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Supporting Information for

Top-down constraints on methane point source emissions from animal agriculture and waste based on GEM airborne measurements in the US Upper Midwest

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Text S1.

Top-down methane point source quantification method

Top-down emission estimates

The airborne sampling consists of a vertically stacked set of circuits around the facility (radius ~1 km) extending from as close to the ground as possible (typically ~60 m) through the extent of the plume (~800 m for summer/spring and ~300 m for winter). The total methane emission E (kg/h) for a given facility is then obtained via summation of the measured advected enhancements as a function of height through the plume, via:

$$E = \sum_{j=1}^{N} \left(F_j \Delta z_j \right) \quad \text{(S1)}$$

where F_j (kg/m/h) is the total advected methane enhancement for vertical layer j and Δz_j (m) is the layer height. F_j is computed for N = 4 evenly-spaced layers by interpolating the individual enhancements $f(z)$ measured on-board for the stacked flight circuits. This interpolation is performed to ensure appropriate weighting of $f(z)$ when the aircraft circuits are not evenly distributed in the vertical. In most cases, the number of individual flight circuits within a given interpolated layer j is 3-4 (range: 1-9). Each $f(z)$ is calculated following Eq. S2:

$$f(z) = \sum_{i} \left(U_{z,\perp}(i) (\rho_z(i) - \bar{\rho}_z) \Delta d_{s,z}(i) \right), z \in [z_1, z_2] \quad \text{(S2)}$$

where i indicates observational time steps, s is flight direction, and z is height above ground level (AGL) determined from on-board altitude measurements and a high-resolution elevation dataset (0.33 arc-second) from the US Geological Survey [*USGS*, 2019]. $U_{z,\perp}$ (m/s) is the wind speed component perpendicular to s at height z; $\rho_z(i)$ is the dry methane density (kg/m^3) for observation i, while $\bar{\rho}_z$ (kg/m^3) is the mean dry methane density over the individual flight circuit; $\Delta d_{s,z}$ (m) is distance between two consecutive observations (~80 m); and z_1 and z_2 (m) are respectively the lowest and highest sampled altitudes in the calculation (see Table S1). The surface-layer flux is set equal to the observed value in the lowest sampled layer j.

For most cases, the stacked flight circuits were each flown at level altitude and connected by ascents/descents while flying into the wind (as in Figure 2), thus minimizing any difference in upwind concentrations between upwind and downwind legs in case of a background vertical gradient in methane. In a few instances (08/17/2017, 08/24/2017, 01/19/2018, 01/20/2018, 01/27/2018, 01/28/2018, and 05/21/2018), the sampling followed more of a spiral configuration. In such cases we account for any methane background gradient by repeating the calculation while assuming the upwind legs to be the i) lower versus ii) upper portions of the stacked circuits, with the resulting average used for the final point source emission estimates.

In cases where the profile extended through the mixed layer into the lower free troposphere, we restrict the calculation to those observations within the mixed layer (as determined by vertical transitions in trace gases such as methane and water vapor). In the case of Dairy C on 08/17/2017, anomalous negative fluxes are derived above ~300 m AGL that are not readily attributable to meteorological effects. We assume this is due to unidentified nearby methane sources, and in this case omit >300 m AGL observations from the calculation.

Top-down emission uncertainties

We estimate the overall top-down flux uncertainty based on the individual contributions from meteorological factors, instrument error, and sampling lag. The first two (collectively ε_{meas}) are calculated following Conley et al. [2017] from the variance in the measured methane enhancements ($\sigma^2_{f(z)}$, see also Eq. S2) and the uncertainties associated with each individual flight circuit n ($\varepsilon_{cir,n}$):

$$\varepsilon_{meas} = \Delta z_j \left[\sum_j \left(\left(\sigma^2_{f(z)} + \sum_n \varepsilon^2_{cir,n} \right) / N_j \right) \right]^{0.5} \quad \text{(S3)}$$

where j indicates the vertically interpolated layers as above, N_j is the number of individual circuits for layer j, and Δz_j is layer height. The circuit uncertainties $\varepsilon_{cir,n}$ are calculated as:

$$\varepsilon_{cir,n} = f(z_n)[(\varepsilon_U/\overline{U}_n)^2 + (\varepsilon_c/\bar{c}_n)^2]^{0.5} \quad \text{(S4)}$$

where $\overline{U}_n$ and $\bar{c}_n$ are the mean wind speed and mean methane mixing ratio for circuit n, and ε_U and ε_c are the corresponding precisions of measurement (1 m/s and 1 ppb, respectively).

Here we also account for lag time uncertainty between the trace gas measurements and other quantities (wind speed and direction, position, etc.), as described in the main text. Measurement and lag time uncertainties are then added in quadrature to arrive at the total flux uncertainty. In cases where point sources were quantified more than once in a season, we use the averaged emission *E* as the best estimate with uncertainty based on the root mean square of the individual errors.

Table S1. Point Sources Quantified by GEM Flights

Type	Facility ID	Herd Size (head)	Date	Mean Wind Direction	Mean Wind Speed (m/s)	Surface Skin Temperature – 850 hPa Air Temperature (K)	Height Range (m)	Top-down Best Emissions and Uncertainty Range (kg/h)	Bottom-up Emissions and Uncertainty Range (kg/h)
Dairy	Dairy A	8,000	08/24/2017	SE (123°)	7.6	6.73	[188, 556]	98 [53, 144]	221 [170, 277]
			01/20/2018	NW (323°)	5.4	-4.42	[42, 181]	125 [92, 153]	166 [106, 218]
			01/28/2018	NW (334°)	6.2	-4.42	[56, 263]	112 [83, 141]	
			05/22/2018	SE (147°)	3.4	6.48	[169, 991]	216 [175, 260]	217 [170, 267]
			06/01/2018	SE (115°)	12.8	6.23	[117, 438]	127 [96, 168]	235 [170, 302]
	Dairy B	7,000	08/17/2017	NW (331°)	9.2	6.55	[187, 755]	28 [11, 120]	193 [149, 242]
			01/19/2018	SW (235°)	7.4	-4.13	[66, 691]	55 [39, 63]	145 [93, 190]
			01/28/2018	NW (337°)	4.8	-4.13	[30, 233]	101 [79, 122]	
			05/22/2018	SE (115°)	2.1	5.84	[90, 787]	108 [72, 143]	189 [149, 233]
	Dairy C	6,500	08/17/2017	NW (331°)	8.9	6.80	[157, 716]	26 [23, 32]	179 [138, 225]
			01/20/2018	NW (323°)	4.5	-4.30	[34, 174]	131 [107, 148]	135 [86, 177]
			01/28/2018	N (341°)	6.7	-4.30	[44, 259]	130 [106, 154]	
			05/22/2018	SE (145°)	4.3	6.61	[96, 719]	77 [47, 107]	176 [138, 217]
	Dairy D	6,500	08/24/2017	SE (116°)	9.3	6.55	[138, 439]	78 [61, 95]	179 [138, 224]
			01/19/2018	SW (232°)	5.9	-4.16	[47, 184]	148 [105, 169]	135 [87, 177]
			01/28/2018	N (346°)	4.5	-4.16	[48, 405]	116 [82, 141]	
			05/22/2018	SE (119°)	2.8	5.98	[105, 925]	45 [9, 71]	176 [138, 217]
			06/01/2018	E (110°)	13.0	5.90	[77, 423]	138 [93, 171]	191 [138, 245]
	Dairy E	6,000	01/20/2018	NW (313°)	6.5	-4.34	[48, 278]	70 [20, 94]	124 [80, 163]
			01/28/2018	NW (336°)	6.2	-4.34	[56, 349]	93 [74, 116]	
			05/22/2018	SE (144°)	3.4	6.41	[160, 1006]	169 [105, 233]	163 [127, 201]
			06/01/2018	SE (116°)	13.1	6.19	[117, 243]	80 [52, 112]	176 [127, 227]
Beef CAFO[1]	Beef CAFO[1] A	11,925	01/20/2018	NW (298°)	1.4	-4.68	[39, 174]	59 [33, 81]	70 [55, 85]
			01/28/2018	N (4°)	7.8	-4.68	[32, 219]	48 [40, 55]	
			05/23/2018	S (174°)	11.2	4.20	[99, 533]	69 [54, 83]	72 [58, 86]
			05/31/2018	NW (331°)	5.9	4.20	[101, 619]	78 [65, 91]	
	Beef CAFO[1] B	10,500	08/22/2017	NW (305°)	10.1	6.80	[167, 545]	26 [-13, 76]	61 [48, 73]
			01/18/2018	SW (239°)	5.4	-3.89	[58, 176]	2.3 [-7, 52]	58 [46, 71]
			01/27/2018	NW (305°)	6.7	-3.89	[65, 209]	74	

								[37, 88]	
			05/22/2018	S (166°)	3.0	5.78	[159, 653]	137 [107, 168]	61 [48, 73]
			05/31/2018	NW (330°)	6.5	5.78	[200, 618]	84 [57, 116]	
Swine CAFO[1]	Swine CAFO[1] A	20,080	05/23/2018	S (171°)	9.1	4.97	[134, 650]	84 [71, 104]	57 [39, 60]
			05/31/2018	NW (309°)	5.7	4.97	[141, 589]	162 [128, 195]	
			06/02/2018	NW (311°)	11.2	5.32	[148, 418]	242 [195, 285]	77 [39, 79]
	Swine CAFO[1] B	28,588	05/26/2018	W (263°)	4.0	4.64	[149, 694]	6 [-12, 37]	78 [51, 86]
Sugar Plant	Sugar Plant A		08/17/2017	NW (334°)	6.9	7.69	[249, 727]	146 [128, 160]	473 [464, 622]
			01/19/2018	NW (298°)	9.5	-5.92	[112, 296]	38 [-11, 96]	471 [463, 620]
			05/21/2018	SE (137°)	6.6	6.78	[161, 544]	161 [126, 198]	471 [463, 620]
	Sugar Plant B		01/19/2018	NW (307°)	11.6	-6.69	[63, 818]	41 [31, 53]	569 [424, 574]

[1]CAFO: concentrated animal feeding operation.