A. Validation methods

The goal in applying validation methods to this dataset was to evaluate the potential level of bias present when an analyst designates plume associations (“whole site”, “animal”, and “manure”) for the purpose of separating out emissions by source. Two validation techniques were compared with the primary dataset to identify areas of potential bias: 1) a simple model employing only wind direction with aircraft position and 2) a “hands-off” analysis allowing almost all of the data collected to be considered as valid.

For the simple model, sections were drawn upon a map of each site based on their likelihood of representing a plume from a distinct source or the whole site, given a consistent wind originating from a particular range of directions (Figure S1). When the aircraft flew into one of these sections, if the wind direction at the point of highest concentration during an enhancement fell into a specified range, a source designation was made. Emission estimates for each source were summed together and compared to the main dataset. Figure S1 visualizes sections of Dairy 2 defined in the model, overlaid on top of all the transects flown by the aircraft at this site.

In contrast to the primary analysis, the “hands-off” approach analyzed a broad set of data, containing all well-correlated plumes ($R^2 > 0.5$) that had resulted in positive emissions at times when the tracer gas was flowing. Data that had undergone minor quality requirements (“minor QA”) overall provided averaged emissions that closely resemble the primary dataset for Dairy 1 (4,161 ± 535; n = 159) and Dairy 2 (6,242 ± 1,855; n = 275).

Using the minor QA dataset, this model yielded similar plume designations with a high success rate (Table S1; 115 out of 153 for Dairy 1; 65 out of 80 for Dairy 2). Within this selected set of plumes, emission estimates for Dairy 1 whole site ($97\%_{ARI}$) and cow ($94\%_{ARI}$) categories closely matched ARI findings. Model-based whole site estimates for Dairy 2 ($116\%_{ARI}$) also resembled ARI results. Given the variability and scarcity of measurements related to manure management, it is difficult to have much confidence in the manure estimates. Seeing how the model performed exceedingly well considering only a couple of basic factors, it makes sense to nominate wind direction and sampling point as major determinants of plume origin. While these factors ignore many nuances that still require human judgement, it is clear that the results tend to agree with findings from the manually filtered dataset.

B. CH_4:CO_2 ratios

Some studies utilize CO_2 measurements combined with the CH_4:CO_2 ratio to quantify enteric CH_4 emissions from ruminants (Madsen et al., 2010; Haque et al., 2014). One study compared breath samples between cows with varied feed concentrates and saw a similar ratio (~0.1 CH_4:CO_2) in both scenarios (Haque et al., 2014). Another study that measured cow breath while feeding (using two different methods to collect samples) saw a mean CH_4:CO_2 ratio of 0.09-0.11 across two experiments comparing multiple methods (Huhtanen et al., 2015). Values of CH_4:CO_2 from this study compare favorably to other studies of dairy cows (Table S1). Using all the plumes from this project provided a significant dataset of CH_4:CO_2 ratios (n = 123 at Dairy 1; n = 106 at Dairy 2) that range from 0.08 ± 0.01 (Dairy 1) to 0.13 ± 0.04 (Dairy 2).

Agreement between this study, the literature, and ARI findings indicate that not only is it possible to quickly obtain repeatable CH_4:CO_2 ratios using a mobile lab, but it is also feasible when flying above at rapid speeds. Average ratios of CH_4:CO_2 were calculated from plumes well-correlated ($R^2 > 0.5$) between CH_4 with both C_2H_6 and CO_2 (Table S1). This study provided more CH_4:CO_2 ratios than the ground-based study with similar results. Average CH_4:CO_2 ratios sorted by source in the minor QA dataset follow the manual QA dataset as well, with an average of 0.08 ± 0.01 (n = 123) at Dairy 1 and 0.13 ± 0.04 (n = 106) at Dairy 2. These ratio values only apply to whole site estimates since this dataset did not sort data into source categories.

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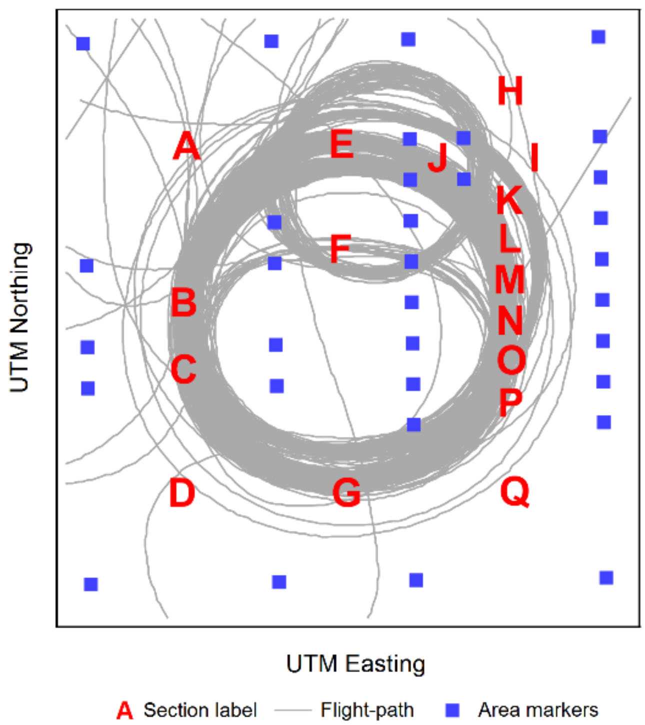

Figure S1. Map of Dairy 2 divided into sections selected to distinguish sources on-site through unique associations of wind direction and physical location.

Table S1. Comparison of methane:carbon dioxide (CH_4:CO_2) ratios between sites and measurement methods.

Site	Tracer-Plane	ARI*
	[CH_4:CO_2 ratio (ppm:ppm) ± 95% C.I. (count)]	
Dairy 1		
Whole-Site	0.13 ± 0.03 (n = 12)	0.05 ± 0.03 (5)
Animal housing	0.06 ± 0.01 (18)	0.06 ± 0.004 (7)
Dairy 2		
Whole-Site	0.12 ± 0.11 (3)	0.08 ± 0.13 (9)
Animal housing	0.04 ± 0.02 (9)	0.11 ± 0.04 (4)

*Ground-based measurement emission estimates from Arndt et al., 2018.

Table S2. Predicted methane (CH_4) emissions and methane:carbon dioxide (CH_4:CO_2) ratios from the simple model using the minor QA dataset, compared to Aerodyne ground-based measurements (Arndt et al., 2018).

Source	Count [n]	Emission [kg CH_4 d^{-1}]	Fraction* [% ARI]	CH_4:CO_2 [ppm:ppm]
Dairy 1				
Whole-Site	45	6,747	97%	0.09
Animal housing	50	2,873	94%	-
Liquid Manure	20	20,931	349%	-
Dairy 2				
Whole-Site	54	4,285	116%	0.06
Animals	-	-	-	-
Liquid Manure	11	4,758	222%	-

*Ground-based measurement emission estimates from Arndt et al., 2018.