Assessment of Regional Methane Emissions Inventories through Airborne Quantification in the San Francisco Bay Area

SUPPLEMENTARY INFORMATION

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APPENDIX 1 – AIRBORNE FLUX ESTIMATION METHOD

Airborne Mass Fluxes

In this study, emission rates are measured using an airborne mass balance technique that combines trace gas concentrations of methane, carbon dioxide and/or ethane with coincident meteorological measurements. This project deployed an instrumented Mooney aircraft owned and operated by Scientific Aviation to conduct emissions measurements from target sources. The aircraft is outfitted with a data system that measures and records real-time aircraft speed, position, altitude, and horizontal winds (accuracy < 0.5 m s-1), as well as ambient temperature, pressure, and relative humidity. A flight-optimized Picarro (Santa Clara, CA) cavity ringdown spectrometer and an on-board data system provide fast-response, in-situ concentration measurements of CH_4, CO_2, and water vapor. Many of the flights also included instrumentation to measure ethane (C_2H_6) concentrations to provide a reliable source marker for fossil-fuel derived CH_4 sources, since decomposition-induced biological sources of CH_4 such as landfills, wastewater treatment and composting do not emit C_2H_6. We use the C_2H_6 data qualitatively to confirm the source / origin of the observed CH_4 emissions.

The method used by Scientific Aviation and its application to quantify emissions from facilities and sources have been described previously in greater detail.[25,26] In brief, the measurement approach is based on mass balance principles and Gauss's Law as described in

Conley et al.[25] For each target site, the aircraft flies a closed loop pattern around a facility, beginning at the minimum safe altitude (often ~60m above ground level) and climbing progressively higher, until real-time data indicate the aircraft is above the plume (Figure S1). Additional loops are flown above the plume to confirm the extent of vertical mixing. The flight path traces a cylinder around the target site and using the observed horizontal wind and trace gas concentration enhancements (upwind-minus-downwind concentrations over background), Gauss's Law is applied to estimate the flux divergence through the cylinder. Details of the calculations to estimate the average emission rate for each sampled site using mass-balance horizontal flux divergences are presented elsewhere.[25,26]

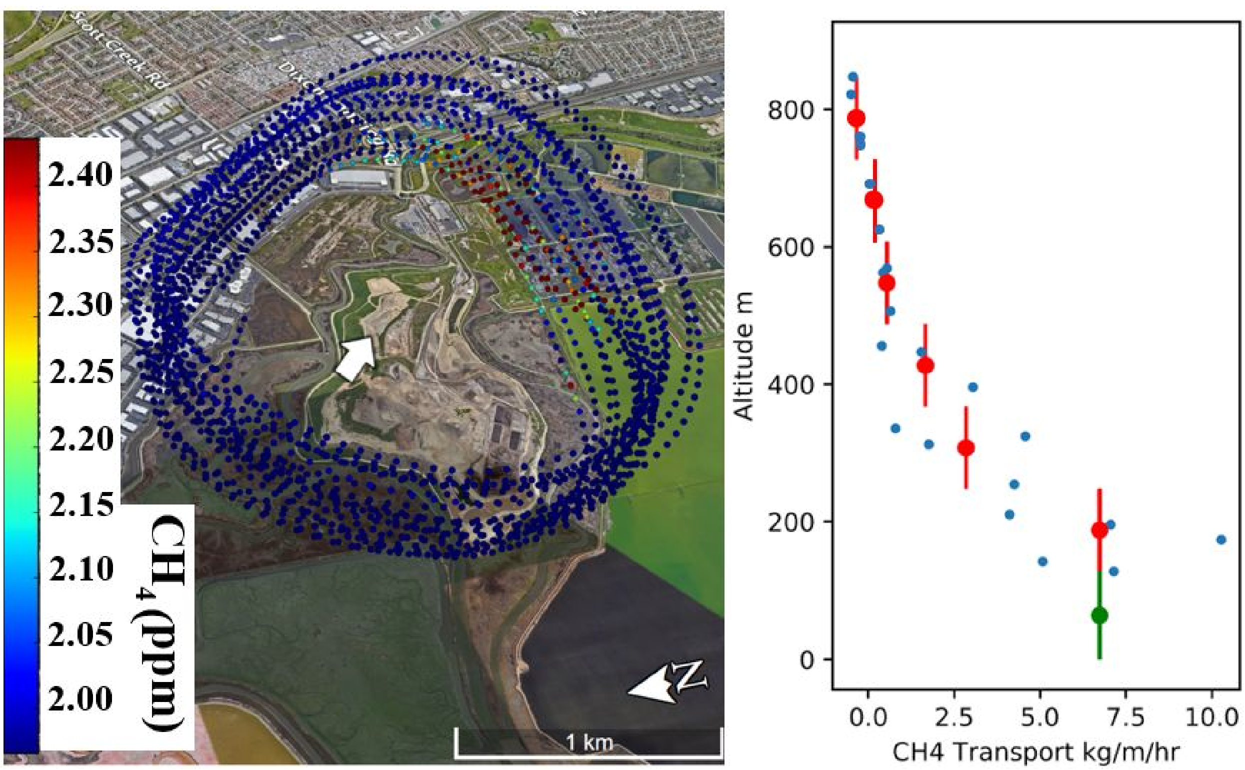

Figure S1. Example aircraft data (A) Flight path colored by methane concentrations over Newby Island landfill, showing the enhancements in CH_4 are downwind of the landfill (white arrow indicates wind direction at the time of the flight); and, (B) Vertical profile of horizontal methane flux divergences for the flight path where blue circles represent each horizontal loop's flux divergences, red bars and circles represent the altitude bins and binned averages, respectively, and the green circle is the assumed flux divergence for the layer between the lowest flight altitude and the surface representing the - "ground contribution term".

The flux contribution of the emission plume flowing underneath the lowest aircraft sampling altitude and the ground surface is estimated by investigating the Large Eddy Simulation (LES) flux divergence profiles in the lowest layer and evaluating the accuracy of the approach against coordinated controlled release and real-time power plant CO_2 emissions releases.[25]

Uncertainty Estimation

Controlled release tests have been applied before to verify the accuracy of the cylindrical mass balance method and have generated accuracies within 15% or better with an average accuracy of 6%.[25] Since field-measurement flights often take place in less than optimal conditions as compared to the controlled release tests, the Scientific Aviation method estimates uncertainties for each target measurement. The main uncertainty term arises from the prediction of flux below the lowest flight altitude, and this is minimized by prior LES studies that have been used to determine a downward distance that provides very little change in the plume flux divergence from the lowest loop to the ground.[25] The uncertainty in the final emission rate produced by the Scientific Aviation method is determined by summing in quadrature terms representing a binning error from the standard deviations of the horizontal flux divergences for the individual closed-path loops within each altitude bin (assumed to provide an estimate of variability from turbulence), error due to the number of loops flown around a facility, uncertainty term due to not capturing the horizontal efflux below the lowest leg of the aircraft profile, and the uncertainty due to the lack of consistency of the wind velocity during the observations over a target facility.[25,26] The uncertainty in horizontal flux for each closed-loop path is estimated from the uncertainty in the wind measurement and the uncertainty in the chemical species measurement, summed in quadrature.[25]

The successful application of the Scientific Aviation method for quantifying emissions relies upon favorable wind conditions and adequate vertical mixing. Ideal wind conditions are moderate and steady in both speed and direction. Completely calm conditions, very high wind speeds, or rapidly varying wind directions are not ideal for this method and introduce large errors. The highest degree of confidence in the results require sampling most of the plume at and above the lowest flight altitude and below the top of the boundary layer to ensure complete sampling of the entire plume. Under ideal conditions, emissions uncertainties ≤10% can be achieved. Conditions that are less than ideal increase the uncertainty in the resulting flux. For this study, the mean of the individual emissions uncertainties across all site-assessments was 25% with a range of 9-76%.

APPENDIX 2 – ESTIMATION OF ACTUAL DAY-TO-DAY VARIATION

Although not shown, there was no appreciable trend or seasonality. The data were clustered in time, specifically during the 2nd and 4th quarters of the calendar year; there were only two observations in the 1st quarter and none in the 3rd. There was significant day-to-day variation, however, on the order of four times the measurement error variance. This analysis addresses the question of whether there is evidence of day-to-day variation in refinery emissions evident above measurement error. We note that as long as this variation is “random” in the sense that the choice of days the airplane flights occurs is independent of the actual emissions, then the above estimates should be valid.

There are several approaches. We can analyze the data on the original, linear scale or the log scale, and we can consider the uncertainties as fixed and known or themselves only estimates of the true measurement errors.

Approach 1 – Original scale

Each of the 41 measurements came with a corresponding uncertainty estimate. Let observations from a particular refinery be $Y_1, Y_2, \ldots, Y_n$, and let $u_1, u_2, \ldots, u_n$ be the corresponding uncertainties. Assume the Y_j have the same mean, μ, but different variances, σ_j^2, $j=1,\ldots,n$. let $S = \sum_{j=1}^{n}(Y_j - \bar{Y})^2$. Then, after some algebra, the uncertainty, E(S), is given by $E(S) = \frac{n-1}{n}\sum\sigma_j^2$ or $\frac{E(S)}{n-1} = \overline{\sigma^2}$. If all the σ_j^2 were equal ($\sigma_j^2 \equiv \sigma^2$), and σ^2 were known, then S/σ^2 would have an approximately chisquare distribution. If we instead divide S by the mean of the σ_j^2, then $S/\overline{\sigma^2}$ would likely be longer-tailed, but perhaps a chisquare distribution would be a good first approximation, an upper bound, at least. Alternatively, to roughly account for the fact that the u_j are also estimates of σ_j, we could assume that $F = \frac{S}{(n-1)\overline{u^2}}$ has an F distribution, with n-1 df in the numerator, and n df in the denominator. The table below shows the results by refinery, and also where the refineries are pooled.

Table S1. Summary of statistical tests run on the refinery emissions data.

Tests for actual variation					**p-values**	
Refinery	**# obs**	**S/(n-1)**	$\overline{u^2}$	**F**	**Chisquare**	**F**
Marathon	7	29,244	8,182	3.6	0.002	0.073
Chevron	7	28,938	28,233	1.0	0.407	0.488
Phillips	10	29,977	15,158	2.0	0.038	0.162
Shell	6	343,928	19,519	17.6	<0.001	0.003
Valero	11	23,618	11,324	2.1	0.022	0.131
Combined	41	75,760	15,809	5.1	<0.001	<0.001

Individual tests are clearly significant only for the Shell refinery, though borderline significant for Marathon and Valero. However, combining all five refineries does produces a highly significant result and an F-statistic of roughly 5. But note that the numerator of Approach 1 incorporates the uncertainty variance plus the true day-to-day variance. This suggests that the day-to-day variance is roughly 5 -1 = 4 times the measurement error.

Approach 2 – Log scale

As demonstrated in Appendix 3, the data appear to fit a log-normal model reasonably well. The model incorporates both measurement error, Var(U), and the actual day-to-day variance, Var(W). The fitted model estimated a true variance of $\hat{\sigma}^2 = 0.347$. This compares with an estimated mean of the log uncertainties = 0.09. The estimated ratio of Var(W) to Var(U) is thus 0.347/0.09= 3.8. This ratio is similar to that found in Approach 1.

APPENDIX 3 – ANALYSIS OF REFINERY METHANE EMISSIONS DATA

There are 41 aircraft measurements from the 5 Bay Area refineries from which to develop estimates of their total hourly average methane (CH_4) emissions and the uncertainty of these estimates. Two statistical methods yield essentially the same estimates, and simulations also show a similar range of values as traditional confidence intervals. We estimate total CH_4 emissions from the five refineries at 19,000 mt/yr, with a 95% confidence interval of 15,500 mt/yr to 22,800 mt/yr.

Data

A total of 41 measurements were made between 2015 and 2018. Figure S2 shows a dot plot by refinery.

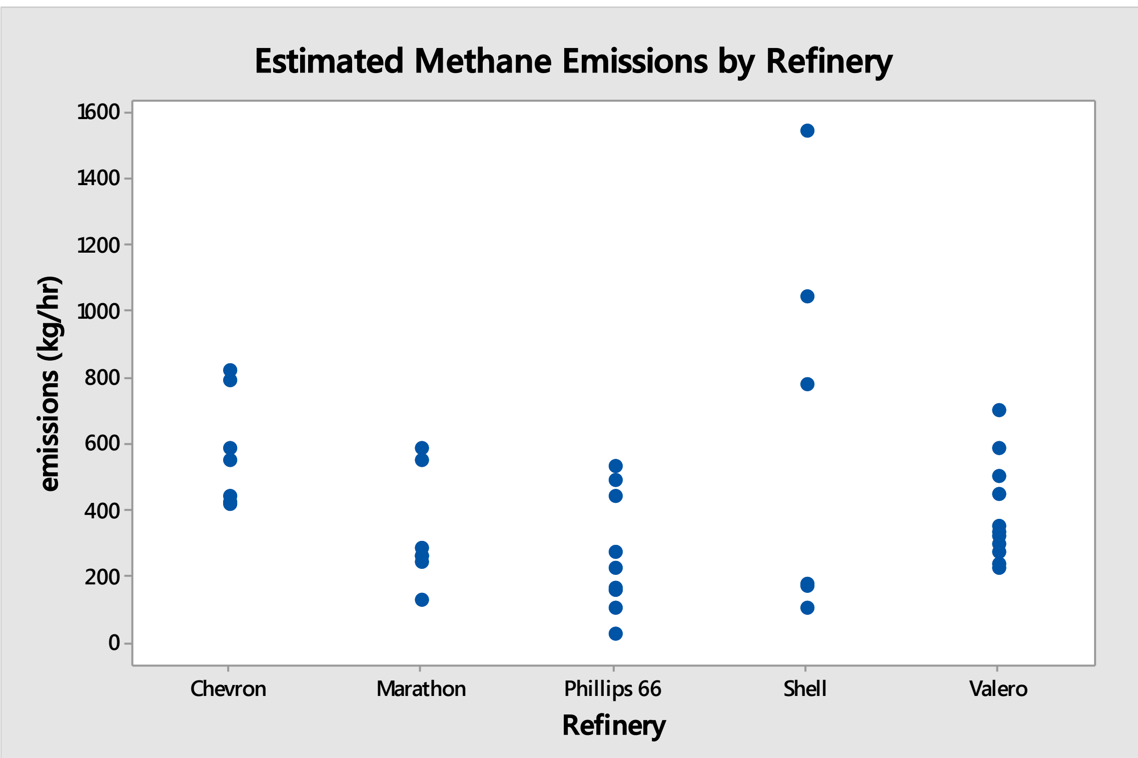

Figure S2. Estimated Methane Emissions by Refinery.

Estimation Methods

The goal was to estimate total emissions, T, from the 5 refineries, or $T = m_1 + m_2 + m_3 + m_4 + m_5$, where m_i = estimated mean hourly CH_4 emissions from the ith refinery. The simplest estimate would assume that the emissions rates from the refineries were equal: $m_i \equiv m$, and estimate the total emissions as $T = 5m$, where m is the arithmetic mean of the 41 measurements. However, a 1-way analysis of variance found statistical differences among the refineries ($p=0.044$). Figure S3 shows the refinery means and 95% confidence intervals. Thus, from this point, we do the analysis estimating separate means for each refinery.

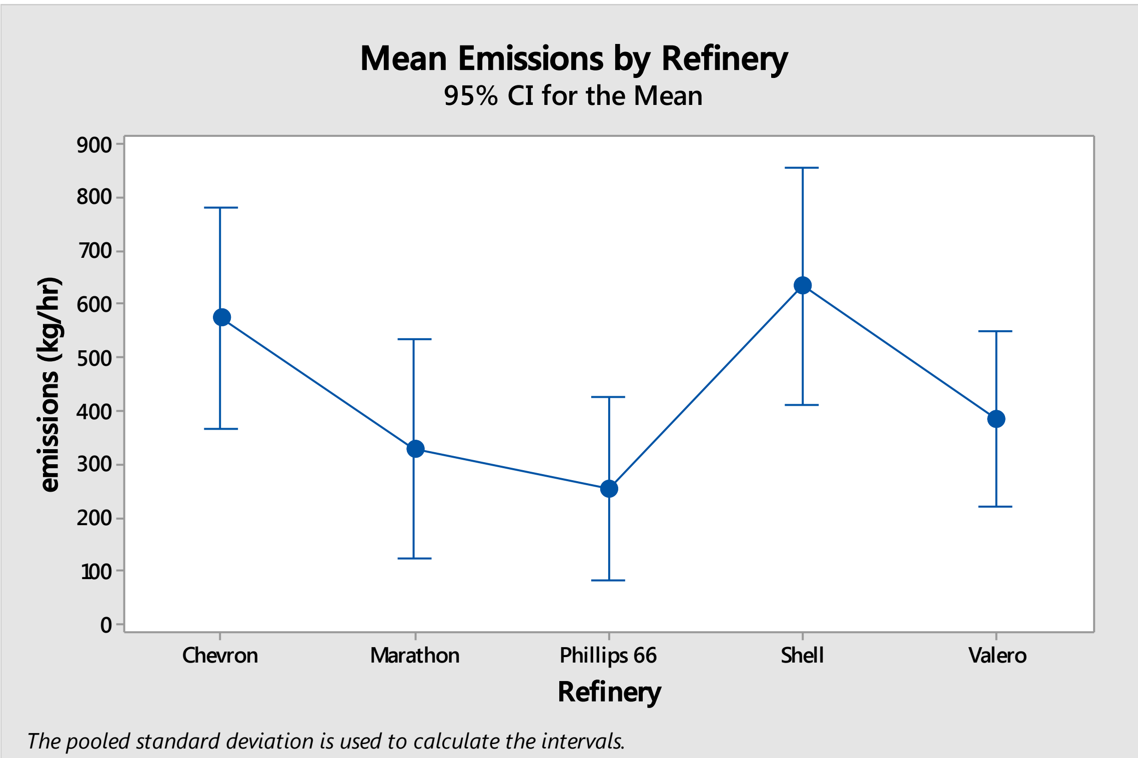

Figure S3. Means and 95% confidence intervals by refinery.

Because the measurement uncertainties differ, we considered using mean estimates with inverse variance weighting, which can produce more efficient estimators. But, as shown later in Figure S5, the uncertainties are correlated with the individual values. Thus, a weighted average would produce biased results. So, here we use simple arithmetic means.

Let $\bar{y}_i$ be the arithmetic mean of the n_i measurements from refinery i, and estimate $T = \bar{y}_1 + \bar{y}_2 + ... + \bar{y}_5$. Assuming that the variances are equal, we can compute a pooled variance estimate: $s^2 = \frac{\sum_{i=1}^{5}\sum_{j=1}^{n_i}(y_{ij} - \bar{y_i})^2}{41 - 5}$, where y_{ij} is the jth observation from refinery i. The estimated standard error of T is the square root of $\tau^2 = s^2\sum_{i=1}^{5}\frac{1}{n_i}$. This method provides an unbiased estimator as long as the observations can be considered to be collected at random. A 100*(1-α)% confidence intervals based on the t-distribution is:

$$T \pm t_{41,\alpha/2} * \tau \quad (1)$$

Bootstrap estimate of uncertainty

We simulate T by sampling from each refinery's observations with replacement. Specifically, we can randomly select y_{ij}^* from $\{y_{i1}, y_{i2}, \ldots, y_{in_i}\}$, with replacement, $j=1,\ldots,n_i$, for $i = 1,\ldots,5$, (in other words, generating new random samples of size 41 from the original data) and compute $T^* = \bar{y}_1^* + \ldots + \bar{y}_5^*, where\ \bar{y}_i^* = \frac{1}{n_i}\sum_{j=1}^{n_i} y_{ij}^*$. This yields the same estimated mean and variance but provides different (and probably more realistic) lower and upper confidence interval bounds due to the skewness of the underlying data.

Parametric approach

Generally, we can get more efficient estimators (that is, having less uncertainty) if we know the approximate distribution of the data and we use a parametric approach tailored to that distribution. Also, for these data the individual measurements have different uncertainties, so it would be preferable if these could be incorporated into the analysis.

The simplest situation would be if the data were normally distributed. A normal scores plot[1] suggests the data are right-skewed, i.e., non-normal (Figure S4). Because the data are a mixture of distributions with different means, a simple test of normality such as the Shapiro-Wilk test can't be applied. Instead we constructed a test using the mean of Shapiro-Wilk statistics for each refinery, i.e., $S = \sum_{i=1}^{5} W_i/5$, where W_i is the Shapiro-Wilk statistic for

[1] A normal scores plot shows the data versus the expected values of the order statistics from a sample of the same size of standard normal random variables. If the data were normal, the points should lie close to a straight line. If the points are concave as in Figure S4a, this indicates the data are right-skewed.

refinery i, i=1,…,5. We simulated the value of S 100,000 times, using pseudo-random normal variates. The observed value of S was 0.87, with a p-value of 0.037, based on the simulation.

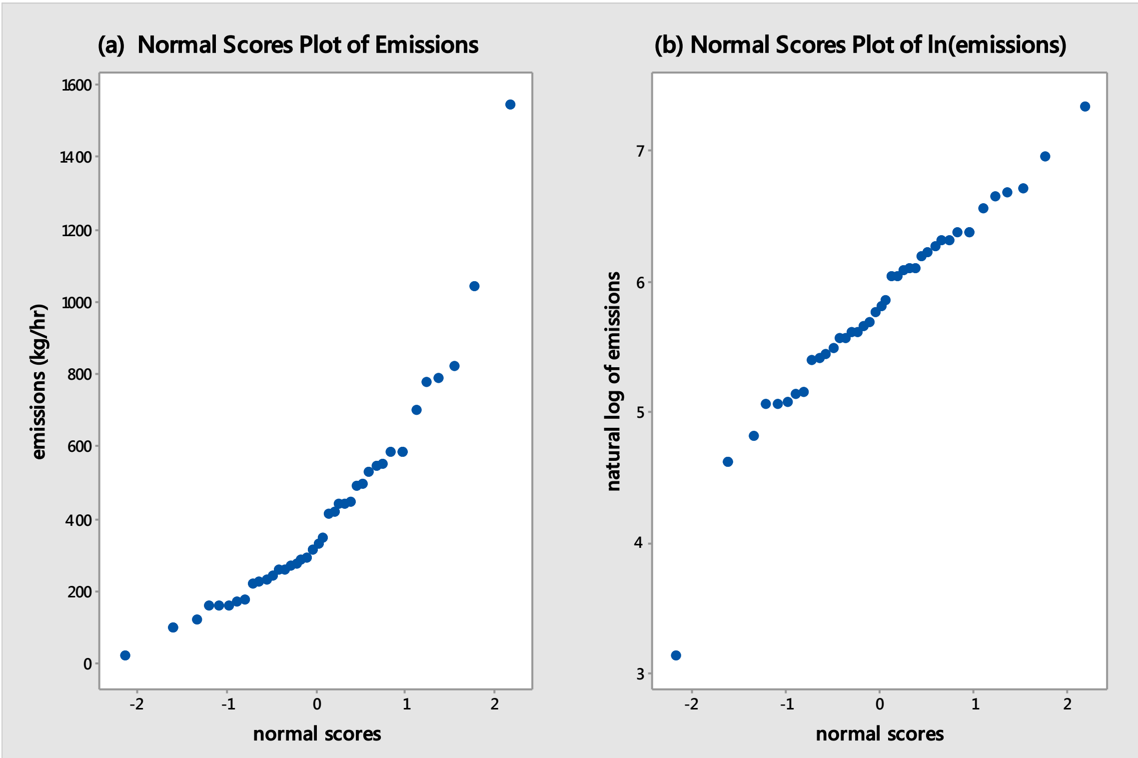

Figure S4. Normal scores plots of the (a) estimated emissions, and, (b) their log values.

For the natural logs of the data (Figure S4b), the figure doesn't suggest non-normality. The value of S for the logs of the data was 0.90, with a p-value of 0.17. Here, normality can't be rejected, so we assume the data are log-normally distributed.

A second issue is how to deal with the uncertainties. Figure S5a shows that uncertainties increase as emissions increase. Figure S5b shows the ratios of uncertainty to emissions. Here there is no significant correlation, so that the percent uncertainty appears independent of the magnitude of the emissions.

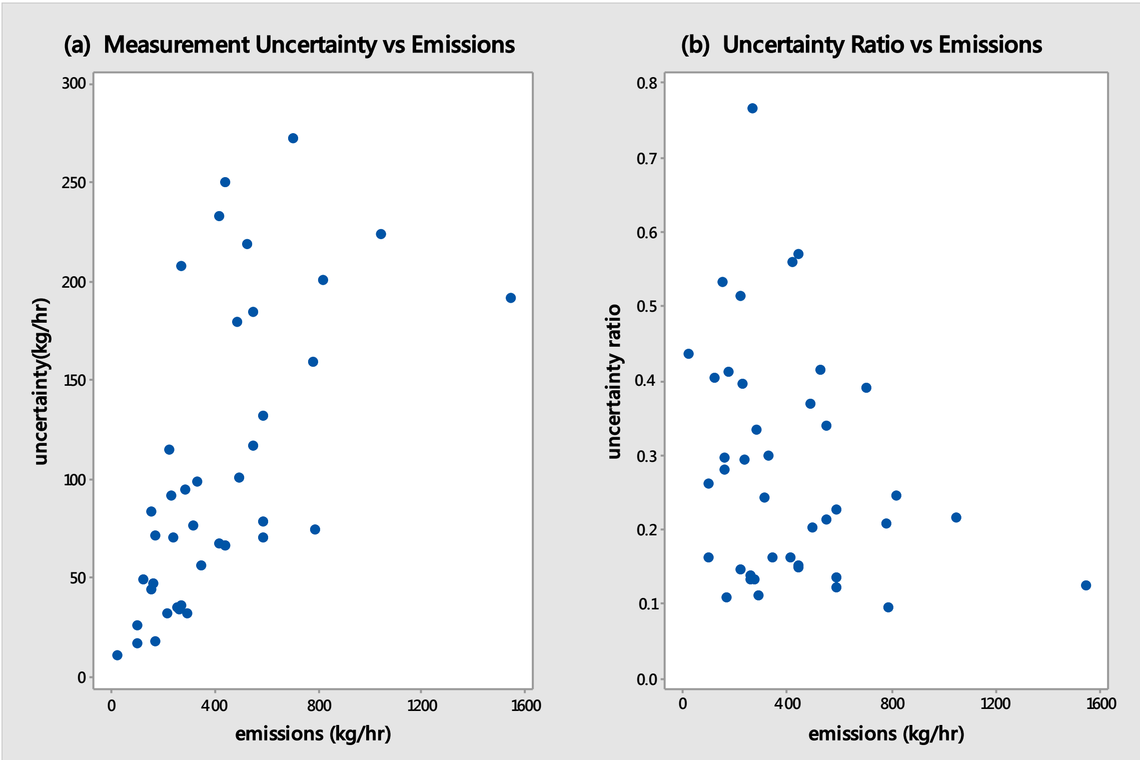

Figure S5. Estimated refinery emissions plotted against (a) absolute value of uncertainties, and, (b) uncertainties as percentage of magnitude of emissions.

Given the results of Figure S5, it is reasonable to model the measurement error as multiplicative, that is, that a measured value is the product of the true emissions times measurement error. This dovetails with the assumption that the data are log-normally distributed, since the error becomes additive on the log scale. We model the data on the original scale as Y = WU, where U = measurement error and W = true (but unobserved) actual emissions. Assume that both W and U are log-normally distributed and that they are statistically independent. Also assume that E(U) = 1, so that the measurement provides an unbiased estimator of W, that is, E(Y|W) = W. The goal will be to estimate E(W).

Since U is lognormally distributed, $E(U) = e^{\vartheta + \frac{\sigma^2}{2}} = 1$ by assumption. Or, taking logs, $\vartheta + \frac{\sigma^2}{2} = 0$, so that we need $\vartheta = -\frac{\sigma^2}{2}$. With this constraint, we have $Var(U) = \left(e^{\sigma^2} - 1\right)e^{2\vartheta + \sigma^2}$ $= e^{\sigma^2} - 1$. We've been given the measurement error of Y, say σ_Y, which suggests that the measurement error (sd) of U given Y would be σ_Y/Y. Combining this with Var(U), we get $Var(U|Y) = e^{\sigma^2} - 1 = \frac{\sigma_Y^2}{Y^2}, or\ \sigma^2 = \ln\left(1 + \frac{\sigma_Y^2}{Y^2}\right)$. Let's first consider an individual refinery with n observations. Let $\ln(W_j) \sim \text{Normal}(\mu, \sigma^2)$, j = 1, 2, …, n. Based on the assumptions above, $\ln(U_j) \sim \text{Normal}(-\frac{\sigma_j^2}{2}, \sigma_j^2)$, where $\sigma_j^2 = \ln\left(1 + \frac{\sigma_{Y_j}^2}{Y_j^2}\right)$, for the jth observation. We take σ_j^2 as known. Then X=ln(Y) = ln(W) + ln(U) is the sum of two independent normal random variables so its distribution is also normal, with mean $\mu - \frac{\sigma_j^2}{2}$, and variance $\sigma^2 + \sigma_j^2$. We can write the joint likelihood of the X_j as: $L = \prod_{j=1}^{n} \frac{1}{\sqrt{2\pi(\sigma^2 + \sigma_j^2)}} e^{-\frac{(x_j - \mu + \frac{\sigma_j^2}{2})^2}{2(\sigma^2 + \sigma_j^2)}}$. Taking logs:

$$l = \ln(L) = -\frac{n}{2}\ln(2\pi) - \sum_{j=1}^{n} \ln(\sigma^2 + \sigma_j^2) - \sum_{j=1}^{n} \frac{(x_j - \mu + \frac{\sigma_j^2}{2})^2}{2(\sigma^2 + \sigma_j^2)}.$$

Taking the derivative of the log likelihood with respect to μ and setting it equal to 0, we can find its maximum likelihood estimator (mle): $\hat{\mu} = \sum_{j=1}^{n} u_j(x_j + \frac{\sigma_j^2}{2})$, where $u_j = \frac{1}{\hat{\sigma}^2 + \sigma_j^2}\left[\sum_{k=1}^{n} \frac{1}{\hat{\sigma}^2 + \sigma_k^2}\right]^{-1}$, $and\ \hat{\sigma}^2\ is\ the\ mle\ of\ \sigma^2$. Since $E(W) = e^{\mu + \frac{\sigma^2}{2}}$, the mle of E(W) is just $e^{\hat{\mu} + \frac{\hat{\sigma}^2}{2}}$.

This approach could be applied, computing each mean separately although this involves estimating 10 parameters with only 41 observations. Another approach is to estimate μ separately by refinery but assume that σ^2 is the same. This requires estimating 6 parameters – still a lot, but not unreasonable. We call this estimate T_2. An R script was written to compute the estimates of the μ_i and σ^2 by maximizing the log likelihood. Calculating the uncertainty of this estimator is difficult. Instead we derived a confidence interval by simulating the distributions of W and U.

Results

Estimate 1. Using separate arithmetic means for the 5 refineries. Here the estimated T_1 = 2,171, with a pooled standard deviation estimate of 267. The estimated se is: 214. Using equation (1), a 95% confidence interval for T_1 is 1,720 kg/hr to 2,600 kg/hr.

Bootstrap simulation of Estimate 1. Based on a simulation of 10,000 bootstrap samples T_b has a 95% confidence interval of 1,720 kg/hr to 2,670 kg/hr.

Estimate 2. Assuming observations log-normally distributed with different μ and same σ^2 across refineries. The estimated μ_i were 5.72, 6.39, 5.33, 6.02, and 5.90 for Marathon, Chevron, Phillips, Shell, and Valero, respectively, $and\ \hat{\sigma}^2 = 0.347.\ So\ T_2 = \sum_{i=1}^{5} e^{\hat{\mu}_i + \frac{\hat{\sigma}^2}{2}} = 2{,}240\ kg/yr.$ A simulation was performed here also, yielding a 95% confidence interval of 1,770 kg/yr to 2,840 kg/yr.

Discussion

Estimates using the log-normal yielded somewhat higher totals than arithmetic means. But the two estimates were well within the 95% confidence interval of the other. The uncertainty ranges were similarly wide. There was no appreciable benefit in terms of reduced variability to using a parametric approach. Thus, we use the simpler Estimate 1 for the point estimate 2,194 kg/hr, or 24*365/1000*2171 = 19,000 mt/year, rounded to the nearest hundred, and the 95% confidence interval 1,680 to 2,720 kg/hr, or 15,500 to 22,800 mt/year based on the bootstrap simulation.

APPENDIX 4 – ESTIMATION OF UNCERTAINITY IN SECTOR-WIDE EMISSIONS

For landfills and POTWs, we estimate total emissions, T, as $T_S + T_U$, where T_S and T_U represent the total emissions for sampled and unsampled facilities, respectively. For the uncertainty of T, it is noted that T_S and T_U are not statistically independent because they are both linear functions of the $\bar{y}_i$: $T_S = \sum_{i \in \boldsymbol{S}} a_i \bar{y}_i$, where $a_i = 1$, and $T_U = \sum_{i \in S} b_i \bar{y}_i, \mathit{where}\ b_i = (\sum_{j \in \boldsymbol{U}} x_j)\frac{x_i}{\sum_{j \in \boldsymbol{S}} x_j^2}$ $= constant * x_i$. Using the general formula for the variance of two random variables yields $Var(T) = Var(T_S) + 2*Cov(T_S,T_U) + Var(T_U)$. The general formula for the covariance of two linear combinations of random variables, $U = \sum a_i X_i \ and\ V = \sum b_i X_i,\ is\ Cov\ (U,V) = \sum_i \sum_j a_i b_j Cov(X_i,X_j)$. In the present case, we assume the $\bar{y}_i$are mutually independent and have the same variance, σ^2, say. So $Cov(T_S,T_U) = \sum_{i \in \boldsymbol{S}} a_i b_i \sigma^2$ $= \sigma^2 \sum_{i \in \boldsymbol{S}} a_i b_i$.

Letting $\boldsymbol{a'} = (a_1, a_2, \dots, a_k)$ and $\boldsymbol{b}' = (b_1, b_2, \dots, b_k)$, where k = number of elements in $\boldsymbol{S}$, $Cov (T_S,T_U) = \sigma^2 \boldsymbol{a'b}$. Also $Var(T_S) = Cov(T_S,T_S) = \sigma^2 \boldsymbol{a'a}$, and similarly,. $Var(T_U) = Cov (T_U,T_U) = \sigma^2 \boldsymbol{b'b}$. Thus, the correlation between T_S and T_U is $\rho = \frac{\sigma^2 \boldsymbol{a'b}}{\sqrt{\sigma^2 \boldsymbol{a'a} * \sigma^2 \boldsymbol{b'b}}} = \frac{\boldsymbol{a'b}}{\sqrt{\boldsymbol{a'a} * \boldsymbol{b'b}}}$, which is just the cosine of the angle between **a** and **b**. Since a = the **1** vector, and b = constant * x_i, the correlation is the cosine of the angle between **1** and **x**, where $\boldsymbol{x'} = (x_1, x_2, \dots, x_k)$. Then $Cov(T_S,T_U) = \rho\ Var(T_S)\ Var(T_U)$. Using the general formula for the standard deviation of the sum of two variables, we estimate $\sigma_T = \sqrt{\sigma_{T_S}^2 + 2\rho\sigma_{T_S}\sigma_{T_U} + \sigma_{T_U}^2}$.

APPENDIX 5 – ALTERNATE ANALYSIS OF COMPOSTING SECTOR EMISSIONS USING EMISSION-TO-THROUGHPUT RATIOS (MIDPOINT METHOD)

With observations from only 2 compost facilities, we can only get a rough idea of the total CH_4 emissions from compost facilities and an even rougher estimate of uncertainty. One way or the other, we want to scale up the estimated emissions from these 2 to all of the facilities using throughput as a surrogate. One approach is a regression through the origin. Since only the slope is fit, that leaves 1 degree of freedom to estimate uncertainty.

An alternative, shown here, is to ignore any correlation between these 2 facilities and their corresponding throughput estimates, and scale up based on the ratio of total throughput (T) to the throughput from these 2 facilities ($T_0 = T_1 + T_2$, where T_1 and T_2 are the throughputs from the 2 facilities), and estimating C = total CH_4 emissions from $C_0 = C_1 + C_2$ by $C = T/T_0 * C_0$. Let C_1 and C_2 be the mean of the CH_4 measurements from the 2 facilities. If we consider them as randomly selected from the set of all compost facilities, then they are independent with mean μ = mean emissions from the facilities and variance σ^2.

To estimate uncertainty, we use the usual sample standard deviation, s, where $S^2 = \frac{\sum_1^2 (c_i - \bar{c})^2}{2-1}$. This works out to $S^2 = (c1 - c2)^2/2$, so $s = |c1 - c2|/\mathrm{sqrt}(2)$. $C_0 = C_1 + C_2$, so variance of $C_0 = \sigma^2 + \sigma^2 = 2\sigma^2$, and its standard deviation is $\mathrm{sqrt}(2)*\sigma$. So we estimate the standard deviation of C_0 as $\mathrm{sqrt}(2)*s = |c1 - c2|$. Then the standard error of C is just T/T_0*s.

Results

x_1 = 328 kg/hr = 2.88 Gg CH4/yr

x_2 = 175 kg/hr = 1.54 Gg CH4/yr

T_1 = 533 thousand metric tons/yr

T_2 = 80 thousand metric tons/yr

T_0 = 1546 thousand metric tons/yr

➔ Estimated total CH_4 emissions = C = 1546/(533+80)*(2.88+1.54) = 11.1 Gg CH_4/yr.

➔ Standard error of C = 1546/613*|2.88-1.54| = 3.4 Gg CH_4/yr.

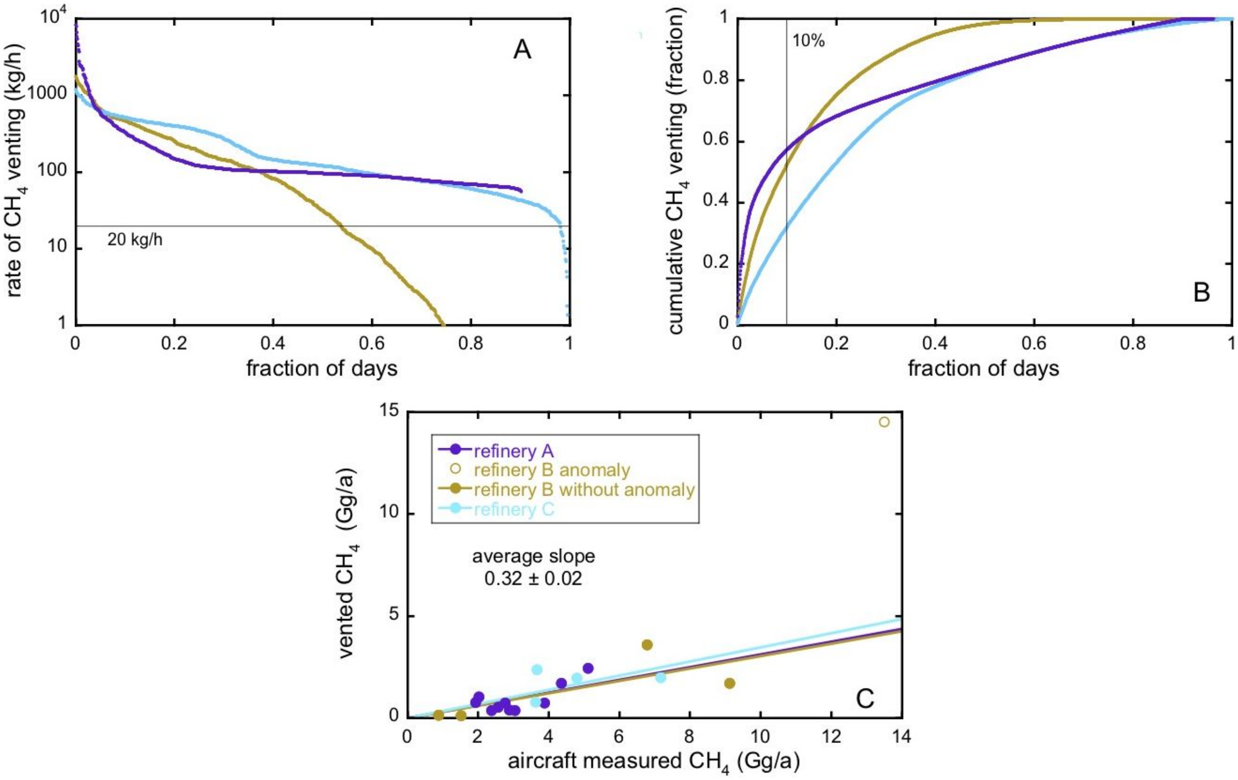

Figure S6 – CH_4 emissions derived from refinery reported H_2 venting data at three SFBA refineries not equipped with Pressure Swing Adsorption technology and with atmospheric vents at H_2 plants. The panels plot the averaged vented daily CH_4 emission rates (Figure S6A) and cumulative fractional total of CH_4 emissions released into the atmosphere (Figure S6B) against the fraction of total days that CH_4 is vented in a ~3-4 year period (2015-18). Figure S6C shows the correlation between aircraft-measured facility-scale CH_4 emissions and the corresponding daily vented CH_4 emission rate derived from H_2 plant data. The total number of days for which H_2 venting data is available is 1387 (Refinery A), 1096 (Refinery B) and 1124 (Refinery C) days.

Table S2: List of airplane-measured facility-scale CH_4 and CO_2 emissions rate over five SFBA refineries between 2015-18. The "Campaign" column indicates the agency (CEC – California Energy Commission, ARB – California Air Resources Board, BAAQMD – Bay Area Air Quality Management District) that funded the corresponding measurement flight.

Facility name	Campaign	Date	Methane Mass Rate	Measurement Uncertainty	Carbon Dioxide Mass Rate	Measurement Uncertainty
			(kg/h)			
Phillips Rodeo	CEC	2/16/2015	271	207	179416	48152
	CEC	5/12/2015	488	179	407069	33287
	CEC	5/15/2015	222	114	262140	53836
	CEC	6/16/2015	527	218	151568	16948
	CEC	5/23/2016	23	10	176504	11204
	BAAQMD	10/10/2016	159	47	299541	73368
	BAAQMD	12/19/2016	157	44	404220	42517
	ARB	10/5/2017	156	83	208256	120456
	BAAQMD	5/9/2018	100	26	257660	35230
	ARB	10/6/2018	439	66	301174	40438
Tesoro Martinez	CEC	5/13/2015	283	94	155080	59413
	CEC	5/15/2015	239	70	240533	16313
	CEC	5/23/2016	258	35	274634	13303
	BAAQMD	10/10/2016	259	34	313391	91139
	BAAQMD	12/20/2016	585	70	467486	55990
	ARB	10/6/2017	546	184	497760	87407
	BAAQMD	4/11/2018	122	49	156154	56323
Valero Benicia	CEC	2/16/2015	700	272	417444	20589
	CEC	5/13/2015	315	76	445250	34918
	CEC	5/15/2015	220	32	218778	28701
	CEC	6/16/2015	231	91	395421	21012
	CEC	5/20/2016	584	78	306688	15534
	CEC	5/23/2016	293	32	213983	7079
	CEC	6/10/2016	329	98	NA	NA
	BAAQMD	10/5/2016	347	56	369182	67994
	BAAQMD	10/10/2016	272	36	308881	32478
	BAAQMD	4/11/2018	497	100	485409	65732
	ARB	10/6/2018	443	66	302806	40508
Shell Martinez	BAAQMD	10/5/2016	100	16	564717	112847
	BAAQMD	11/9/2016	1540	191	777818	100606
	BAAQMD	11/11/2016	173	71	198460	58674
	BAAQMD	4/11/2018	1041	223	342567	57897
	BAAQMD	6/21/2018	774	159	534424	74239
	ARB	10/7/2018	168	18	618500	44940

Facility name	Campaign	Date	Methane Mass Rate	Measurement Uncertainty	Carbon Dioxide Mass Rate	Measurement Uncertainty
			(kg/h)			
Chevron Richmond	BAAQMD	10/10/2016	585	132	428456	147778
	BAAQMD	11/9/2016	787	74	472533	74238
	ARB	10/5/2017	440	250	351743	137663
	BAAQMD	4/21/2018	418	233	527907	213887
	BAAQMD	5/9/2018	414	67	675761	79344
	ARB	10/5/2018	819	200	596532	138009
	ARB	10/6/2018	548	116	429953	59157

Table S3: List of SFBA's landfill facilities with permit-based prior CH_4 inventory emission estimates. Facilities sampled using airplane approach are highlighted in orange.

Facility	City	Methane emissions (metric tons)		
		2016	2017	2018
Waste Management of Alameda County (Altamont)	Livermore	8768	8340	7957
Keller Canyon Landfill Company	Pittsburg	6426	3936	3936
International Disposal Corp of CA (Newby Island)	Milpitas	6075	7725	8442
Browning-Ferris Industries of CA, Inc (Ox Mountain)	Half Moon Bay	5651	6012	6012
Redwood Landfill Inc	Novato	3899	4201	4304
Guadalupe Rubbish Disposal	San Jose	3279	3010	2231
Kirby Canyon Recycling and Disposal Facility	San Jose	3062	3332	4164
Republic Services Vasco Road, LLC	Livermore	2703	2678	2845
Potrero Hills Landfill, Inc	Suisun City	2472	4201	4033
Republic Services of Sonoma County, Inc	Petaluma	2363	4596	4710
Waste Management of Alameda County	Fremont	2215	2236	2168
West Contra Costa County Landfill	Richmond	2146	2146	2146
Cypress Amloc Land Co , Inc	Colma	998	790	695
City of Mountain View (Shoreline Landfill)	Mountain View	992	1129	1164
Waste Management Inc	San Leandro	646	603	615
Acme Fill Corporation	Martinez	547	547	483
Recology Pacheco Pass	Gilroy	544	484	475
Clover Flat Resource & Recovery Park	Calistoga	415	415	446
Napa-Vallejo Waste Management Authority	Napa	406	383	413
City of Santa Clara	Santa Clara	398	350	12
Zanker Road Resource Management,Ltd	San Jose	361	361	361
TRC	Antioch	314	296	291
Turk Island Solid Waste Disposal Site	Union City	305	252	229
Cal-Pox, Inc	San Rafael	234	234	234
Sunquest Properties Inc	Brisbane	223	223	223
City of Berkeley/Engr Div/Public Works	Berkeley	166	196	23
Zanker Road Material Processing Facility	San Jose	166	168	168
Shoreline Amphitheatre	Mountain View	130	130	93
City of Alameda, Maint Serv Center	Alameda	110	110	0
City of San Jose (Singleton Road Landfill)	San Jose	103	83	66
Bayview Business Park Owner's Association	San Rafael	66	66	66
Pleasanton Garbage Service, Inc	Pleasanton	50	55	18
Spirit HD Colma CA, LP	Colma	37	21	13
City of Palo Alto Landfill	Palo Alto	19	19	20
City of Burlingame, Waste Water Treatment Plant	Burlingame	9	9	7
County of Santa Clara	San Jose	7	14	36
City of Menlo Park	Menlo Park	4	4	4
City of Sunnyvale/Public Works Dept	Sunnyvale	4	4	4
	Total	**56313**	**59359**	**59107**

Table S4: Summary of airplane-measured facility-scale emissions rate over ten SFBA landfills from year 2015-19. The "Campaign" column indicates the agency (ARB – California Air Resources Board, CalRecycle – California Department of Resource Recycling and Recovery, BAAQMD – Bay Area Air Quality Management District) that funded the corresponding measurement flight.

Facility	Campaign	Date	Methane Mass Rate	Measurement Uncertainty	BAAQMD Inventory	EPA GHGRP
			(kg/h)			
Altamont	BAAQMD	11/09/2016	2132	424	1001	812
	BAAQMD	12/19/2016	1983	295	1001	812
	ARB	10/06/2017	2977	653	952	778
	ARB & CalRecycle	12/07/2017	1358	547	952	778
	BAAQMD	05/11/2018	2010	267	908	778
	ARB & CalRecycle	08/27/2018	2077	240	908	778
Vasco	BAAQMD	12/20/2016	536	121	309	144
	BAAQMD	05/11/2018	807	157	325	160
Keller Canyon	BAAQMD	10/12/2016	931	211	734	700
	ARB	10/06/2017	640	209	449	605
	BAAQMD	03/26/2018	1339	277	449	605
	BAAQMD	3/28/2018	1130	355	449	605
	BAAQMD	6/21/2018	396	145	449	605
	BAAQMD	8/23/2018	1127	171	449	605
	ARB	10/5/2018	1361	209	449	605
	BAAQMD	6/27/2019	1115	156	449	605
Newby Island	BAAQMD	11/11/2016	2893	1173	693	640
	ARB	10/05/2017	2075	587	882	769
	BAAQMD	04/20/2018	2214	402	964	769
	BAAQMD	08/25/2018	2442	387	964	769
	ARB	10/05/2018	3287	487	964	769
Potrero Hills	ARB	10/06/2017	2292	385	480	394
	ARB & CalRecycle	12/07/2017	2004	417	480	394
	BAAQMD	04/20/2018	1799	532	460	394
	ARB & CalRecycle	08/23/2018	1718	252	460	394
	ARB	10/05/2018	919	286	460	394
Redwood	BAAQMD	12/20/2016	660	166	445	507
	ARB & CalRecycle	11/17/2017	140	42	480	618
	BAAQMD	04/11/2018	505	102	491	618
	BAAQMD	05/09/2018	422	52	491	618

Sonoma County	BAAQMD	07/17/2018	796	122	538	314
Kirby Canyon	BAAQMD	07/18/2018	383	166	475	439
Guadalupe	BAAQMD	07/18/2018	187	63	255	468
Half Moon Bay (Ox Mountain)	ARB	10/06/2018	2939	574	686	533

Table S5: List of SFBA's publicly owned treatment works (POTWs) with their annual throughput of waste effluent processed. Facilities sampled by the airplane for this study are highlighted in orange.

Facility	City	Effluent Throughput		
		2016	2017	2018
		million liters of wastewater		
San Jose-Santa Clara Regional Wastewater Facility	San Jose	127777	141361	146827
San Francisco South East Treatment Plant	San Francisco	94673	94673	96649
East Bay Municipal Utility District	Oakland	82798	91505	91505
Central Contra Costa Sanitary District	Martinez	48885	48885	60790
Union Sanitary District	Union City	30866	32237	33679
City of Santa Rosa Wastewater Treatment	Santa Rosa	23996	23996	29189
San Francisco, City & County, PUC	San Francisco	20082	22818	24420
Palo Alto Regional Water Quality Control Plant	Palo Alto	25987	28440	23988
Silicon Valley Clean Water	Redwood City	20646	23106	19169
Fairfield-Suisun Sewer District	Fairfield	17863	22451	18272
City of Sunnyvale Water Pollution Control	Sunnyvale	16497	18060	18060
Delta Diablo Sanitation District	Antioch	17114	18321	17455
Hayward Waste Water Treatment Plant	Hayward	14497	14407	17099
Oro Loma Sanitary District	San Lorenzo	17231	20754	16913
Vallejo Sanitation & Flood Control District	Vallejo	13336	16736	16814
San Mateo Water Quality Control Plant	San Mateo	15348	18008	15307
Dublin San Ramon Services District - Wastewater TP	Pleasanton	14388	14388	14617
Central Marin Sanitation Agency	San Rafael	11430	19800	14126
Napa Sanitation District - Soscol	Napa	11534	13953	13953
South San Francisco-San Bruno Water Quality Plant	South San Francisco	11659	11848	11848
West County Wastewater District	Richmond	10573	13805	11318
City of Livermore Sewage Treatment Plant	Livermore	7640	8070	8070
City of Richmond Water Pollution Control District	Richmond	6905	10251	8014
North San Mateo County Sanitation Dist	Daly City	72453	6560	7858
Novato Sanitary District	Novato	5290	6534	7813
City of Petaluma, Dept of Water Resources & Convs	Petaluma	7283	8521	7037

San Leandro Water Pollution Control Plant	San Leandro	6449	7173	6727
Sonoma County Water Agency	Sonoma	3865	5194	5663
City of Brentwood	Brentwood	4690	4980	4820
Sewerage Agency of South Marin	Mill Valley	4141	4296	4296
City of Burlingame, Waste Water Treatment Plant	Burlingame	4153	4823	3734
City of Pacifica Calera Creek Water Recycling	Pacifica	3372	3676	3459
Pinole-Hercules Wastewater Treatment Plant	Pinole	3431	4079	3426
North Point Wet Weather Facility, SFPUC	San Francisco	4318	6592	3409
Las Gallinas Valley Sanitary District	San Rafael	3361	4517	3305
City of Benicia	Benicia	409	3355	2724
Town of Windsor	Windsor	2261	2562	2628
City of Millbrae Wastewater Treatment Plant	Millbrae	2227	2486	2486
City of American Canyon	American Canyon	2230	1904	1904
Town of Discovery Bay	Discovery Bay	1031	1605	1817
Mt View Sanitary District	Martinez	1707	2041	1813
Sausalito-Marin City Sanitary District	Sausalito	1803	2246	1595
Marin County Sanitary Distr No 5	Tiburon	958	1039	1039
City of Calistoga	Calistoga	643	897	1034
San Francisco International Airport	San Francisco	852	852	943
Rodeo Sanitary District	Rodeo	741	999	818
Town of Yountville	Yountville	649	667	667
City of St Helena	Saint Helena	647	717	569
Treasure Island - US Navy BRAC PMO-W	San Francisco	382	428	496
Pacific Union College	Angwin	na	na	142
USCG Training Center	Petaluma	124	131	100
Adventist Health St Helena	Saint Helena	na	na	46
U S Veterans Administration Medical Center	Livermore	14324	9	9
Discovery Bay Community Services District	Discovery Bay	38	na	na
	Total	**815555**	**816759**	**810461**

Table S6: Summary of airplane-measured facility-scale CH_4 emissions rate over four SFBA wastewater treatment plants (POTWs) from 2016-18. The "campaign" column indicates the agency (ARB – California Air Resources Board, BAAQMD – Bay Area Air Quality Management District) that funded the corresponding measurement flight.

Facility	Campaign	Date	Methane Mass Rate	Measurement Uncertainty
			kg/h	
San Jose Santa Clara	BAAQMD	04/19/2018	204	54
	BAAQMD	04/20/2018	290	69
	ARB	10/05/2018	427	80
EBMUD Oakland	BAAQMD	11/11/2016	82	314
	BAAQMD	12/19/2016	91	26
	BAAQMD	04/19/2018	130	24
	BAAQMD	05/10/2018	83	21
San Francisco Southeast	BAAQMD	12/20/2016	229	56
	BAAQMD	04/19/2018	62	10
	BAAQMD	05/10/2018	80	16
Central Contra Costa	BAAQMD	04/20/2018	120	22

Table S7: List of SFBA's composting operations with their annual throughput of green and food waste accepted (or maximum permitted). Facilities sampled by the airplane for this study are highlighted in orange.

Facility	City	2018 throughput (thousand metric tons)
Z-Best Composting Facility	Gilroy	533
Newby Island Compost Facility	San Jose	146
Composting Facility (Altamont Landfill)	Livermore	140
Redwood Landfill	Novato	128
WM Earthcare of Marin	Novato	121
Zanker Road Class III Landfill	San Jose	87
WCCSLF Organic Materials Processing	Richmond	80
South Valley Organic Composting Facility (Recology Pacheco Pass)	Gilroy	37
City of Napa Material Diversion Facility	Napa	35
East Bay Municipal Utility District	Oakland	33
Monterey Mushrooms - Morgan Hill	Morgan Hill	30
Potrero Hills Compost Facility	Suisun City	18
West Marin Compost	Nicasio	18

Grab N` Grow (Soiland Co.)	Santa Rosa	15
Upper Valley Disposal Service (Napa Recycling and Waste Services)	Saint Helena	14
Vision Recycling Green Waste Composting	Livermore	13
Dolcini Brothers Composting Operation Ag	Petaluma	13
Goodyear Road Compost Facility	Benicia	10
Laguna Subregional Compost Facility	Santa Rosa	10
Carneros River Ranch	Petaluma	8
West Marin Compost Project- Drop Off	Nicasio	7
Blue Line Transfer	South SF	6
Oliveira Enterprises, Inc.	Byron	5
PSSI Ag. Material Storage / Handling Op.	Stanford	4
Poncia Fertilizer	Santa Rosa	3
Altamont Resource + Recovery Facility	Livermore	3
Countryside Mushrooms, Inc.	Gilroy	3
Upper Valley Research Composting Op. #2	Saint Helena	3
B and D Mushrooms, Inc.	San Martin	3
Point Reyes Compost Co. LLC	Point Reyes Station	2
Reichert Duck Farm	Petaluma	2
Royal Oaks Mushrooms	Morgan Hill	2
Bolinas-Stinson Resource Rcvry. Project	Bolinas	2
South Valley Organic Composting Facility	Gilroy	2
Global Mushrooms Farm	Gilroy	2
South Valley Mushroom Farm	Morgan Hill	2
Buchli Station	Napa	2
Oso Vineyard	Pope Valley	1
Clover Flat Resource Recovery Park	Calistoga	1
Joseph Phelps Vineyards	Saint Helena	1
Yount Mill Composting	Oakville	0
Mont Emei Vineyards, LLC	Napa	0
Opus One	Oakville	0
Tierra Vegetables	Santa Rosa	0
Graton Waste Water Treatment Plant	Graton	0
Thermopile Research Composting Operation	Nicasio	0
Central Compost Site2	Petaluma	0
Zero Waste to Energy Development Co. AD	San Jose	0

	Total	1546

Table S8: Summary of airplane-measured facility-scale CH_4 emissions rate over two large SFBA composting operations.

Facility	Date	Methane Mass Rate	Measurement Uncertainty
		kg/h	
ZBEST Composting	05/10/2018	374	75
	07/18/2018	251	46
	08/25/2018	360	31
West Contra Costa Composting	05/11/2018	175	37

Table S9: Summary of airplane-measured facility-scale CH_4 emissions rate over two SFBA dairy facilities.

Facility	Date	Methane Mass Rate	Measurement Uncertainty
		kg/h	
Tresch dairy farm	10/12/2016	10	4
McClelland dairy	10/12/2016	38	7