation water. According to the producer on dairy 1, approximately 95 million L of anaerobic lagoon water was used to irrigate the surrounding fields during the 2016 growing season. On dairy 2, the irrigation water used over the growing season was estimated to be equal to the amount of wash water produced in the milking parlor and from the sprinklers in the concrete holding pen outside of the milking parlor (~110 million L). No records of the timing of irrigation events or the amount of water used during each irrigating event were available. Irrigation would have been scheduled based on evapotranspiration of the crops over the growing season and therefore vary depending on crop growth and weather. Due to this lack of data, we assumed a daily use of irrigation water in our calculation. The amount of VS flow from the irrigation water was estimated by multiplying the amount of irrigation water by the VS content of the part of the anaerobic lagoon in which the irrigation pumps were located. For dairy 1 and 2, the VS content was 8,158 and 7,824 mg of VS /L, respectively. A detailed description of the on-farm manure flows can be found in Supplemental File S4 (<https://doi.org/10.3168/jds.2017-13881>).

### Calculation of Intake and CH<sub>4</sub> Emissions

Enteric CH<sub>4</sub> emissions were estimated following the US EPA (2017a) methodology using observed DMI and dietary GE content. Details on the calculation can be

found in the Supplemental File S5 (<https://doi.org/10.3168/jds.2017-13881>). The 95% confidence interval (CI) of the enteric CH<sub>4</sub> emissions (−11 and +18%) were obtained from Table A-280 (US EPA, 2017a).

Manure CH<sub>4</sub> emissions from liquid manure storage (settling basins, anaerobic lagoon, overflow pond, and flush pond) were calculated on a monthly basis following US EPA (2017a) methodology. For both dairies, CH<sub>4</sub> emissions from liquid manure storage were estimated using the US EPA (2017a) methodology for anaerobic lagoons because US EPA (2017a) assumes that dairies that use flush systems, such as dairy 1 and 2, manage the collected manure in anaerobic lagoons. We computed CH<sub>4</sub> emissions from liquid storage for VS flow rates based on US EPA (2017a) assumptions as well as on-farm practices. The 95% CI of the CH<sub>4</sub> emissions from anaerobic lagoons (−18 and +20%) were obtained from Table A-280 (USEPA, 2017a). For more details on the calculation see Supplemental File S5 (<https://doi.org/10.3168/jds.2017-13881>).

### Methane Emission Measurements

For both dairies, ambient air temperature and relative humidity were measured on site during the monitoring period (measurements made at 15-min intervals), whereas precipitation data were retrieved from California weather databases hosted on the University of California Agriculture and Natural Resources website (UC-IPM, 2017) using a California Irrigation Management Information System station within 5 km of each dairy farm.

During the summer measurement period on dairy 1, the prevailing wind was from the south between 1200 and 1600 h and from the north between 1600 and 1200 h. The ambient temperature was  $24.6 \pm 8.1^\circ\text{C}$  (mean  $\pm$  SD), the wind speed was  $3.0 \pm 1.5$  m/s, the relative humidity was  $49.8 \pm 23.2\%$ , and precipitation was 0 mm. During the winter measurement period, the prevailing wind was from the south or north depending on the day. The ambient temperature was  $7.8 \pm 2.8^\circ\text{C}$ , the wind speed was  $4.3 \pm 2.3$  m/s, the relative humidity was  $86.1 \pm 11.3\%$ , and it rained for 6 of the 15 d of measurements (every day from Jan 20 to 26) with a total precipitation of 93 mm over the monitoring period.

During the summer measurement period on dairy 2, the prevailing wind was from the north-northwest. The ambient temperature was  $26.7 \pm 6.8^\circ\text{C}$  (mean  $\pm$  SD), the wind speed was  $3.0 \pm 1.5$  m/s, the relative humidity was  $27.5 \pm 14.5\%$ , and precipitation was 0 mm. During the winter measurement period, the prevailing wind was from the south and north or north-northwest depending on the day. The ambient temperature was  $13.9 \pm 3.0^\circ\text{C}$ , the wind speed was  $2.9 \pm 2.0$  m/s, the

relative humidity was  $90.0 \pm 9.2\%$ , and it rained for 6 of the 16 d of measurements (February 2, 3, 6, 7, 9, and 10) with a total precipitation of 38 mm over the monitoring period.

### Open-Path Measurements with Inverse Dispersion Modeling

The Northwest Irrigation and Soils Research Laboratory (Kimberly, ID) of the USDA Agricultural Research Service conducted ground measurements using open-path Fourier transform infrared spectrometry coupled with inverse dispersion modeling to estimate  $\text{CH}_4$  emission rates. On-farm measurement locations are indicated for dairy 1 and dairy 2 in Figure 1 and Figure 2, respectively. These locations were chosen to

acquire separate measurements of the  $\text{CH}_4$  emissions from discrete sources (animal housing, settling basin, anaerobic lagoon, overflow pond, and flush pond). Measurement dates and  $\text{CH}_4$  emissions (kg/d) for each location can be found in Supplemental Table S1 (<https://doi.org/10.3168/jds.2017-13881>). The concentration of  $\text{CH}_4$  was measured using open-path Fourier transform infrared spectrometry (Air Sentry, Cerex Monitoring Solutions, Atlanta, GA, and ABB-Bomem MB-100, MDA, Atlanta, GA; Griffiths et al., 2009; Shao et al., 2010). Spectra were acquired continuously and averaged over 5-min intervals. Background concentrations were measured at each dairy before the onset of the study as well as at a remote (nonagricultural affected) location for comparison. Experiments performed with the open-path units demonstrated that background

**Table 2.** United States Environmental Protection Agency (EPA; US EPA, 2017a) manure management assumptions compared with on-farm manure management practices on dairy 1 and 2

US EPA assumptions	On-farm practices
100% of dairy cow manure is captured by flush and stored in an anaerobic lagoon	28 to 100% of dairy cow manure was captured by the flush and stored in the liquid manure storage system <sup>1</sup>
Manure separator removes 40% of VS <sup>2</sup> captured by the flush water <sup>3</sup>	Not measured
Dry lots are not a manure management system for dairy cows	30% of dairy cows were managed in dry lots on dairy 1, and $\geq 40\%$ of cows had access to dry lots on both dairies
100% of replacement heifer manure is stored in dry lots	0 to 87% of replacement heifer manure remained in dry lots <sup>4</sup>
Anaerobic lagoons are not a manure management system for replacement heifers	13 to 100% of replacement heifer manure was captured by flush water and entered the liquid manure storage system
Calf manure management is not accounted for	13–100% of calf manure was captured by flush water and entered the liquid manure storage system on dairy 1 <sup>5</sup>
Anaerobic lagoons are the manure management system for dairy cows on flush dairies	Anaerobic lagoons were only part of the liquid manure storage on both dairies; manure was also stored in settling basins, flush ponds, and overflow ponds
Does not account for the use of bedding	100% of bedding was captured by the flush <sup>6</sup>
Evacuation of natural crusts on liquid storage is not part of the manure management system	Evacuation of natural crust of settling basins on dairy 1
Removal of VS from liquid storage by irrigation water is unaccounted for	9 to 13% of excreted VS were removed from liquid storage by irrigation water <sup>7</sup>
Assumes complete emptying of anaerobic lagoon once per year in October	Anaerobic lagoons have not been emptied since commissioning

<sup>1</sup>Assumes that dairy cows spend at least 6.6 h/d in areas that collect manure in liquid form, calculated as 2.5 h/d milking (personal communication with farmer) plus  $4.1 \pm 0.2$  h/d (mean  $\pm$  SD) spent feeding according to Grant (2009), Legrand et al. (2009), and Gomez and Cook (2010).

<sup>2</sup>VS = volatile solids.

<sup>3</sup>Cortney Itle, Eastern Research Group Inc., Chantilly, VA, personal communication.

<sup>4</sup>Assumes that replacement heifers spend at least 3.1 h in the feed alleyway that collects manure in liquid form. Heifers spend  $3.1 \pm 0.1$  h/d (mean  $\pm$  SD) feeding according to Greter et al. (2008), DeVries and von Keyserlingk (2009), and Greter et al. (2012).

<sup>5</sup>Assumes that calves spend the same amount of time in the feed alley as replacement heifers.

<sup>6</sup>Between 2.5 and 3.0 kg of VS/dairy cow per day were added as bedding during dry weather conditions (summer) and between 1.5 and 2.0 times that during wet weather conditions (winter).

<sup>7</sup>Anaerobic lagoon water contains VS that are removed from liquid storage when it is used for irrigation.



concentrations were stable and did not fluctuate. In addition, the on-farm concentration data at each location was filtered for wind direction to isolate times when no upwind source of CH<sub>4</sub> was present to verify that background concentrations were consistent over time. Quantitative determinations of CH<sub>4</sub> concentrations were performed by partial least squares regression of the open-path Fourier transform infrared spectra (Griffiths et al., 2009; Shao et al., 2010), and the detection limit of CH<sub>4</sub> was less than 0.01 ppmv (parts per million by volume). Concentration data were processed to produce 15-min average mixing-ratio concentrations at the source areas (*C*).

The wind environment at the dairy was described by simple Monin-Obukhov similarity theory relationships defined by  $u^*$ ,  $L$ ,  $z_0$ , and  $\beta$ , as provided by 3-dimensional sonic anemometers (RM Young Model 81000 ultrasonic anemometer, Traverse City, MI), where  $u^*$  is the friction velocity,  $L$  is the Obukhov stability length,  $z_0$  is the surface roughness length, and  $\beta$  is wind direction [see Flesch et al. (2004) for details of how these parameters were calculated from a sonic anemometer]. The sonic anemometer was placed on top of a 3-m tower at each location, where there were minimal flow disturbances from structures upwind, to capture a more idealized wind flow of the area, as suggested by Flesch et al. (2005). Wind parameters were calculated for each 15-min period (corresponding to *C* observations). Methane concentrations and on-site barometric pressure, humidity, air temperature, wind direction, and wind speed were used to calculate CH<sub>4</sub> emissions for each 15-min interval, which is described in the Supplemental File S6 (<https://doi.org/10.3168/jds.2017-13881>).

Methane emissions from all liquid storage facilities on dairy 1 (settling basins, anaerobic lagoon, and overflow pond) and dairy 2 (settling basins, anaerobic lagoon, and flush pond) were added together and reported as liquid manure storage to compare with liquid manure storage emissions estimated by US EPA methods for anaerobic lagoons. The 95% CI was determined using a bootstrapping method that resampled (with replacement) the distribution of each measurement by measurement location (housing area, settling basins, anaerobic lagoon, overflow pond, and flush pond) 10,000 times (Efron and Tibshirani, 1993).

### **Vehicle Measurements with Tracer Flux Ratio Method**

Aerodyne Research Inc. conducted ground measurements employing the tracer flux ratio (TFR) method (Lamb et al., 1995; Mønster et al., 2014; Roscioli et al., 2015) using a mini Aerodyne Mobile Laboratory (Herndon et al., 2005). Details on the mobile labora-

tory setup can be found in Supplemental File S7 and Supplemental Figure S1 (<https://doi.org/10.3168/jds.2017-13881>). Mixing ratios of various species were measured every second using 3 Aerodyne single-laser quantum cascade laser spectrometers [CH<sub>4</sub>, acetylene (C<sub>2</sub>H<sub>2</sub>), and ethane (C<sub>2</sub>H<sub>6</sub>)] and a nondispersive infrared LI-6262 gas analyzer (CO<sub>2</sub> and H<sub>2</sub>O) from LI-COR Biosciences Inc. (Lincoln, NE). Additional details of the measurement systems and analysis protocols are described in Supplemental File S7. Site-wide emissions were quantified using TFR measurements. Analysis of the relative correlation with dual tracers during the TFR measurements was used to apportion the CH<sub>4</sub> burden from the 2 major sources of CH<sub>4</sub>: animal housing and liquid manure storage. Tracer gases (C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>6</sub>) were placed strategically near individual sources (Figures 1 and 2) and released at rates ranging from 10 to 40 L/min. Using a different tracer near each emission source offered spatial distinction in real-time, with the goal of having tracer gas and CH<sub>4</sub> merge together. Optimal placement of tracers depended on wind conditions, site access and road access. Vehicle transects occurred on public roads downwind of each site at various distances (up to 6 km away). Transects used to distinguish the relative contributions of individual sources within dairy farms were typically closer than whole-facility transects, except for the settling basin on dairy 2. The close-in transects at the dairy farms involved additional uncertainty. The tracer was released at a single point, but the housing and liquid manure storage areas, the main subjects of this study, were distributed over tens or hundreds of meters. For TFR to effectively quantify area emissions with a point source release, the molar ratio of emissions to tracer must be conducted sufficiently downwind for co-dispersion to be met. Effectively, when doing the close-in transects, the method is susceptible to spatial mismatch between emissions and tracer plumes. The effect of this spatial mismatch is reduced as the transect downwind distance increases.

At dairy 2, low wind speeds and poor road access limited the quality of data for certain sections of the farm. No TFR plumes were captured for the anaerobic lagoon area during attempted close-in transects on June 21. The settling basin estimate from dairy 2 involved analysis of distant plumes by determining a fraction of each tracer's overlap with the overall CH<sub>4</sub> plume (dual-sum method). This fraction was applied to the average whole-site CH<sub>4</sub> plumes on dairy 2 to deliver an emission estimate of the settling basin. Supplemental Table S2 (<https://doi.org/10.3168/jds.2017-13881>) includes details on source area, date, times, number of plumes, and downwind distances of vehicle-based measurements at dairy 1 and 2. Supplemental Figure S2

(<https://doi.org/10.3168/jds.2017-13881>) provides an example of real-time results during a transect at dairy 2. Supplemental Figure S3 (<https://doi.org/10.3168/jds.2017-13881>) displays enhancements of tracer gas and target CH<sub>4</sub> emissions measured along various roads from ~0.8 to 6.0 km away from dairy 2. With increasing distance, tracer plumes more closely simulate the site emission plumes. Ideally, 2 tracers would be used to model a single source (to provide an internal data quality indicator), but given the presence of multiple area sources on each dairy, a single tracer was used to overlap with each source.

Uncertainties were expressed as 95% CI of the pool of valid plume intercepts. Plumes, defined as enhancements of CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, and C<sub>2</sub>H<sub>6</sub>, were validated by applying acceptance criteria described in Supplemental File S7 (<https://doi.org/10.3168/jds.2017-13881>). Essentially, this was done by comparing the downwind molar ratio of the 2 tracers to the ratio of their known flow rates. For hours containing fewer than 2 data points, method-based factor errors derived from a previous study (Roscioli et al., 2015) were employed to calculate an upper and lower 95% confidence limit. Applied to this work, these hourly averages were assigned upper and lower limit error factors of 1.85 to 0.53, respectively. For example, an hourly averaged CH<sub>4</sub> emission of 100 kg/h would be bounded by 185 to 53 kg/h at 95% confidence.

#### **Aircraft Measurements with Closed-Path Method**

The University of California, Davis, and Scientific Aviation Inc. (Boulder, CO) conducted the airborne CH<sub>4</sub> measurements of the dairies using a series of concentric, closed flight paths around the facilities. Emission rates were estimated by applying Gauss's Theorem (Conley et al., 2017). To summarize the method, the aircraft flew a series of concentric closed paths around the source, measuring CH<sub>4</sub> mixing ratio, pressure, temperature, and horizontal wind (vector components). Using pressure and temperature, the CH<sub>4</sub> mixing ratio was converted to a CH<sub>4</sub> density (kg/m<sup>3</sup>), which was then multiplied by the wind speed to achieve a CH<sub>4</sub> flux vector. The instantaneous flux into the closed path was the dot product of the CH<sub>4</sub> flux vector and the unit normal to the flight path. Next, the normal components of the CH<sub>4</sub> flux were summed around the closed path to yield the net flux per unit altitude entering the closed path. Each closed path was then summed in the vertical to yield the total net flux into the volume inscribed by the flight path. As the volume is bounded by the earth's surface at the bottom and the flight path includes altitudes above the level of the plume, the

net flux into the volume can be assumed equal to the source strength.

A total of 5 measurements (between 1100 and 1900 h) were conducted at dairy 1 and a total of 6 measurements (between 1130 and 1800 h) were conducted at dairy 2. Specifics for each of the flights can be found in Supplemental Table S3 (<https://doi.org/10.3168/jds.2017-13881>) and an example of the laps for afternoon flight at dairy 2 is provided in Supplemental Figure S4 (<https://doi.org/10.3168/jds.2017-13881>).

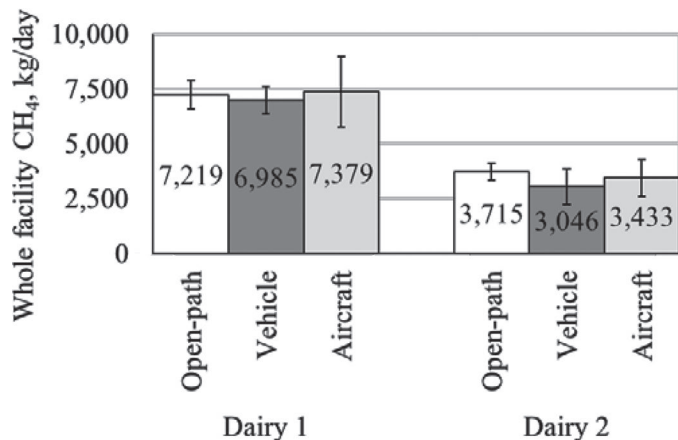
#### **Comparison Among Measurement Techniques**

Unlike the automatic daily emissions readings of open-path, both vehicle and aircraft measurements were not continuous and required the engagement of a research team during measurements. Furthermore, the aircraft measurements were limited to daytime because the boundary layer collapses at night, precluding flight at the appropriate altitude. Due to budget constraints, CH<sub>4</sub> measurements by the vehicle and aircraft technique were limited to 10 and 4 h per day, respectively. We extrapolated their hourly emissions to daily emissions by multiplying the hourly emissions by 24 to compare them with open-path measurements, which measured emissions over a 24-h period. This extrapolation is not ideal, as there can be diurnal fluctuations in emissions; however, it was the only method available to normalize the data for comparison. Uncertainties of each measurement technique were expressed as 95% CI. Statistical significance ( $P \leq 0.05$ ) was declared when the 95% CI of 2 measurements did not overlap.

## **RESULTS AND DISCUSSION**

### **All Measurement Techniques Estimated Similar Whole-Facility CH<sub>4</sub> Emissions**

Whole-facility CH<sub>4</sub> emissions estimated by the different measurement techniques (open-path, vehicle, and aircraft) were not different within each farm ( $P > 0.05$ ; Figure 3). During the summer measurement periods, on-farm emission estimates among all measurement techniques ranged between 6,985 and 7,219 kg of CH<sub>4</sub>/d and between 3,046 and 3,715 kg of CH<sub>4</sub>/d for dairy 1 and 2, respectively. The uncertainties associated with open-path, vehicle, and aircraft measurements on dairy 1 ( $\pm 9$ ,  $\pm 9$ , and  $\pm 22\%$ , respectively) and 2 ( $\pm 11$ ,  $\pm 27$ , and  $\pm 25\%$ , respectively) were greater than the variation among measurement techniques. Table 3 summarizes the characteristics of the measurement techniques. Hourly open-path estimates for the dairy cow housing (dairy 1) combined over 9 consecutive days during the winter measurements showed CH<sub>4</sub> emission peaks af-



**Figure 3.** Whole-facility methane (CH<sub>4</sub>) emissions on dairy 1 and 2 during summer measurement periods in 2016. Emissions were estimated by open-path, vehicle, and aircraft. Error bars represent the 95% CI.

ter the morning and afternoon feeding (Supplemental Figure S5; <https://doi.org/10.3168/jds.2017-13881>). These peaks were in accordance with previous studies, which found that enteric CH<sub>4</sub> emissions increased after feeding and were lower at night (Gao et al., 2011; Hegarty, 2013; Hammond et al., 2016). Consequently, the vehicle and aircraft techniques were expected to overestimate daily whole-facility CH<sub>4</sub> emissions because both measured emissions primarily during the day and not during the night. In addition, vehicle and aircraft might have captured CH<sub>4</sub> emissions from manure stockpiles (of VS recovered from the separator or lagoon evacuation) that were not captured by the open-path measurements. However, emissions from manure stockpiles were likely very low, as Ding et al. (2016) estimated that only  $23.3 \pm 14.8$  g of CH<sub>4</sub>/head per day (mean  $\pm$  SD) were emitted from manure stockpiles on a dry lot dairy in China. Nevertheless, no difference was detected ( $P > 0.05$ ) in the whole-facility emissions, perhaps because the variability associated with the dif-

ferent measurement techniques was greater than the diurnal variability in CH<sub>4</sub> emissions of the farms.

As we found no difference in whole-facility CH<sub>4</sub> emissions among the measurement techniques, it can be assumed that the open-path estimates captured all significant on-farm CH<sub>4</sub> sources on both dairies. Figure 4 shows the contribution of the different on-farm CH<sub>4</sub> sources on dairy 1.

For animal housing, vehicle estimates were numerically greater than open-path estimates (Table 4), which might have been the result of limited nighttime measurements by the vehicle when housing emissions were low. For manure storage, vehicle estimates were within 5% of open-path estimates. Manure storage estimates were similar between the 2 techniques, possibly due to the lack of a diurnal trend in manure emissions (Supplemental Figures S5 and S6; <https://doi.org/10.3168/jds.2017-13881>).

### Open-Path CH<sub>4</sub> Estimates Broadly Tracked Monthly US EPA Estimates

Open-path estimates for the 2 measurement periods (summer 2016 and winter 2017) were used to assess US EPA CH<sub>4</sub> emission predictions from animal housing and liquid manure storage. As the main objective was to evaluate US EPA prediction equations for enteric and manure CH<sub>4</sub> emissions, enteric and manure CH<sub>4</sub> emissions were estimated from on-farm DMI rather than predicted DMI using US EPA (2017a) methodology. This approach allowed us to reduce variation in US EPA (2017a) CH<sub>4</sub> predictions. This is especially important for the present study, as US EPA (2017a) emission factors are based on Holstein cattle and both of the studied dairies housed primarily Jersey cattle, which have a lower DMI (kg/d), and thus emission factor. For example, the US EPA (2017a) DMI estimate for a dairy cow (including lactating and dry cows) in California was 12.8 and 8.2% greater than the average

**Table 3.** Characteristics of methane emission measurements by open-path, vehicle, and aircraft methods in the present study

Characteristic	Measurement technique		
	Open-path	Vehicle	Aircraft
Farm access	Yes	Yes	No
Night measurements	Yes	Yes	No
Continuous 24-h measurements	Yes	No	No
Separation of emission sources	Yes	Yes <sup>1</sup>	No
Whole-facility emissions	Yes <sup>2</sup>	Yes	Yes
Uncertainty in this study, <sup>3</sup> %	$\pm 9$ to 11	$\pm 9$ to 27	$\pm 22$ to 25

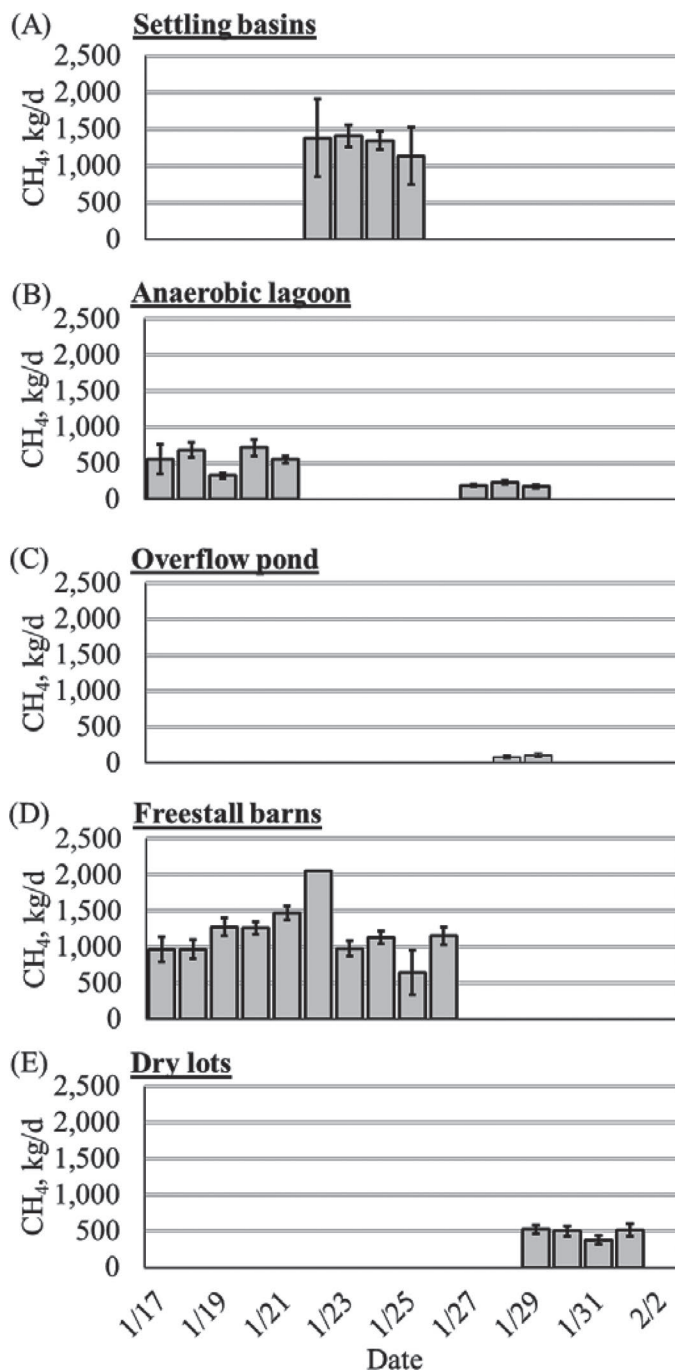
<sup>1</sup>On dairy 2, only the settling basin but not anaerobic lagoon emissions could be measured separately.

<sup>2</sup>Only as sum of the individual source areas.

<sup>3</sup>Associated with 95% CI.

DMI for dairy 1 and dairy 2, respectively (21.2 vs. 18.8 and 19.6 kg/dairy cow per day).

Daily open-path  $\text{CH}_4$  estimates from animal housing during the summer and winter measurement periods



**Figure 4.** Daily methane ( $\text{CH}_4$ ) emissions estimated by the open-path method from the different source areas on dairy 1 during the winter measurement period in 2017. The source areas presented in the figure are settling basins (A), anaerobic lagoon (B), overflow pond (C), freestall barns (housing—dairy cows) (D), and dry lots (housing—replacement heifers) (E). Error bars represent the 95% CI. When no error bar is present, only 1 measurement was conducted.

were similar within farms and to monthly US EPA estimates ( $P > 0.05$ ; data not shown). Methane emissions from animal housing are primarily from enteric fermentation, with minor contributions from manure and bedding stored in the freestall barns and dry lots. As neither farm had major changes in diet composition, DMI or cattle numbers, which are drivers of enteric fermentation, enteric  $\text{CH}_4$  emissions were expected to be comparable between summer and winter measurement periods. Similarly, no difference in summer and winter  $\text{CH}_4$  emissions from the manure and bedding in the freestall barns and dry lots were expected because they are relatively low and other authors have not observed an effect of season on these emission sources. Rahman et al. (2013) found no difference in  $\text{CH}_4$  flux rates or fluxes of beef cattle pen surfaces among months over a 7-mo study in North Dakota. Borhan et al. (2011) found no effect of season on daily  $\text{CH}_4$  emissions for a freestall dairy barn (manure lane and bedding area) and its loafing pen.

Appuhamy et al. (2016) reported that the IPCC (2006b) Tier 2 model, which the US EPA (2017a) methodology is based on, was associated with a notable mean bias for over-predicting enteric  $\text{CH}_4$  emissions from lactating cows and thus was not ranked in the top 10 models for predicting enteric  $\text{CH}_4$  emissions. However, the US EPA (2017a) methodology predicted  $\text{CH}_4$  from animal housing (enteric  $\text{CH}_4$  emissions) similar to open-path measurements in our study. Thus, the findings of the present study indicate that the US EPA methodology, which is based on IPCC (2006b) guidelines, can be used to estimate  $\text{CH}_4$  emissions from animal housing if DMI data are available. The correlation between DMI and enteric  $\text{CH}_4$  emissions has been well documented in the literature (Hristov et al., 2013; Moraes et al., 2014; Niu et al., 2018). Although these findings should be confirmed by further studies, it appears that  $\text{CH}_4$  emissions from liquid manure storage could be estimated by subtracting housing  $\text{CH}_4$  emissions, predicted from DMI and the US EPA (2017a) model, from whole-facility  $\text{CH}_4$  emissions. This would allow estimation of manure  $\text{CH}_4$  emissions in cases where measurement techniques cannot distinguish between  $\text{CH}_4$  emissions from housing and liquid manure storage because of farm layout or the technique itself.

Open-path  $\text{CH}_4$  estimates from liquid manure storage (sum of  $\text{CH}_4$  emissions of all liquid manure storage facilities on each farm) broadly tracked both monthly US EPA (based on VS flow assumptions from US EPA, (2017a) and adjusted monthly US EPA estimates [based on VS flows determined from on-farm practices; Tables 2 and 5; Supplemental File S4 (<https://doi.org/10.3168/jds.2017-13881>); Figure 5]. The monthly US EPA estimate was similar to the open-path estimates

**Table 4.** Contributions of different source areas to whole-facility methane (CH<sub>4</sub>) emissions on dairy 1 and dairy 2 estimated by open-path and vehicle measurements during the summer measurement period

Item	Animal housing, kg/d	Liquid manure storage, kg/d		
		Total	Lagoon	Settling basin
Dairy 1				
Open-path	2,330 ± 466	5,732 ± 466	1,566 ± 313	4,166 ± 833
Vehicle	2,601 ± 811	5,994 ± 579	ND <sup>1</sup>	ND
Dairy 2				
Open-path	1,275 ± 255	2,550 ± 510	220 ± 44	2,219 ± 444
Vehicle	1,636 ± 513	ND	ND	2,141 ± 637

<sup>1</sup>Not determined.

on both dairies during the summer measurement period ( $P > 0.05$ ) but not during the winter measurement period ( $P \leq 0.05$ ). During the winter measurement period, the monthly US EPA estimate was 0.6 and 2.3 times that measured on dairy 1 and 2, respectively. The adjusted monthly US EPA estimates were also similar to the open-path CH<sub>4</sub> estimates on both dairies during the summer measurement periods ( $P > 0.05$ ). During the winter measurement periods, the adjusted monthly US EPA estimate was similar to open-path estimates on dairy 1 ( $P > 0.05$ ) but was 2.6 times the open-path estimates on dairy 2 ( $P \leq 0.05$ ). Although the relative differences between the open-path measurement and the monthly and adjusted monthly US EPA estimates was large, the numerical difference was relatively small considering yearly emission estimates. It was expected that adjusted monthly US EPA estimates would be more similar to the open-path estimates, as they were based on VS flows that were calculated from on-farm practices, which were different from US EPA (2017a) assumptions (Table 2). Based on calculations of on-farm practices, we assumed that more VS were stored in the liquid manure systems than based on US EPA assumptions. On dairy 1, US EPA assumed that 66% of VS excreted on the farm were stored in the liquid manure system, whereas, based on calculations of on-farm practices, 91% of the total VS stored in

the liquid manure system (Table 5). On dairy 2, the corresponding estimates were 44 and 49%, respectively. The lower amount of VS stored in the liquid manure system (27%) based on US EPA (2017a) assumptions compared with on-farm practices on dairy 1 indicates that future studies should be conducted to validate US EPA assumptions.

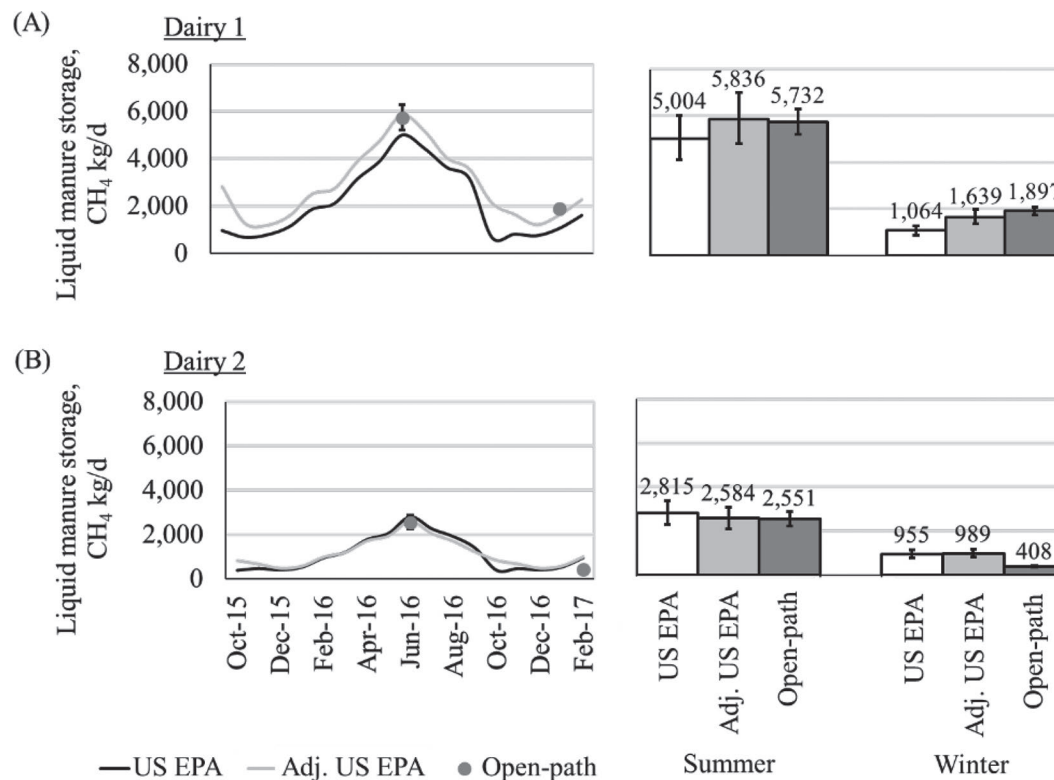
In contrast to the present study, other measurements of liquid manure storage (settling basin and anaerobic lagoon or manure tanks) in the western United States and Canada have indicated that US EPA predictions of annual manure CH<sub>4</sub> emissions may be underestimating on-farm emissions by up to 60% (Baldé et al., 2016; Leytem et al., 2017). A review of field-based studies on greenhouse gas emissions from dairy manure management also suggested that current CH<sub>4</sub> emission factors generally underestimate emissions from liquid manure-management systems (Owen and Silver, 2015).

The great variability between open-path CH<sub>4</sub> emission estimates from liquid manure storage during the summer and winter measurement periods shows that short-term measurements of CH<sub>4</sub> emissions from liquid manure systems cannot be used to evaluate annual US EPA estimates or other BU inventories. Thus, we concluded that only TD inventory estimates based on long-term measurements (Miller et al., 2013; Jeong et al., 2016) can be used to validate BU inventories, and

**Table 5.** Daily manure and bedding volatile solids (VS) flow alone and as percent of manure VS excreted on dairy 1 and dairy 2 as predicted by US Environmental Protection Agency (US EPA) methodology and on-farm practices<sup>1</sup>

Item	US EPA, kg/d		On-farm practices, kg/d		US EPA, % daily manure VS excreted		On-farm practices, % daily manure VS excreted	
	Dairy 1	Dairy 2	Dairy 1	Dairy 2	Dairy 1	Dairy 2	Dairy 1	Dairy 2
Manure VS excreted by all cattle	24,443	18,629	24,443	18,629	100	100	100	100
Manure and bedding VS								
Captured by flush	19,295	16,250	24,443	19,061	79	87	100	102
Removed by manure separator	0	6,500	0	7,624	0	35	0	41
Removed by irrigation water	0	0	2,122	2,358	0	0	9	13
Removed by lagoon emptying	3,095	1,543	0	0	13	8	0	0
Stored in liquid manure system	16,200	8,207	22,321	9,079	66	44	91	49

<sup>1</sup>Assumes the US EPA and on-farm manure management practices outlined in Table 2.



**Figure 5.** Monthly CH<sub>4</sub> emission estimates from liquid manure storage based on US Environmental Protection Agency (EPA) and on-farm volatile solid (VS) flows (adj. US EPA) compared with the open-path estimates during the summer and winter measurement periods. Graphs (A) and (B) represent CH<sub>4</sub> emissions from liquid manure storage on dairy 1 and 2, respectively. Error bars represent the 95% CI.

that TD inventories based on short-term measurements (Peischl et al., 2013; Wecht et al., 2014a,b; Trousdell et al., 2016; Cui et al., 2017) should not be used. However, publication of a spatially and temporally gridded US EPA emission inventory could allow for short-term comparisons of emission estimates.

Miller et al. (2013) and Jeong et al. (2016), using top-down approaches, both attributed greater annual CH<sub>4</sub> emissions than the BU inventory estimates (US EPA and CARB) to ruminants and dairy livestock, respectively. Jeong et al. (2016) estimated that dairy livestock emitted more CH<sub>4</sub> during winter than summer. This is the opposite of our findings, and the US EPA (2017a) inventory calculations, which predict an exponential increase of CH<sub>4</sub> emissions from liquid manure storage with increasing temperatures during summer. Although we did not measure whole-facility CH<sub>4</sub> emissions over a full year, the large on-farm variability in CH<sub>4</sub> emissions from liquid manure storage indicated that the observed discrepancies between the TD and BU inventories were most likely caused by differences in CH<sub>4</sub> emissions from liquid manure storage and not enteric fermentation, assuming that TD measurements correctly attribute emissions to dairy livestock.

#### **Open-Path Whole-Facility CH<sub>4</sub> Estimates Varied Between the Dairies**

Whole-facility, animal housing, and liquid manure storage CH<sub>4</sub> emissions for dairy 1 were greater than for dairy 2 (Table 6). Dairy 1 was expected to have greater CH<sub>4</sub> emissions because it had 1.5 times more animal units (AU; Table 1), consuming 1.4 times more DM (76,448 vs. 53,906 kg of DMI/d) and producing 1.3 times more manure VS (24,443 vs. 18,629 kg of VS/d). As DMI is the driver for enteric CH<sub>4</sub> emissions and manure VS are the substrate for manure CH<sub>4</sub> emissions, we expected differences in daily enteric CH<sub>4</sub> emissions similar to the difference in DMI, as well as a difference in daily manure CH<sub>4</sub> emissions similar to the difference in manure VS production. On dairy 1, daily CH<sub>4</sub> emissions from animal housing were 1.3 times greater than on dairy 2, and thus similar to the difference in DMI (Table 6). However, liquid manure storage CH<sub>4</sub> emissions on dairy 1 were, on average, 3.5 times greater than on dairy 2, and thus much greater than the 1.3 times difference that we expected based on the difference in VS production. Data were normalized to account for the differences in AU, DMI, and VS excre-

tion to further investigate variation in CH<sub>4</sub> emissions between the farms.

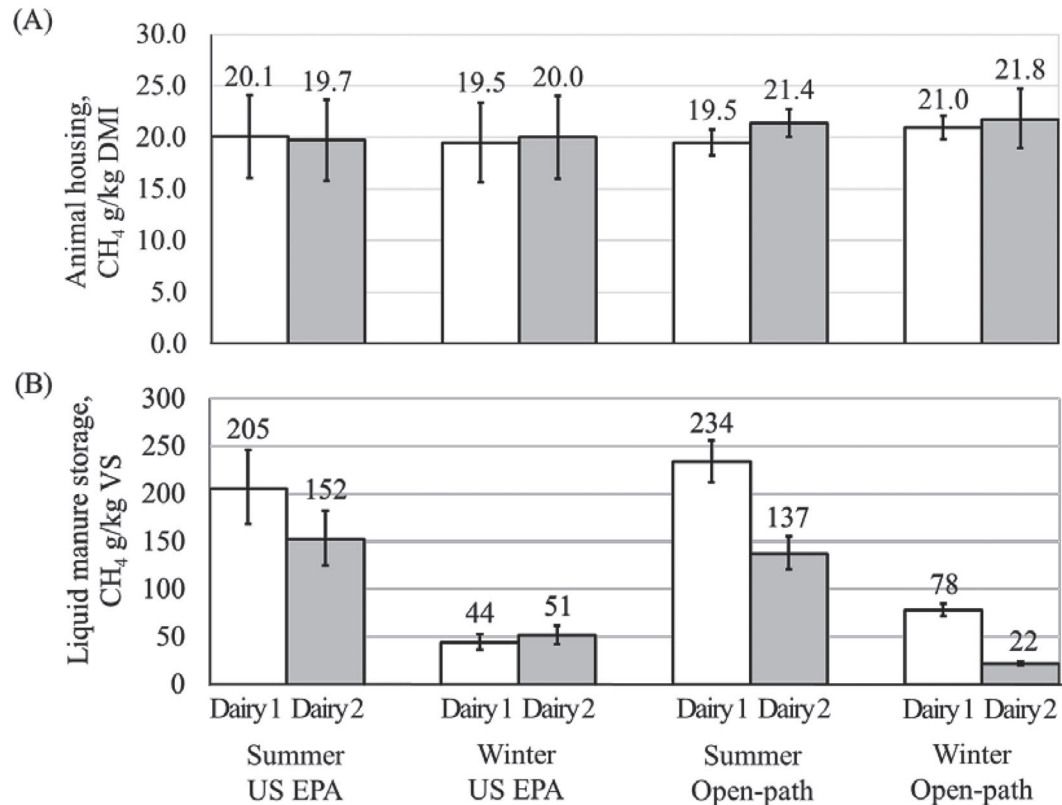
Emissions from animal housing (g of CH<sub>4</sub>/AU) were 1.2 times greater on dairy 2 than dairy 1 during the summer measurement period ( $P \leq 0.05$ ), but similar between both farms during the winter measurement period ( $P > 0.05$ ). This difference between the farms might have resulted from the relatively greater DMI on dairy 2 compared with dairy 1 during the summer measurement period (21.3 vs. 20.1 kg of DMI/lactating cow per day) compared with the winter measurement period (20.3 vs. 19.9 kg of DMI/lactating cow per day). This hypothesis is supported by similar CH<sub>4</sub> yields (g of CH<sub>4</sub>/kg of DMI) from animal housing across the farms and seasons (Figure 6). On dairy 1 and 2, CH<sub>4</sub> yields from animal housing averaged 20.2 g of CH<sub>4</sub>/kg of DMI (95% CI = 19.0–21.4) and 21.6 g of CH<sub>4</sub>/kg of DMI (95% CI = 19.4–23.6), respectively.

Emissions from liquid manure storage (g of CH<sub>4</sub>/AU) were 1.5 and 3.2 times greater on dairy 1 than on dairy 2 during the summer and winter measurement periods, respectively. Consequently, the whole-facility CH<sub>4</sub> emissions per AU on dairy 1 was greater than on dairy 2, likely driven by greater VS stored in liquid manure. Similar patterns were observed for CH<sub>4</sub> emissions normalized by VS loading from liquid manure storage [CH<sub>4</sub> (g/d)/VS (kg/d) produced by all cattle; Figure 6]. Methane emissions normalized by VS loading from liquid manure storage on dairy 1 were 1.7 and 3.5 times greater than on dairy 2 during the summer and winter measurement periods, respectively. Dairy 1 stored a relatively greater proportion of VS in liquid than dairy 2 because it did not have a manure separator and housed a greater proportion of dairy cows in freestall barns from which manure and bedding VS were collected by flushing. VanderZaag et al. (2018) found that a screw press separator that removed 78% of VS reduced the CH<sub>4</sub> emission potential by 81% on a volume basis. In addition, differences in manure handling could have contributed to the reduced emissions on dairy 2. The primary settling basin on dairy 2 was shallower (approximately 3 vs. 6 m on dairy 1) and therefore may have had a lower CH<sub>4</sub>-generation potential. According to IPCC (2006a), shallow anaerobic waste water lagoons (<2 m in depth) have a CH<sub>4</sub> conversion factor between 0.0 and 0.3, and deep anaerobic waste water lagoons (>2 m in depth) have a CH<sub>4</sub> conversion factor between 0.8 and 1.0. In addition, the settling basins on dairy 2 were cleaned out more recently; thus, they had less inoculum than the settling basins on dairy 1 and may have had reduced CH<sub>4</sub>-generation potential (Vanderzaag et al., 2013). The possibility exists that different chemicals used in the wash water from the milking parlor could have an effect on CH<sub>4</sub> emissions;

**Table 6.** Average daily methane (CH<sub>4</sub>) emissions per animal unit (AU) for the whole facility, animal housing, and liquid manure storage as estimated by US Environmental Protection Agency (US EPA) methodology and open-path measurements for dairy 1 and dairy 2 during the summer and winter measurement periods (summer 2016 and winter 2017; 95% CI appear between parentheses)

Item	Unit	Dairy 1				Dairy 2			
		US EPA		Open-path		US EPA		Open-path	
		Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter
Whole facility CH <sub>4</sub>	kg/d	6,538 (5,467, 7,816)	2,548 (2,193, 3,028)	7,219 (6,592, 7,857)	3,491 (3,250, 3,732)	3,889 (3,261, 4,648)	2,035 (1,744, 2,420)	3,715 (3,336, 4,123)	1,585 (1,399, 1,779)
	g/AU <sup>1</sup>	1,424 (1,191, 1,703)	555 (478, 660)	1,592 (1,454, 1,733)	752 (700, 803)	1,259 (1,056, 1,505)	659 (565, 784)	1,237 (1,111, 1,373)	500 (441, 561)
Animal housing CH <sub>4</sub>	kg/d	1,504 (1,339, 1,775)	1,479 (1,316, 1,745)	1,487 (1,393, 1,583)	1,594 (1,510, 1,679)	1,059 (941, 1,253)	1,077 (959, 1,271)	1,164 (1,092, 1,236)	1,177 (1,026, 1,336)
	g/AU	328 (292, 387)	322 (287, 380)	328 (307, 349)	343 (325, 361)	343 (305, 406)	349 (311, 412)	388 (363, 412)	371 (323, 421)
Liquid manure storage CH <sub>4</sub>	kg/d	5,034 (4,128, 6,041)	1,069 (877, 1,283)	5,732 (5,198, 6,275)	1,897 (1,740, 2,053)	2,830 (2,320, 3,396)	957 (785, 1,149)	2,551 (2,245, 2,886)	408 (373, 443)
	g/AU	1,097 (899, 1,316)	233 (191, 280)	1,264 (1,147, 1,384)	408 (375, 442)	916 (752, 1,100)	310 (254, 372)	849 (747, 961)	129 (118, 140)

<sup>1</sup>AU = animal unit (equivalent to 454 kg of live weight).



**Figure 6.** Summer and winter methane ( $\text{CH}_4$ ) emissions predicted by US Environmental Protection Agency (US EPA) methodology and estimated by open-path for dairy 1 and dairy 2 during the summer and winter measurement periods. Figure (A) shows animal housing  $\text{CH}_4$  emissions over DMI and (B) shows liquid manure storage  $\text{CH}_4$  emissions normalized by volatile solids (VS) loading on each dairy. Error bars represent the 95% CI.

however, we were unable to get detailed information on all chemicals used in the milking parlors.

The ratio of enteric to manure  $\text{CH}_4$  emissions on dairy 1 were 21:79 and 46:54 during summer and winter, respectively; the corresponding values on dairy 2 were 31:69 and 74:26. These results suggest that manure management on dairy 1 might have contributed more than the 55% of whole-facility  $\text{CH}_4$  emissions estimated by the current inventory (CARB, 2017).

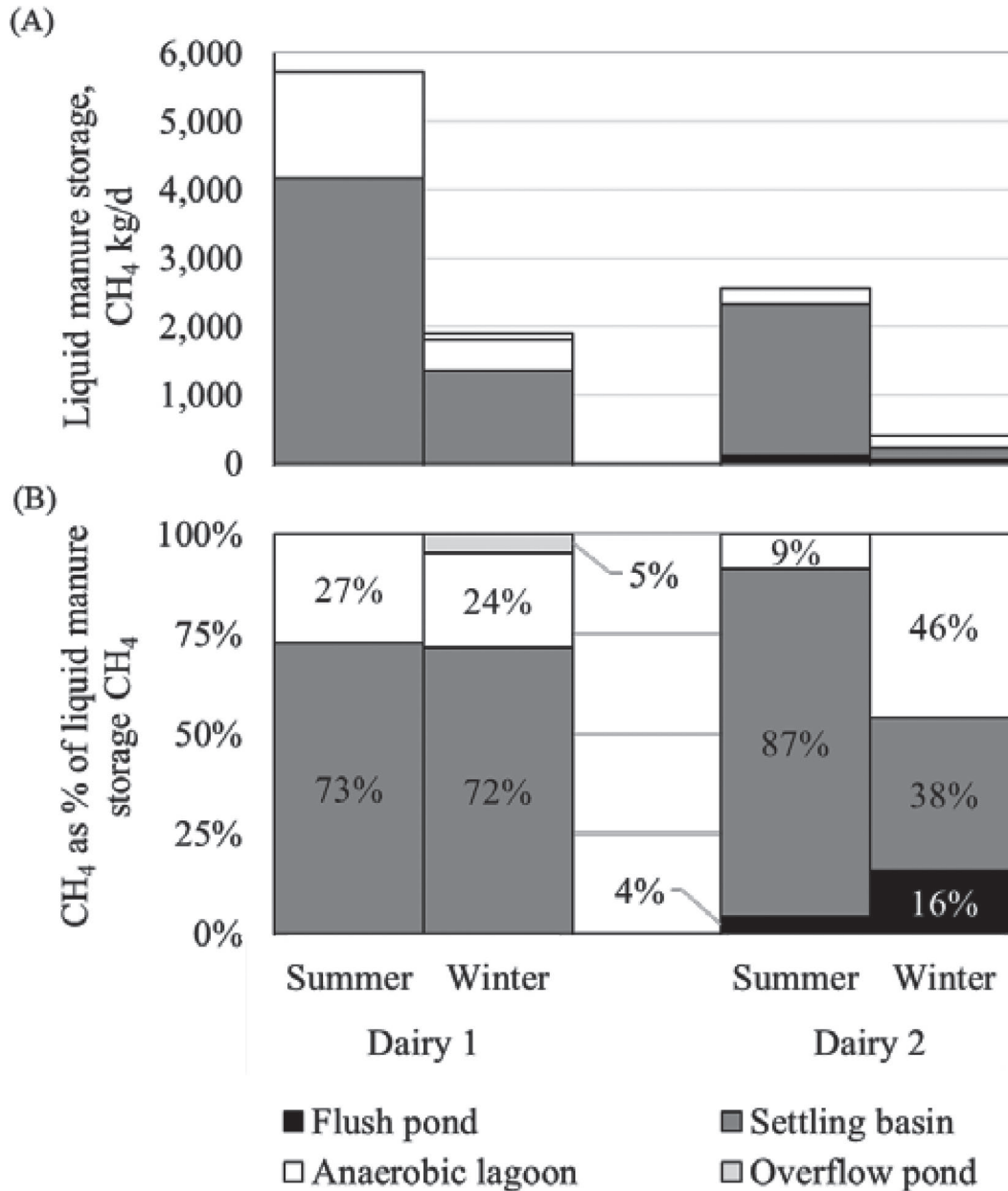
#### **Methane Emissions Varied Between and Within Source Area**

Different  $\text{CH}_4$  emissions were observed between and within on-farm sources (Figures 4 and 7). On dairy 1, open-path estimated greater  $\text{CH}_4$  emissions for the freestall barns to the west [1,148 kg of  $\text{CH}_4$ /d (95% CI = 1,106–1,192)] than for the dry lots to the east [446 kg of  $\text{CH}_4$ /d (95% CI = 404–487)]. The US EPA (2017a) methodology estimated  $\text{CH}_4$  emissions similar to the open-path estimates for the freestall barns [1,296 kg of  $\text{CH}_4$ /d (95% CI = 1,129–1,497)], but less  $\text{CH}_4$  emissions for the dry lots [188 kg of  $\text{CH}_4$ /d (95% CI = 167–222)].

A prediction equation for enteric  $\text{CH}_4$  emissions from heifers, developed by Jiao et al. (2013), estimated similar  $\text{CH}_4$  emissions to US EPA [283 kg of  $\text{CH}_4$ /d (95% CI = 201–366)] for the dry lots to the east. It is possible that manure  $\text{CH}_4$  emissions from dry lots were greater because the lots were muddy with patches of standing water. However, this hypothesis cannot be validated because dry lot emissions were not measured during the summer measurement period. In addition, it is possible that the prediction equation for heifers underestimated their emissions.

The various parts of the liquid manure storage systems on dairy 1 (2 settling basins, anaerobic lagoon, and overflow pond) and dairy 2 (1 settling basin, anaerobic lagoon, and flush pond) contributed different amounts of  $\text{CH}_4$  to the overall liquid manure storage emissions during the summer and winter measurement periods (Figure 7). On dairy 1, the settling basins were the largest source of  $\text{CH}_4$  emissions from liquid manure storage during the summer and winter measurement periods despite significantly lower total  $\text{CH}_4$  emissions from liquid manure storage during winter. On dairy 2, the settling basin was the largest source of  $\text{CH}_4$  emis-





**Figure 7.** Methane (CH<sub>4</sub>) emissions from different liquid manure storage components (settling basin, anaerobic lagoon, overflow pond, and flush pond). Emissions are shown per day (A) and as percentage of total liquid manure emissions (B).

sions from liquid manure storage during the summer measurement period, whereas the anaerobic lagoon was the largest source of CH<sub>4</sub> emissions during the winter measurement period. Leytem et al. (2017) and Borhan et al. (2011) observed that settling basins generated more CH<sub>4</sub> than anaerobic lagoons during the summer but not the winter measurement period on 2 different dairies. Leytem et al. (2017) suggested that CH<sub>4</sub> emissions from settling basins may be more affected by ambient temperature than CH<sub>4</sub> emissions from anaerobic lagoons because of their relatively smaller storage

volume. It is possible that a different emission pattern was observed on dairy 1 because 2 sequential settling basins were used, whereas dairy 2 and the dairies studied by Leytem et al. (2017) and Borhan et al. (2011) used only 1 settling basin. In addition, each of the settling basins on dairy 1 were relatively large and about twice the size of the settling basin on dairy 2. Thus, the settling basins on dairy 1 may have functioned similar to an anaerobic lagoon.

Anaerobic lagoon emissions were more than 2 times greater during the first week compared with the second

week of the winter measurements on dairy 1 (596 vs. 207 kg of CH<sub>4</sub>/d; Figure 4B). Storms occurred each day during the first week of measurements from January 18 to 22 and may have caused agitation of the anaerobic lagoon, resulting in greater CH<sub>4</sub> emissions. Baldé et al. (2016) found that agitation, such as rain events, led to increased emissions in a concrete storage tank; however, those authors noted that the increased emissions may just be over the short-term and not substantially increase overall emission rates. Baldé et al. (2016) estimated that CH<sub>4</sub> emission surges caused by agitation events increased the daily CH<sub>4</sub> emission rate by 3 to 8%. VanderZaag et al. (2014) noted that high emissions during agitation were offset by very low emissions after agitation, suggesting that the overall effect of high fluxes on total emissions is minimal. Although CH<sub>4</sub> surges just slightly increased daily CH<sub>4</sub> emissions when long-term measurements were conducted, these surges could cause an overestimation of CH<sub>4</sub> emissions during short-term measurements.

In summary, the variation between and within CH<sub>4</sub> emission sources indicate that it is important to take representative measurements of all housing and liquid manure storage facilities on a dairy. Furthermore, all liquid manure storage areas need to be assessed repeatedly to make sure that representative CH<sub>4</sub> emissions are measured and the possibility of overestimating or underestimating emissions is reduced due to CH<sub>4</sub> surges or decreased emissions after CH<sub>4</sub> surges.

## CONCLUSIONS

Whereas the number of measurements was limited in this study, we were able to compare different techniques for measuring whole-facility CH<sub>4</sub> emissions and found similar results among techniques. The open-path and vehicle techniques were able to distinguish between daily CH<sub>4</sub> emissions from animal housing (primarily enteric fermentation) and liquid manure storage, the 2 main CH<sub>4</sub> sources on the studied farms. The open-path results for CH<sub>4</sub> emissions from animal housing were similar to monthly US EPA estimates, which were predicted using farm DMI and US EPA methodology. This indicates that US EPA estimates for CH<sub>4</sub> produced by enteric fermentation may be used to predict CH<sub>4</sub> emissions from manure storage by subtracting US EPA enteric CH<sub>4</sub> estimates from whole-facility CH<sub>4</sub> emissions (housing and liquid manure storage) when separate emissions estimates are not available. Open-path estimates for CH<sub>4</sub> emissions from liquid manure storage broadly tracked monthly US EPA estimates. The proportion of CH<sub>4</sub> contributed by the different liquid manure storage facilities changed over time. On 1 dairy, US EPA assumptions estimated that 27% more

VS were stored in the liquid manure system than based on calculations of on-farm management practices. In the future, US EPA VS flow assumptions should be validated and possible improvements could be made by including different manure management systems, such as anaerobic lagoons for replacement heifers and dry lots, for dairy cows. Furthermore, CH<sub>4</sub> emissions from liquid manure should be measured on a monthly basis or more frequently throughout the year to validate the US EPA CH<sub>4</sub> prediction for liquid manure management, as other research has found that the US EPA methodology may over- or underestimate monthly CH<sub>4</sub> emissions. Substantial differences in CH<sub>4</sub> emissions from liquid manure storage (g of CH<sub>4</sub>/kg of VS) between the 2 dairies indicated that CH<sub>4</sub> emissions from California dairy farms could be significantly reduced by changes in manure management practices. However, future research is needed to evaluate the potential tradeoffs between manure stored in dry and liquid systems.

## ACKNOWLEDGMENTS

C. Arndt's postdoctoral fellowship at Environmental Defense Fund and part of A. N. Hristov's time were funded by a gift from Sue and Steve Mandel and the Kravis Scientific Research Fund (New York, NY). The measurements were funded by a gift from Sue and Steve Mandel and the Robertson Foundation (New York, NY). I. Faloona was supported by the California Agricultural Experiment Station, Hatch project CA-D-LAW-2229-H. We thank E. Kebreab, M. Hess, and their laboratories at University of California, Davis, for assistance with sample preparation and CH<sub>4</sub> analysis, respectively.

## REFERENCES

- Allen, M. S. 2000. Effects of diet on short-term regulation of feed intake by lactating dairy cattle. *J. Dairy Sci.* 83:1598–1624. [https://doi.org/10.3168/jds.S0022-0302\(00\)75030-2](https://doi.org/10.3168/jds.S0022-0302(00)75030-2) (Review).
- Appuhamy, J. A. D. R. N., J. France, and E. Kebreab. 2016. Models for predicting enteric methane emissions from dairy cows in North America, Europe, and Australia and New Zealand. *Glob. Chang. Biol.* 22:3039–3056. <https://doi.org/10.1111/gcb.13339>.
- Baldé, H., A. C. VanderZaag, S. Burt, L. Evans, C. Wagner-Riddle, R. L. Desjardins, and J. D. MacDonald. 2016. Measured versus modeled methane emissions from separated liquid dairy manure show large model underestimates. *Agric. Ecosyst. Environ.* 230:261–270. <https://doi.org/10.1016/j.agee.2016.06.016>.
- Bergamaschi, P., M. Corazza, U. Karstens, M. Athanassiadou, R. L. Thompson, I. Pison, A. J. Manning, P. Bousquet, A. Segers, A. T. Vermeulen, G. Janssens-Maenhout, M. Schmidt, M. Ramonet, F. Meinhardt, T. Aalto, L. Haszpra, J. Moncrieff, M. E. Popa, D. Lowry, M. Steinbacher, A. Jordan, S. O'Doherty, S. Piacentino, and E. Dlugokencky. 2015. Top-down estimates of European CH<sub>4</sub> and N<sub>2</sub>O emissions based on four different inverse models. *Atmos. Chem. Phys.* 15:715–736. <https://doi.org/10.5194/acp-15-715-2015>.

- Borhan, M. S., S. Capareda, S. Mukhtar, W. B. Faulkner, R. McGee, and C. B. Parnell. 2011. Determining seasonal greenhouse gas emissions from ground-level area sources in a dairy operation in central Texas. *J. Air Waste Manag. Assoc.* 61:786–795. <https://doi.org/10.3155/1047-3289.61.7.786>.
- CARB. 2017. Documentation of California's 2000–2015 GHG inventory—Index. California Air Resource Board, Sacramento, CA. Accessed Mar. 17, 2018. [https://www.arb.ca.gov/cc/inventory/doc/doc\\_index.php](https://www.arb.ca.gov/cc/inventory/doc/doc_index.php).
- CDFA. 2017. California cost of milk production Annual 2016. California Department of Food and Agriculture, Sacramento, CA. Accessed Mar. 17, 2018. [https://www.cdfa.ca.gov/dairy/pdf/Annual/2016/COP\\_Annual2016Data.pdf](https://www.cdfa.ca.gov/dairy/pdf/Annual/2016/COP_Annual2016Data.pdf).
- Conley, S., I. Faloon, S. Mehrotra, M. Suard, D. H. Lenschow, C. Sweeney, S. Herndon, S. Schwietzke, G. Pétron, J. Pifer, E. A. Kort, and R. Schnell. 2017. Application of Gauss's theorem to quantify localized surface emissions from airborne measurements of wind and trace gases. *Atmos. Meas. Tech.* 10. <https://doi.org/10.5194/amt-10-3345-2017>.
- Cui, Y. Y., J. Brioude, W. M. Angevine, J. Peischl, S. A. McKeen, S.-W. Kim, J. A. Neuman, D. K. Henze, N. Bousseres, M. L. Fischer, S. Jeong, H. A. Michelsen, R. P. Bambha, Z. Liu, G. W. Santoni, B. C. Daube, E. A. Kort, G. J. Frost, T. B. Ryerson, S. C. Wofsy, and M. Trainer. 2017. Top-down estimate of methane emissions in California using a mesoscale inverse modeling technique: The San Joaquin Valley. *J. Geophys. Res.* <https://doi.org/10.1002/2016JD026398>.
- Deshpande, B., L. Hunsaker, M. Vayssières, K. Lutter, K. Eslinger, K. Scott, and A. Huang. 2014. California's 2000–2012 Greenhouse gas emissions inventory—Technical support document. California Air Resource Board, Sacramento.
- DeVries, T. J., and M. A. G. von Keyserlingk. 2009. Short communication: Feeding method affects the feeding behavior of growing dairy heifers. *J. Dairy Sci.* 92:1161–1168. <https://doi.org/10.3168/jds.2008-1314>.
- Ding, L., Q. Lu, L. Xie, J. Liu, W. Cao, Z. Shi, B. Li, C. Wang, G. Zhang, and S. Ren. 2016. Greenhouse gas emissions from dairy open lot and stockpile in northern China: A case study. *J. Air Waste Manag. Assoc.* 66:267–279. <https://doi.org/10.1080/10962247.2015.1124058>.
- Eaton, A. D., L. S. Clesceri, E. W. Rice, and A. E. Greenberg. 2005. Standard Methods for the Examination of Water and Wastewater. 21st ed. American Public Health Association, American Water Works Association, Water Environment Federation. Port City Press, Baltimore, MD.
- Efron, B., and R. J. Tibshirani. 1993. An Introduction to the Bootstrap: Monographs on Statistics and Applied Probability. Vol. 57. Chapman Hall-CRC, New York, NY.
- Flesch, T. K., J. D. Wilson, and L. A. Harper. 2005. Deducing ground-to-air emissions from observed trace gas concentrations: A field trial with wind disturbance. *J. Appl. Meteorol.* 44:475–484. <https://doi.org/10.1175/jam2214.1>.
- Flesch, T. K., J. D. Wilson, L. A. Harper, B. P. Crenna, and R. R. Sharpe. 2004. Deducing ground-to-air emissions from observed trace gas concentrations: A field trial. *J. Appl. Meteorol.* 43:487–502. [https://doi.org/10.1175/1520-0450\(2004\)043<0487:dgefot>2.0.co;2](https://doi.org/10.1175/1520-0450(2004)043<0487:dgefot>2.0.co;2).
- Gao, Z., H. Yuan, W. Ma, J. Li, X. Liu, and R. L. Desjardins. 2011. Diurnal and seasonal patterns of methane emissions from a dairy operation in north china plain. *Adv. Meteorol.* 7. <https://doi.org/10.1155/2011/190234>.
- Gomez, A., and N. B. Cook. 2010. Time budgets of lactating dairy cattle in commercial freestall herds. *J. Dairy Sci.* 93:5772–5781. <https://doi.org/10.3168/jds.2010-3436>.
- Grant, R. 2009. Stocking density and time budgets. Pages 7–17 in 2009 Proceedings of Western Dairy Management Conference, March 11–13, Reno, NV.
- Greter, A. M., T. J. DeVries, and M. A. G. von Keyserlingk. 2008. Nutrient intake and feeding behavior of growing dairy heifers: Effects of dietary dilution. *J. Dairy Sci.* 91:2786–2795. <https://doi.org/10.3168/jds.2008-1052>.
- Greter, A. M., M. A. G. von Keyserlingk, and T. J. DeVries. 2012. Ration composition affects short-term diurnal feeding patterns of dairy heifers. *Appl. Anim. Behav. Sci.* 140:16–24. <https://doi.org/10.1016/j.applanim.2012.04.013>.
- Griffiths, P. R., L. Shao, and A. B. Leytem. 2009. Completely automated open-path FT-IR spectrometry. *Anal. Bioanal. Chem.* 393:45–50. <https://doi.org/10.1007/s00216-008-2429-6>.
- Hammond, K. J., L. A. Crompton, A. Bannink, J. Dijkstra, D. R. Yáñez-Ruiz, P. O'Kiely, E. Kebreab, M. A. Eugène, Z. Yu, K. J. Shingfield, A. Schwarm, A. N. Hristov, and C. K. Reynolds. 2016. Review of current in vivo measurement techniques for quantifying enteric methane emission from ruminants. *Anim. Feed Sci. Technol.* 219:13–30. <https://doi.org/10.1016/j.anifeeds.2016.05.018>.
- Hegarty, R. S. 2013. Applicability of short-term emission measurements for on-farm quantification of enteric methane. *Animal* 7:401–408. <https://doi.org/10.1017/S1751731113000839>.
- Herndon, S. C., J. T. Jayne, M. S. Zahniser, D. R. Worsnop, B. Knighton, E. Alwine, B. K. Lamb, M. Zavala, D. D. Nelson, J. B. McManus, J. H. Shorter, M. R. Canagaratna, T. B. Onasch, and C. E. Kolb. 2005. Characterization of urban pollutant emission fluxes and ambient concentration distributions using a mobile laboratory with rapid response instrumentation. *Faraday Discuss.* 130:327–339. <https://doi.org/10.1039/b500411j>.
- Hristov, A., J. Oh, C. Lee, R. Meinen, F. Montes, T. Ott, J. Firkins, A. Rotz, C. Dell, and A. Adesogan. 2013. Mitigation of Greenhouse Gas Emissions in Livestock Production: A Review of Technical Options for Non-CO<sub>2</sub> Emissions. Food and Agriculture Organization of the United Nations, Rome, Italy.
- Hristov, A. N., M. Harper, R. Meinen, R. Day, J. Lopes, T. Ott, A. Venkatesh, and C. A. Randles. 2017. Discrepancies and uncertainties in bottom-up gridded inventories of livestock methane emissions for the contiguous united states. *Environ. Sci. Technol.* 51:13668–13677. <https://doi.org/10.1021/acs.est.7b03332>.
- IPCC. 2006a. Wastewater treatment and discharge. Pages 6.1–6.28 in IPCC Guidelines for National Greenhouse Gas Inventories Volume 4: Agriculture, Forestry and Other Land Use. IGES, Cambridge, United Kingdom.
- IPCC. 2006b. Emissions from livestock and manure management. Pages 10.1–10.87 in IPCC Guidelines for National Greenhouse Gas Inventories Volume 4: Agriculture, Forestry and Other Land Use. IGES Cambridge, United Kingdom.
- IPCC. 2013. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom. <https://doi.org/10.1017/CBO9781107415324>.
- Jeong, S., S. Newman, J. Zhang, A. E. Andrews, L. Bianco, J. Bagley, X. Cui, H. Graven, J. Kim, P. Salameh, B. W. LaFranchi, C. Priest, M. Campos-Pineda, E. Novakovskaia, C. D. Sloop, H. A. Michelsen, R. P. Bambha, R. F. Weiss, R. Keeling, and M. L. Fischer. 2016. Estimating methane emissions in California's urban and rural regions using multitower observations. *J. Geophys. Res.* 121:13,031–13,049. <https://doi.org/10.1002/2016JD025404>.
- Jiao, H. P., T. Yan, D. A. McDowell, A. F. Carson, C. P. Ferris, D. L. Eason, and D. Wills. 2013. Enteric methane emissions and efficiency of use of energy in Holstein heifers and steers at age of six months. *J. Anim. Sci.* 91:356–362. <https://doi.org/10.2527/jas.2012-5259>.
- Lamb, B. K., J. B. McManus, J. H. Shorter, C. E. Kolb, B. Mosher, R. C. Harriss, E. Allwine, D. Blaha, T. Howard, A. Guenther, R. A. Lott, R. Siverson, H. Westburg, and P. Zimmerman. 1995. Development of atmospheric tracer methods to measure methane emissions from natural gas facilities and urban areas. *Environ. Sci. Technol.* 29:1468–1479. <https://doi.org/10.1021/es00006a007>.
- Legend, A. L., M. A. G. von Keyserlingk, and D. M. Weary. 2009. Preference and usage of pasture versus free-stall housing by lactating dairy cattle. *J. Dairy Sci.* 92:3651–3658. <https://doi.org/10.3168/jds.2008-1733>.
- Leytem, A. B., D. L. Bjorneberg, A. C. Koehn, L. E. Moraes, E. Kebreab, and R. S. Dungan. 2017. Methane emissions from dairy

- lagoons in the western United States. *J. Dairy Sci.* 100:6785–6803. <https://doi.org/10.3168/jds.2017-12777>.
- Maynard, L. A., J. K. Loosli, H. F. Hintz, and R. G. Warner. 1979. *Animal Nutrition*. 7th. McGraw Hill Inc., New York, NY.
- Miller, S. M., S. C. Wofsy, A. M. Michalak, E. A. Kort, A. E. Andrews, S. C. Biraud, E. J. Dlugokencky, J. Eluszkiewicz, M. L. Fischer, G. Janssens-Maenhout, B. R. Miller, J. B. Miller, S. A. Montzka, T. Nehrkorn, and C. Sweeney. 2013. Anthropogenic emissions of methane in the United States. *Proc. Natl. Acad. Sci. USA* 110:20018–20022. <https://doi.org/10.1073/pnas.1314392110>.
- Mønster, J. G., J. Samuelsson, P. Kjeldsen, C. W. Rella, and C. Scheutz. 2014. Quantifying methane emission from fugitive sources by combining tracer release and downwind measurements—A sensitivity analysis based on multiple field surveys. *Waste Manag.* 34:1416–1428. <https://doi.org/10.1016/j.wasman.2014.03.025>.
- Moraes, L. E., A. B. Strathe, J. G. Fadel, D. P. Casper, and E. Kebreab. 2014. Prediction of enteric methane emissions from cattle. *Glob. Chang. Biol.* 20:2140–2148. <https://doi.org/10.1111/gcb.12471>.
- Niu, M., E. Kebreab, A. N. Hristov, J. Oh, C. Arndt, A. Bannink, A. R. Bayat, A. F. Brito, T. Boland, D. Casper, L. A. Crompton, J. Dijkstra, M. A. Eugène, P. C. Garnsworthy, M. N. Haque, A. L. F. Hellwing, P. Huhtanen, M. Kreuzer, B. Kuhla, P. Lund, J. Madsen, C. Martin, S. C. McClelland, M. Mcgee, P. J. Moate, S. Muetzel, C. Muñoz, P. O'kiely, N. Peiren, C. K. Reynolds, A. Schwarm, K. J. Shingfield, T. M. Storlien, M. R. Weisbjerg, D. R. Yáñez-Ruiz, and Z. Yu. 2018. Prediction of enteric methane production, yield, and intensity in dairy cattle using an intercontinental database. *Glob. Change Biol.* 24:3368–3389. <https://doi.org/10.1111/gcb.14094>.
- NRC. 2001. *Nutrient Requirement of Dairy Cattle*. 7th rev. ed. Natl. Acad. Press, Washington, DC.
- Owen, J. J., and W. L. Silver. 2015. Greenhouse gas emissions from dairy manure management: a review of field-based studies. *Glob. Chang. Biol.* 21:550–565. <https://doi.org/10.1111/gcb.12687>.
- Peischl, J., T. B. Ryerson, J. Brioude, K. C. Aikin, A. E. Andrews, E. Atlas, D. Blake, B. C. Daube, J. A. de Gouw, E. Dlugokencky, G. J. Frost, D. R. Gentner, J. B. Gilman, A. H. Goldstein, R. A. Harley, J. S. Holloway, J. Kofler, W. C. Kuster, P. M. Lang, P. C. Novelli, G. W. Santoni, M. Trainer, S. C. Wofsy, and D. D. Parrish. 2013. Quantifying sources of methane using light alkanes in the Los Angeles basin, California. *J. Geophys. Res.* 118:4974–4990. <https://doi.org/10.1002/jgrd.50413>.
- Penn State College of Agricultural Sciences. 2017a. Holstein Growth Chart. The Pennsylvania State University, University Park. Accessed August 2017. <https://extension.psu.edu/growth-charts-for-dairy-heifers#section-1>.
- Penn State College of Agricultural Sciences. 2017b. Jersey growth chart. The Pennsylvania State University, University Park, PA. Accessed August 2017. <https://extension.psu.edu/growth-charts-for-dairy-heifers#section-5>.
- Rahman, S., M. S. Borhan, and K. Swanson. 2013. Greenhouse gas emissions from beef cattle pen surfaces in North Dakota. *Environ. Technol.* 34:1239–1246. <https://doi.org/10.1080/09593330.2012.743598>.
- Robinson, P. H., and D. Meyer. 2010. *Total Mixed Ration (TMR) Sampling Protocol*. University of California. Agriculture and Natural Resources-Publication 8413. ANR Communication Services, Davis, CA.
- Roscioli, J. R., T. I. Yacovitch, C. Floerchinger, A. L. Mitchell, D. S. Tkacik, R. Subramanian, D. M. Martinez, T. L. Vaughn, L. Williams, D. Zimmerle, A. L. Robinson, S. C. Herndon, and A. J. Marchese. 2015. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods. *Atmos. Meas. Tech.* 8:2017–2035. <https://doi.org/10.5194/amt-8-2017-2015>.
- Shao, L., P. R. Griffiths, and A. B. Leytem. 2010. Advances in data processing for open-path fourier transform infrared spectrometry of greenhouse gases. *Anal. Chem.* 82:8027–8033. <https://doi.org/10.1021/ac101711r>.
- Shindell, D., J. C. I. Kuylenstierna, E. Vignati, R. van Dingenen, M. Amann, Z. Klimont, S. C. Anenberg, N. Muller, G. Janssens-Maenhout, F. Raes, J. Schwartz, G. Faluvegi, L. Pozzoli, K. Kupiainen, L. Höglund-Isaksson, L. Emberson, D. Streets, V. Ramanathan, K. Hicks, N. T. K. Oanh, G. Milly, M. Williams, V. Demkine, and D. Fowler. 2012. Simultaneously mitigating near-term climate change and improving human health and food security. *Science* 335:183–189. <https://doi.org/10.1126/science.1210026>.
- State of California. 2016. *Short-lived climate pollutants: Methane emissions: Dairy and livestock: Organic waste: Landfills*. California Legislature, Sacramento.
- Trousdell, J. F., S. A. Conley, A. Post, and I. C. Faloona. 2016. Observing entrainment mixing, photochemical ozone production, and regional methane emissions by aircraft using a simple mixed-layer framework. *Atmos. Chem. Phys.* 16:1615433–1615450. <https://doi.org/10.5194/acp-16-15433-2016>.
- UC-IPM. 2017. California weather database. Agriculture and Natural Resources, University of California, Davis. Accessed May 2017. [http://ipm.ucanr.edu/WEATHER/about\\_weather.html](http://ipm.ucanr.edu/WEATHER/about_weather.html).
- USDA-ERS. 2017. Milk cows and production by state and region (Annual). Accessed May 2017. <https://www.ers.usda.gov/data-products/dairy-data/>.
- US EPA. 2017a. Annexes to the inventory of U.S. GHG emissions and sinks. U.S. Environmental Protection Agency. Inventory of U.S. Greenhouse gas emissions and sinks: 1990–2015. USEPA, Washington, DC, April 2017. Accessed September 2017. [https://www.epa.gov/sites/production/files/2017-02/documents/2017\\_all\\_annexes.pdf](https://www.epa.gov/sites/production/files/2017-02/documents/2017_all_annexes.pdf).
- US EPA. 2017b. Inventory of U.S. greenhouse gases and sinks: 1990–2015. USEPA, Washington, DC, April 2017. Accessed September 2017. [https://www.epa.gov/sites/production/files/2017-02/documents/2017\\_complete\\_report.pdf](https://www.epa.gov/sites/production/files/2017-02/documents/2017_complete_report.pdf).
- VanderZaag, A. C., H. Baldé, A. Crolla, R. J. Gordon, N. M. Ngwabie, C. Wagner-Riddle, R. Desjardins, and J. D. MacDonald. 2018. Potential methane emission reductions for two manure treatment technologies. *Environ. Technol.* 39:851–858. <https://doi.org/10.1080/09593330.2017.1313317>.
- Vanderzaag, A. C., T. K. Flesch, R. L. Desjardins, H. Baldé, and T. Wright. 2014. Measuring methane emissions from two dairy farms: Seasonal and manure-management effects. *Agric. For. Meteorol.* 194:259–267. <https://doi.org/10.1016/j.agrformet.2014.02.003>.
- Vanderzaag, A. C., J. D. Macdonald, L. Evans, X. P. C. Vergé, and R. L. Desjardins. 2013. Towards an inventory of methane emissions from manure management that is responsive to changes on Canadian farms. *Environ. Res. Lett.* 8:035008. <http://stacks.iop.org/ERL/8/035008>.
- Wecht, K. J., D. J. Jacob, C. Frankenberg, Z. Jiang, and D. R. Blake. 2014a. Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data. *J. Geophys. Res.* 119. <https://doi.org/10.1002/2014JD021551>.
- Wecht, K. J., D. J. Jacob, M. P. Sulprizio, G. W. Santoni, S. C. Wofsy, R. Parker, H. Bösch, and J. Worden. 2014b. Spatially resolving methane emissions in California: Constraints from the CalNex aircraft campaign and from present (GOSAT, TES) and future (TROPOMI, geostationary) satellite observations. *Atmos. Chem. Phys.* 14:8173–8184. <https://doi.org/10.5194/acp-14-8173-2014>.