Anthropogenic Impacts on the Atmosphere

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ABSTRACT

This study derives methane emission rates from 92 airborne observations collected over 23 facilities including 5 refineries, 10 landfills, 4 wastewater treatment plants (POTWs), 2 composting operations and 2 dairies in the San Francisco Bay Area. Emission rates are measured using an airborne mass balance technique from a low-flying aircraft. Annual measurement-based sector-wide methane emissions are 19,000 ±2,300 Mg for refineries, 136,700 ±25,900 Mg for landfills, 11,900 ±1,500 Mg for POTWs, and 11,100 ±3,400 Mg for composting. The average of measured emissions for each refinery ranges from 4 to 23 times larger than corresponding emissions reported to regulatory agencies, while measurement-derived landfill and POTW estimates are approximately twice the current inventory estimates. Significant methane emissions at composting facilities indicate that a California mandate to divert organics from landfills to composting may not be an effective measure for mitigating methane emissions unless best management practices are instituted at composting facilities. Complementary evidence from airborne remote sensing imagery indicates atmospheric venting from refinery hydrogen plants, landfill working surfaces, composting stockpiles, etc., to be among the specific source types responsible for the observed discrepancies. This work highlights the value of multiple measurement approaches to accurately estimate facility-scale methane emissions and perform source attribution at sub-facility scales to guide and verify effective mitigation policy and action.

TOC ART

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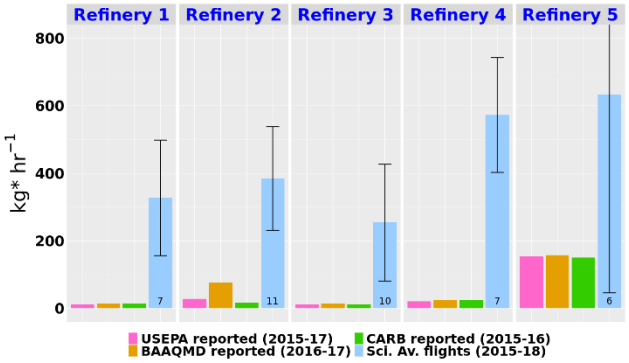
Refinery 1
Refinery 2
Refinery 3
Refinery 4
Refinery 5
800
600
400
200
0
kg* hr−1
7
11
10
7
6
USEPA reported (2015-17)
CARB reported (2015-16)
BAAQMD reported (2016-17)
Sci. Av. flights (2015-18)

INTRODUCTION

Methane (CH_4) is a potent greenhouse gas (GHG) and a short-lived climate pollutant, which is 86 times more effective than carbon dioxide (CO_2) in trapping infrared radiation over a 20-year time frame.[1] CH_4 accounts for approximately 20% of the globally observed GHG-related anthropogenic radiative forcing since pre-industrial times.[1] Due to its short lifetime (~10 years) as compared to that of CO_2 (~100 years),[2] expedited CH_4 emissions reduction efforts can provide near-term climate benefits through reduction in radiative forcing.

In California, which has the country's largest dairy industry, the California Air Resources Board (CARB) estimates that the majority of the statewide methane emissions are from enteric fermentation (28%) and manure management (25%), followed by landfills (21%).[3] Oil and gas (O&G) production/extraction systems account for 16% of statewide CH_4 emissions.[4] In the San Francisco Bay Area (SFBA), landfill emissions account for more than 50% of the bottom-up CH_4 emissions inventory.[5] Although natural gas (NG) transmission and distribution remains a major source in the SFBA (15%), large downstream NG consumers such as refineries account for only ~2% of SFBA CH_4 emissions, per a spatially resolved inventory[6] and industry-reported estimates.[7,8]

Starting in 2005, California adopted a series of regulations aimed at reducing GHG emissions: in 2005, the Governor's Executive Order (EO) S-3-05 to reduce GHG emissions 80% below 1990 levels by year 2050; in 2006, Assembly Bill (AB) 32 to authorize CARB to develop regulations and a cap-and-trade program to reduce statewide GHG emissions to 1990 levels by year 2020; in 2015, EO B-30-15 to reduce GHG emissions 40% below 1990 levels by year 2030, Senate Bill (SB) 32 to codify the goal set by EO B-30-15, and AB 1496 to require monitoring

and measurement of CH_4 hotspots including GHG lifecycle analysis of statewide NG systems; and, in 2016, SB 1383 to reduce CH_4 emissions 40% below 2013 levels by year 2030.

The Bay Area Air Quality Management District (BAAQMD) is the nine-county SFBA's regional air quality regulatory agency. In 2017, BAAQMD's Board adopted the 2017 Clean Air Plan (CAP),[9] a multi-pollutant emissions control strategy that aims to reduce SFBA's GHG emissions to levels consistent with California's targets. Successful implementation of the CAP and the agency's rule development efforts depend on an accurate regional bottom-up emissions inventory of GHGs.

Emissions inventories at regional, state, and federal levels are typically generated using simple combinations of emission factors and activity data, and the evaluation of inventory emissions estimates requires comparison against top-down measurement-based estimates. At the state level, several recent top-down emissions assessment studies have used regional inverse modeling driven by tower and/or satellite column observations to determine that the Central Valley of California is a major source of GHG emissions and strongly assert that bottom-up inventories underestimate CH_4 emissions in this agriculture- and industry-intensive region.[10–15] Evaluations of CH_4 inventories in the Los Angeles basin consistently indicate discrepancies including underestimation of CH_4 emissions[14,16–19] albeit with a decreasing trend[20] and misallocation of emissions within sectors[21]. Recent top-down emissions assessment studies in the SFBA indicate significant underestimation of methane emissions in bottom-up inventories but do not categorically identify which sectors are responsible. Fairley and Fischer[22] scaled local enhancements of CH_4 and carbon monoxide (CO) from 14 BAAQMD measurement sites and combined those with SFBA CO emissions estimates to derive a regional mean CH_4 emissions estimate of 240 ± 60 $GgCH_4\ yr^{-1}$ (2009-2012 period; 1 Gg = 1000 metric tons). Jeong et al.[23] applied a hierarchical Bayesian

inversion approach to CH_4 observations from six tower sites to estimate that SFBA CH_4 emissions range from 166-289 $GgCH_4\ yr^{-1}$ (for year 2015) which is 1.3-2.3 times higher than BAAQMD's recent CH_4 inventory.[6] Jeong et al.[23] also used a multi-species hierarchical Bayesian method to conduct source apportionment analysis combining concurrently measured volatile organic compound (VOC) tracers with CH_4 measurements. They attributed 82% of the CH_4 emissions in SFBA to biological sources (with landfills being the largest source of underestimation) and the remaining fraction to fossil-fuel sources. Taken together, regional and statewide discrepancies between top-down and bottom-up inventory CH_4 estimates suggest inventories undercount CH_4 emissions by approximately a factor of 1.5-2.

These discrepancies, combined with the need for representative emissions data to implement BAAQMD's CAP[9] have focused research efforts on identifying sources of the discrepancy at the sector and facility levels. Aircraft-based mass-balance measurements offer a proven technique to assess emissions inventories. Recently, Lavoie et al.[24] have reported airborne CH_4 emission rates from six facilities in central and midwestern U.S. that are larger than facility-reported estimates by factors of 21-120 and 11-90 for two major end users of NG, power plants and refineries, respectively. Conley et al.[25] developed a new airborne method to quantify trace gas emissions, within 20% accuracy, from facilities in urban areas that often have multiple, closely-spaced potential sources. This approach (described in Appendix 1 in Supplementary Information) was first applied to estimate CH_4 emissions from a subset of California's NG infrastructure by Mehrotra et al.[26] including measurements over 3 SFBA refineries, and by Thorpe et al. [27] for California's underground gas storage (UGS) facilities. In particular, the authors reported an order of magnitude larger emissions from SFBA refineries and over 5 times more than that reported to the USEPA and CARB at UGS facilities.

In this study, we build on the Mehrotra et al. study,[26] adding three more years of facility-level airborne measurements of CH_4 emissions focused on SFBA with a broad sampling of known CH_4 emitters such as landfills and wastewater treatment plants, as well as relatively under-sampled sources such as refineries and previously unsampled sources including composting. Regional sector-specific CH_4 emissions inventories are derived using a combination of methods that include direct airborne measurements (refineries), linear regression (landfills and publicly owned wastewater treatment plants, POTWs), and measured emissions-to-throughput ratios (composting and dairies). Comparisons are provided between measurement-based estimates and reported emissions inventories. Probable sources of "missing" methane are identified for multiple sectors using extensive remote sensing plume imagery generated over California by National Aeronautics and Space Administration Jet Propulsion Laboratory's (NASA-JPL's) Airborne Visible / Infrared Imaging Spectrometer – Next Generation (AVIRIS-NG) instrument and cross-comparisons to other activity data sets.

ANALYSIS AND RESULTS

Measurement Sites

Airborne measurements, using an instrumented Mooney aircraft owned and operated by Scientific Aviation (Boulder, CO), were obtained at 23 facilities (92 flight observations) in the SFBA between 2015 and 2019. An example cylindrical flight path over a facility with color-coded methane concentrations is shown in Figure S1. The sampled sites included refineries (5), landfills (10), wastewater treatment plants (4), composting operations (2) and dairies (2), as summarized in the Supplementary Information. Many sites were sampled multiple times. The aircraft typically spent 20-60 minutes sampling each site and most sampling occurred between 10 am and 5 pm local time. The airborne mass flux estimation approach and the uncertainties

associated with this method are well-documented[25,26,28] and also summarized in the Supplementary Information (Appendix 1).

Sector-Specific Emissions Estimation

Airplane emission rate measurements from each facility were used to estimate SFBA CH_4 emissions for the sampled sectors: refinery, landfill, POTW, composting and dairy. Each airborne measurement took place in a well-mixed planetary boundary layer during daytime within a range of wind speeds with steady wind conditions. The flight measurements represent a "snapshot" in time. Although we did not observe a seasonal trend in emissions data, day to day variations at refineries were statistically significant (see Appendix 2 in Supplementary Information) and indicate that there are process-level activity variations that result in varying emissions (see 'Source Attribution and Discussion' section). We estimated annual emissions at each facility from multiple measurements over a four-year period. Main assumptions for the estimates include that CH_4 emissions across diurnal cycles (daytime versus nighttime) do not vary a lot and that the multi-year record of individual observations capture a representative sampling of the variability in the emissions across days, weeks and months. These assumptions allowed us to generate measurement-based annualized emissions from a limited number of samples. Our subsequent analysis shows a consistent and definite trend in the emissions inventories, and this leads to our major conclusions.

The airborne measurement approach presented in this study is focused on estimation of CH_4 emissions from point-source facilities. This approach is not suitable for a regional evaluation of the ensemble CH_4 emissions from the urban core, a technique that is typically required to estimate diffused areawide emissions from the urban NG distribution system. There is increasing observation-based evidence of underestimation of CH_4 emissions in inventories of

urban NG systems, especially in cities in the East Coast of the U.S. with older infrastructure[29,30]. In this study, we do not provide a measurement-based update to the regional NG distribution system inventory but acknowledge that, based on the outcomes of the above-mentioned studies, the inventory estimates for SFBA are likely underrepresented.

Different statistical approaches were used for different sectors. Since all five SFBA refineries were sampled, estimates were based on airplane measurements only. For the landfill and POTW sectors, regressions were fit to estimate the emissions from the non-sampled facilities and thereby scaled up to the entire sector emissions estimate. For landfills, facility-specific prior bottom-up inventory estimates (Table S3) were used as a covariate in regressions to estimate the emissions from facilities not sampled. For sectors without facility-specific prior inventory CH_4 emissions estimates, throughput or activity data were used as covariates: for POTWs, throughput of effluent processed (Table S5) was used. For composting and dairy sectors, for which we only have two facilities sampled each, we used the ratio of the averaged measured emissions to throughput (organic waste processed and heads of cows, for composting (Table S7) and dairies, respectively) to scale up to the total sector emissions.

Uncertainties were evaluated to investigate true variability versus measurement error. Appendix 2 in the Supplementary Information demonstrates that there was significant day to day variations in the refinery emissions data above measurement uncertainty. The individual uncertainties of airplane measurements varied as well. To test how incorporating these uncertainties would affect the emissions estimates, an alternative analysis was run on a dataset with all the refinery observations (41 data points) using a Gaussian model on the natural logs of the measurements and uncertainties (see Supplementary Information - Appendix 3). Because the results were similar to the unweighted approach, the latter was used for its simplicity. A similar

comparison of methods was also conducted for the landfill measurements (not shown) and indicated no difference in the total emissions resulting from the log-normal versus the arithmetic means approach. Table 1 summarizes the measurement derived CH_4 emissions totals, summing contributions from sampled and non-sampled facilities for source sectors with airborne measurements, including estimates of uncertainties.

Table 1. Measurement-based sector-specific methane emission estimates compared to BAAQMD's annual prior bottom-up inventory estimates (1 Gg = 1000 metric tons).

Source Category		BAAQMD Prior Estimate (Gg)	Airplane Mass Balance	
			Estimate (Gg)	Standard Error (Gg)
Refineries	Total	1.9	19.0	2.3
Landfills	Sampled	47	110.0	8.9
	Non-Sampled*	11	26.7	17.4
	Total	58	136.7	25.9
Wastewater Treatment Plants (POTWs)	Sampled	NA	5.7	1.0
	Non-Sampled*	NA	6.3	0.8
	Total	5	11.9	1.5
Composting Operations	Sampled	NA	4.4	0.9
	Non-Sampled*	NA	6.7	2.0
	Total	NA	11.1	3.4
Dairy Operations	Sampled	NA	0.4	0.2
	Non-Sampled*	NA	4.6	2.6
	Total	9.2	5.0	2.9

*Non-sampled estimates and their uncertainties are derived from the regression analysis and midpoint method described in the sector-specific sections and Appendices 4 and 5, respectively.

Refineries

A total of 41 measurements were made over the 5 refineries from 2015 through 2018. Table S2 provides a summary of the emission rate measurements (in kg h^{-1}) and Figure 1A shows a corresponding scatter plot of observed CH_4 emissions rates by refinery. Individual observations, y_{ij} (i=refinery, j=observation within refinery = 1,…,n_i) are converted to an annual estimate in Gg by refinery and averaged, yielding $\bar{y}_i$. Total emissions are computed as $T = \bar{y}_1 + \bar{y}_2 + \cdots + \bar{y}_5$, while the variance of the total emissions is estimated as $s_T^2 = s^2 \sum_{i=1}^{5} \frac{1}{n_i}$, where

$s^2 = \frac{\sum_{i=1}^{5}\sum_{j=1}^{n_i}(y_{ij}-\bar{y}_i)^2}{41-5}$. Based on the results of the airborne measurements, estimated annual emissions from the refinery sector is 19.0 ±2.3 Gg of CH_4 (Table 1). Figures 1a and 2a show the range in the ratios of measured to inventory (multi-year averaged) CH_4 emissions, emphasizing that the inventory for individual refineries is underestimating averaged emissions 4- to 23-fold.

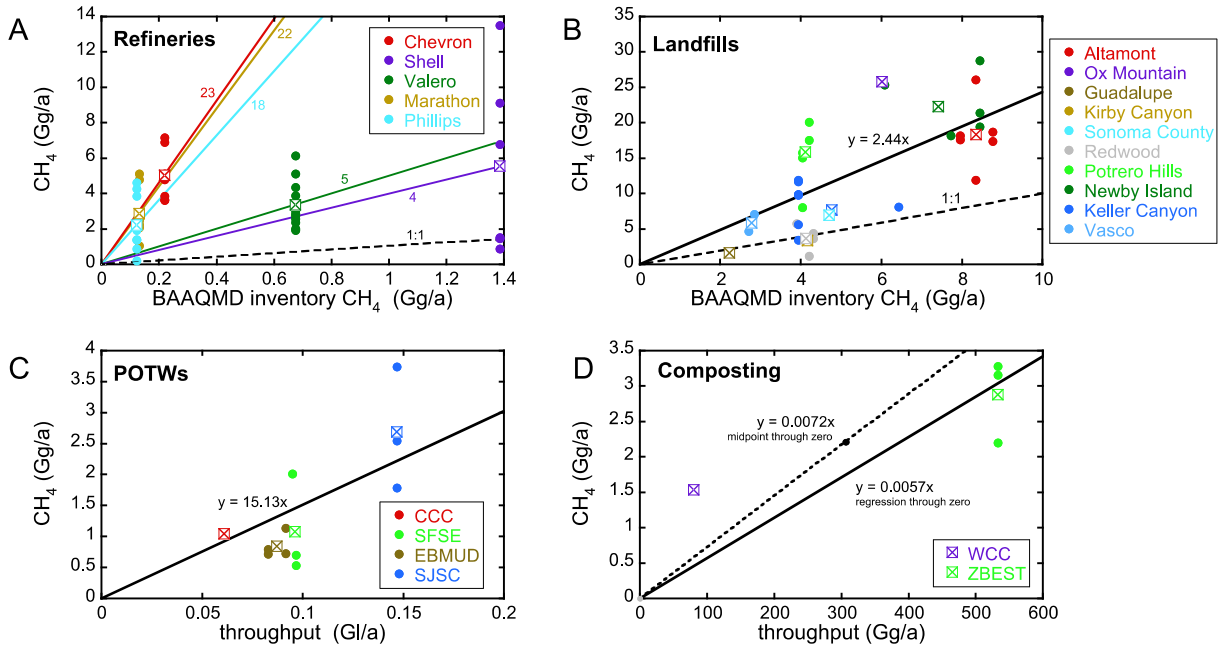

Figure 1. Annual CH_4 emissions inventories from A) refineries), B) landfills, C) wastewater treatment plants (POTWs), and D) composting operations (shown in Gg CH_4) measured by the aerial mass balance method versus prior inventory estimates (A-B) or throughputs (C-D). 1 Gg = 1000 metric tons; 1 Gl = 10^9 liters; throughput = effluent or waste feedstock processed. Filled circles indicate individual aerial mass balance measurements, while crossed squares indicate the averages for each facility. The solid line is the linear regression best fit for the facility averages, forced through zero. The colored lines in A) show the range in ratios of measured to inventory CH_4 for the different refineries. The dashed line in A) and B) is the 1:1 line, shown for refineries and landfills to emphasize the underestimation in the prior emissions inventory. The dotted line for composting is the line between the midpoint of the two facilities and zero.

Landfills

BAAQMD's 2016-18 GHG Emissions Inventory (EI) identifies 38 landfills in the SFBA (Table S3). Figure 1B shows the comparison of airborne landfill CH_4 methane emission estimates versus the prior inventory estimates. Table S4 shows the airborne mass-balance

emissions rates from 34 measurements over 10 active landfills containing approximately 80% of emissions estimated by BAAQMD (Table S3) along with CH_4 emissions self-reported to US EPA by facilities. The sum of the landfill airplane-estimated methane is $T_S = \sum_{i \in \boldsymbol{S}} \bar{y}_i$, where $\bar{y}_i$ is the mean airplane-estimated methane from landfill i and **S** = set of sampled landfills. A regression of facility averages of airborne measurements against inventory estimates is used to estimate the annual emissions from non-sampled landfills. Thereby, a simple linear regression is done with the 10 landfill means, $\bar{y}_i$, as the dependent variable and x_i = prior estimate as the independent variable. The intercept is non-significant (-5.5 with standard error, s.e. = 5.9) so a regression line through the origin is fit. The slope is 2.4 (s.e. = 0.36), so we estimate $T_U = \sum_{i \in \boldsymbol{U}} 2.4 x_i = 2.4 \sum_{i \in \boldsymbol{U}} x_i$, where **U** = set of unsampled landfills. The uncertainty is the slope uncertainty times the sum of the x_i: $standard\ error\ of\ T_U = 0.36 \sum_{i \in \boldsymbol{U}} x_i$. The total emissions combining the estimates from the sampled and non-sampled landfills are: $T = T_S + T_U$. A derivation of the variance of T is presented in Appendix 4 in the Supplementary Information. This approach yields a total (sampled + unsampled) annual estimate of 136.7 ±25.9 Gg of CH_4 for the landfill sector (Table 1).

Publicly Owned Treatment Works (POTWs)

Airplane measurements were made over 4 of SFBA's ~50 publicly owned treatment works (POTWs). The sampled facilities are amongst the largest POTWs in the region by throughput of sewage effluent processed (Table S5), accounting for 46% of the total effluent throughput. Table S6 summarizes the measured CH_4 emission rates from 11 individual measurements over 4 POTWs while Figure 1C shows a comparison of observed CH_4 emission rates with effluent throughput for the 4 POTWs. The BAAQMD prior bottom-up CH_4 emissions inventory for the POTW sector is not sufficiently quantified at the facility level to include all

possible sources. Since bottom-up emissions inventories for POTWs are based on scaling of emission factors with sewage throughput activity data, the overall facility CH_4 emissions are assumed to scale linearly with throughput. For POTWs, we follow the same estimation procedure as for landfills but use facility-specific throughput information as a proxy for prior CH_4 emissions and as the independent variable in the regression analysis. The regression curve is forced through the origin since an assumption is made that in the absence of any activity (wastewater effluent processed), there will be no CH_4 emissions. The measurement-based approach yields a total annual estimate of 11.9 ±1.5 Gg of CH_4 for POTWs (Table 1).

Composting Facilities

There are no prior BAAQMD EI CH_4 estimates for the composting sector (similar to POTWs), a sector where it is generally assumed that an aerobic pathway converts carbon in organic matter to CO_2. Four airborne flux measurements were made over two composting facilities whose combined throughput (Table S7) was approximately 40% of the SFBA total (including those permitted but not operating at full capacity). The measurements indicate significant CH_4 emissions at composting facilities suggesting anaerobic pathways are likely dominant. Table S8 summarizes the measured CH_4 emission rates for these two facilities. Figure 1D shows a comparison of the measurement-derived CH_4 annual emissions estimates versus the annual throughput of waste feedstock processed. Since airborne measurements were made for only two composting facilities, we used two methods for estimating total emissions: 1) a regression analysis similar to that applied to POTWs with organic waste throughput, rather than sewage throughput, assumed as the independent variable that scales linearly with CH_4 emissions; and, 2) a ratio method that uses the midpoint of the average measurements of the two facilities to their throughputs (described in Appendix 5 of the Supplementary Information). An assumption

is made similar to that made for POTWs that in the absence of any activity (composting feedstock processed), there will be no CH_4 emissions, and therefore the regression curve is forced through the origin. The composting sector total emissions are 9.7 ±4.1 Gg of CH_4 using the regression method and 11.1 ±3.4 Gg using the midpoint method. Table 1 shows the values for the midpoint method, since it is more appropriate given the small sample size for analysis. The average value of 11.1 Gg for the sector is similar in magnitude to total emissions from the POTW sector, compared to negligible as previously assumed.

Dairy Operations

Aggregate regional CH_4 EI estimates for the dairy livestock sector are generated by combining information on the total number of milk-yielding cows (heads) from US Department of Agriculture with an emission factor generated from state-specific data.[6] One airborne CH_4 measurement was conducted over each of the two largest dairies in the SFBA with ~900 and ~1000 milk cows (Table S9), accounting for about 9% of SFBA's dairy livestock.[6] Dairy facilities have no emissions or activity data reporting requirements to BAAQMD. Hence, we derive sector-wide CH_4 emissions from the regression analysis combining publicly available cow head data (from company websites, etc.) for these sampled facilities with emission factors from inventory reports[6] as input parameters. Since airborne measurements were made over only two SFBA dairies, we use the same two-method approach as that used for composting to estimate total emissions. An assumption is made similar to that for other sectors that in the absence of any activity (cows in the dairy), there will be no CH_4 emissions, and therefore the regression line is forced through the origin. The two methods result in the same annual emissions, 5.0 Gg of CH_4, although the uncertainties are different: ±1.9 Gg for the regression method, and ±2.9 Gg for the midpoint method. We report the results of the midpoint method in Tables 1 and 2. Although

there is substantial uncertainty, the sector-wide emissions estimate is approximately a factor of 2 lower than that previously inventoried (Table 1).

SOURCE ATTRIBUTION AND DISCUSSION

Refineries

Figure 2A shows a comparison of measured mean hourly CH_4 emission rates for each SFBA refinery (blue bars) to emission rates reported to and derived from three different annual inventory sources: i) emissions reported to BAAQMD, averaged for years 2016 and 2017 (orange bars); ii) CARB's Mandatory Reporting of GHG emissions, averaged for 2015 and 2016 (green); and iii) GHG emissions required under US EPA's 40 CFR Part 98 (pink). Note that at Phillips 66 and Marathon (previously Andeavor and Tesoro) refineries, the flight footprint includes independently permitted GHG sources, an electricity co-generation plant and/or a hydrogen production plant. These sub-facilities are minor CH_4 sources (<5%) in reported inventories (although major sources of CO_2). The CH_4 emissions from these sub-facilities are included in the refinery totals (for all three reported inventories) to ensure a direct comparison with emissions obtained from airplane measurements. Measured average CH_4 emission rates in Figure 2A range from 4-23 times larger than those from reported estimates, with the lower bound of the observed range significantly higher than inventory estimates for all refineries (except Shell whose observed emissions have a larger spread). Thus, there is a large quantity of missing methane not captured in the bottom-up inventories, but which is consistently observed in the measurements conducted at refineries. The measurement-based assessment results in a ten-fold increase in the revised refinery sector SFBA CH_4 estimates (Table 2).

Figure 2B similarly compares CO_2 emissions derived using the airborne mass-balance method to self-reported inventories. Unlike CH_4 emissions, CO_2 emissions are well-characterized in bottom-up inventories. CO_2 is predominantly emitted from combustion sources and relatively

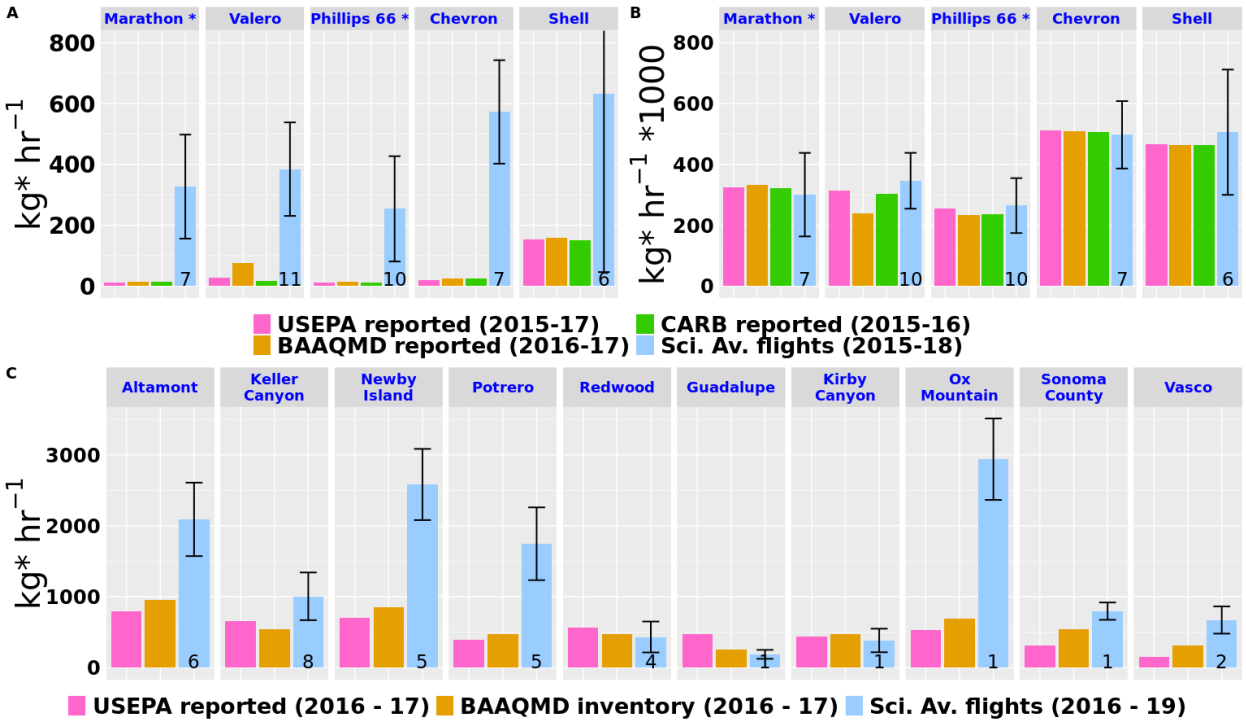

Figure 2. Bar charts comparing A) refinery CH_4 emission rates (kg h^{-1}), B) refinery CO_2 emission rates (10^3 kg h^{-1}), and C) landfill CH_4 emission rates (kg h^{-1}) derived from three self-reported inventories – BAAQMD (orange), CARB (green) and US EPA (pink) - to those derived from airplane measurements (blue). The asterisk symbol (*) indicates refineries that have separately permitted facilities (e.g., hydrogen plant and/or cogeneration power plant) within the refinery complex whose inventory emissions are included in the refinery estimates. The measured rates include emissions contributions from all facilities located within the circular flight transect around the refinery. Whiskers (in black) indicate the standard deviation of the measurements (or measurement uncertainty in case of facilities with a single measurement). The number (within the blue bar) represents the total number of flights over each facility.

simple to estimate from fuel use (tracking carbon) versus CH_4, which is generally emitted from fugitive sources or leaks. The bar chart comparison in Figure 2B indicates that CO_2 emission rates derived from airplane measurements are in good agreement with reported inventories for all

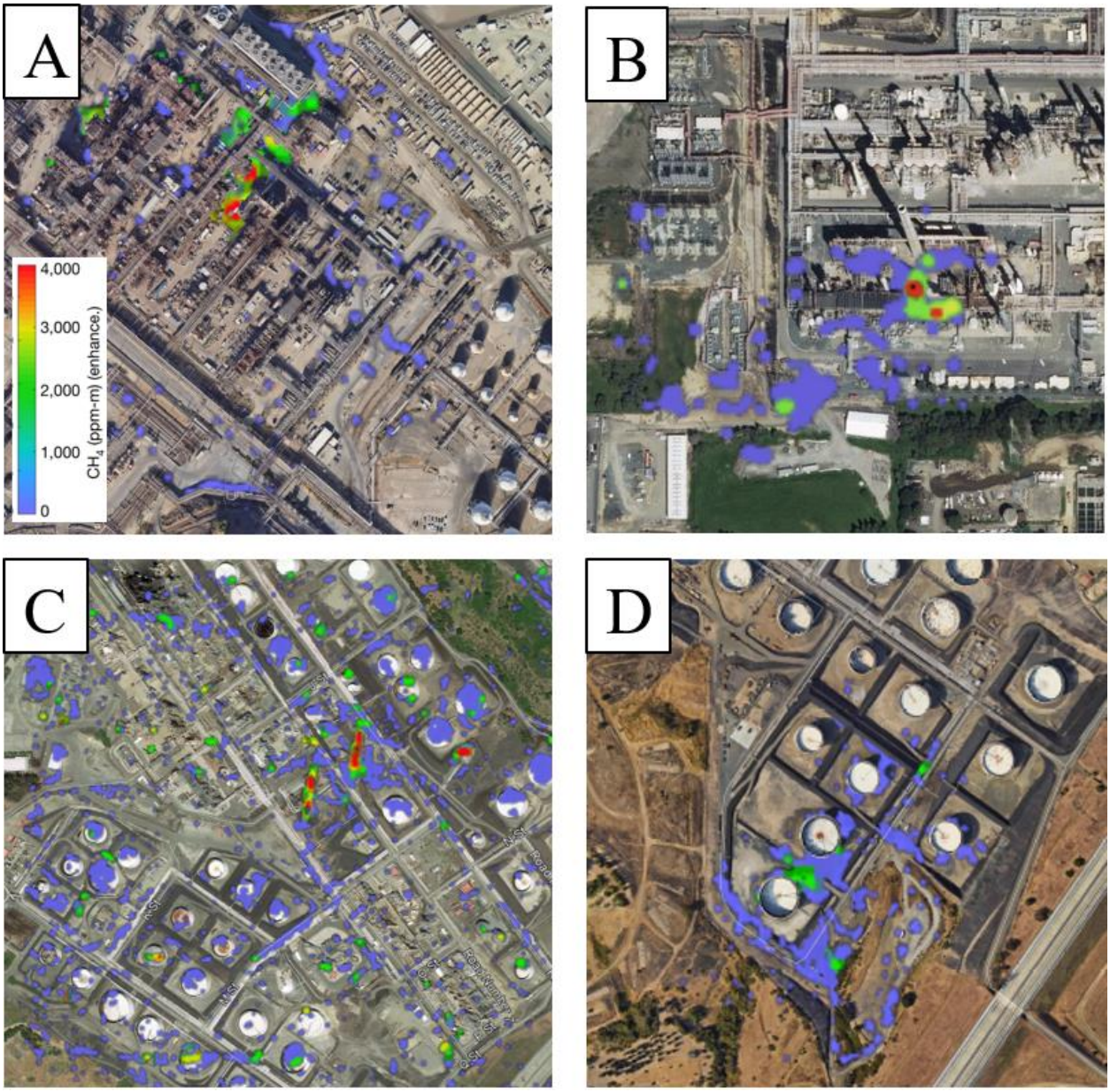

Figure 3. Remote sensing imagery collected using NASA-JPL's Airborne Visible / Infrared Imaging Spectrometer (AVIRIS-NG) over refineries in the Bay Area showing CH_4 plume hotspots coincident with location of hydrogen production plants (A and B), refinery flares (C), and organics storage tanks (D). AVIRIS-NG CH_4 plume imagery has been overlaid on true color imagery. Colors indicate enhancements over background in ppm-m (see text).

refineries. The Mooney aircraft used in this project was equipped with an onboard measurement system (Appendix 1) which is precise enough to detect small downwind concentration enhancements of long-lived tracers such as CO_2 and CH_4 (lifetime of years versus hours) above a

large local background. The close agreement between the top-down airplane measurements and bottom-up inventories for CO_2 (relatively simple to estimate) provides good confidence in the representativeness of the footprint covered and the aerial flux measurements, and underscores the importance of the significantly higher measured CH_4 emission rates compared to the reported inventories seen in Figure 2A.

Recent communications between refinery and BAAQMD staff and evaluation of process-scale activity data indicates that atmospheric venting activity at hydrogen (H_2) production plants and refinery flares are likely significant sources of missing methane observed at refineries as also indicated by remote sensing imagery of methane hotspots over SFBA refineries generated by AVIRIS-NG. [31–33] Figures 3A and 3B show AVIRIS-NG-generated CH_4 hotspot images (red spots in the images) of plumes emanating from H_2 plants at two SFBA refineries. The concentration color scale represents the magnitude of methane enhancements above background in the integrated vertical column of atmosphere between the remote sensing airplane (typically flown 3000 m above ground) and the ground surface. Magnitudes are expressed in conventional units of ppm-m, which affords an equivalence to collapsing the enhancement into a 1 m layer (i.e. 1000 ppm-m is equivalent to 1000 ppm for a 1 m layer).

H_2 gas is an essential feedstock in most petrochemical processes. H_2 is produced in large volumes at refineries using steam reformation that uses NG (>80% CH_4; public information on California NG speciation). Depending on the type of abatement technology applied, there could be residual CH_4 in the product gas following this large-scale synthesis (~5% CH_4 in methanators versus <0.1% when applying Pressure Swing Adsorption – PSA). We used refinery-reported data on daily vented H_2 volumes and vent gas chemical composition (daily to weekly) over the past 3-4 years to calculate daily CH_4 emission rate averages from H_2 production units at three SFBA

refineries that have atmospheric vents and are not equipped with PSA technology. We then plotted the hourly CH_4 emission rates derived from daily averages as a function of fraction of total number of days (~1100-1400; Figure S6A). The fraction of days with CH_4 venting rates above 20 kg h^{-1}, which represents four times the detection limit of the airborne mass balance approach,[25] ranges between 94-98% for two refineries and 54% for the third refinery. This suggests that CH_4 emissions from H_2 venting is a regular and even continous occurrence at some SFBA refineries. A given flight is likely to observe CH_4 emissions from a routine venting event resulting from regular H_2 supply-demand imbalance cycles. Figure S6B reveals that the heavy tailed distribution of CH_4 emissions observed spatially across California's industrial infrastructure by Duren at al., [33] can also manifest temporally, at least amongst refineries. We note that 10% of the days account for a cumulative 30-60% of the H_2 venting-related CH_4 across three refineries. This underscores the importance and need for highly time-resolved activity data for more accurate quantification of refinery emissions inventories. We also compare the CH_4 emission rate averages from three refineries to the airplane-observed facility-total CH_4 emission rates on the corresponding flight days (Figure S6C). Using 19 pairs of data points from three refineries, we determine that the measured and vented CH_4 are correlated at each refinery. The average slope of 0.32 represents the fraction of total airplane-observed CH_4 emissions that can be accounted for by CH_4 released from H_2 venting. Mass balance flights can also occur on days when a high-emitting and anomalous CH_4 venting event (refinery B anomaly, light brown open circle; Figure S6C) is occurring at the H_2 plant that represents a much larger proportion of the airplane-observed facility-total CH_4 emissions. Overall, frequent (planned or unplanned) atmospheric venting of product gas from H_2 plants can be an important source of CH_4 at

refineries and a significant contributor (a third or more) to the refinery methane emissions inventory.

The statewide AVIRIS-NG remote sensing imagery indicates CH_4 plumes are observed over a multitude of sources at complex facilities such as refineries and plumes are often episodic in nature.[32] Figures 3C and 3D show CH_4 plumes observed at locations coincident with those of refinery flares and organics liquid storage tanks, respectively, over two different SFBA refineries. These potentially important sources of CH_4 currently account for minor portions of self-reported CH_4 inventories submitted to US EPA.[34]

Landfills

Figure 2C shows a comparison of the measured mean CH_4 emission rates for 10 SFBA landfills (blue bars) compared to emission rates derived from two different annual inventory sources: i) BAAQMD internal estimates generated from reverse calculations based on permit data records of landfill gas combusted or collected/diverted, averaged for years 2016-18 (orange) and ii) GHG emissions data required under US EPA's 40 CFR Part 98 (pink). The measured mean CH_4 emission rates for 7 out of 10 sampled landfills are 1.5 to 4 times larger than those reported to the EPA or estimated by BAAQMD with activity parameters. For the remaining three, the mean measured rates are similar to prior estimates, agreeing within measurement uncertainty. Some landfills are sampled only once so some caution should be exercised in generating an annual estimate from a single, albeit reliable, measurement. Since landfills are the largest CH_4 emissions source in the SFBA, underestimation of emissions leads to a large difference in the bottom-up (61 Gg) versus measurement-based top-down (137 Gg) regional CH_4 totals (Table 2) making landfills the largest under-reported methane source (by emissions magnitude) in the SFBA.

Recent AVIRIS-NG imagery across California (including SFBA) has shown the active face at several landfills is likely a major CH_4 source.[35] The active face is the current working surface of a landfill where waste is being actively placed. During operational hours, waste is placed using large trucks and heavy machinery and the associated landfill surface is open to the

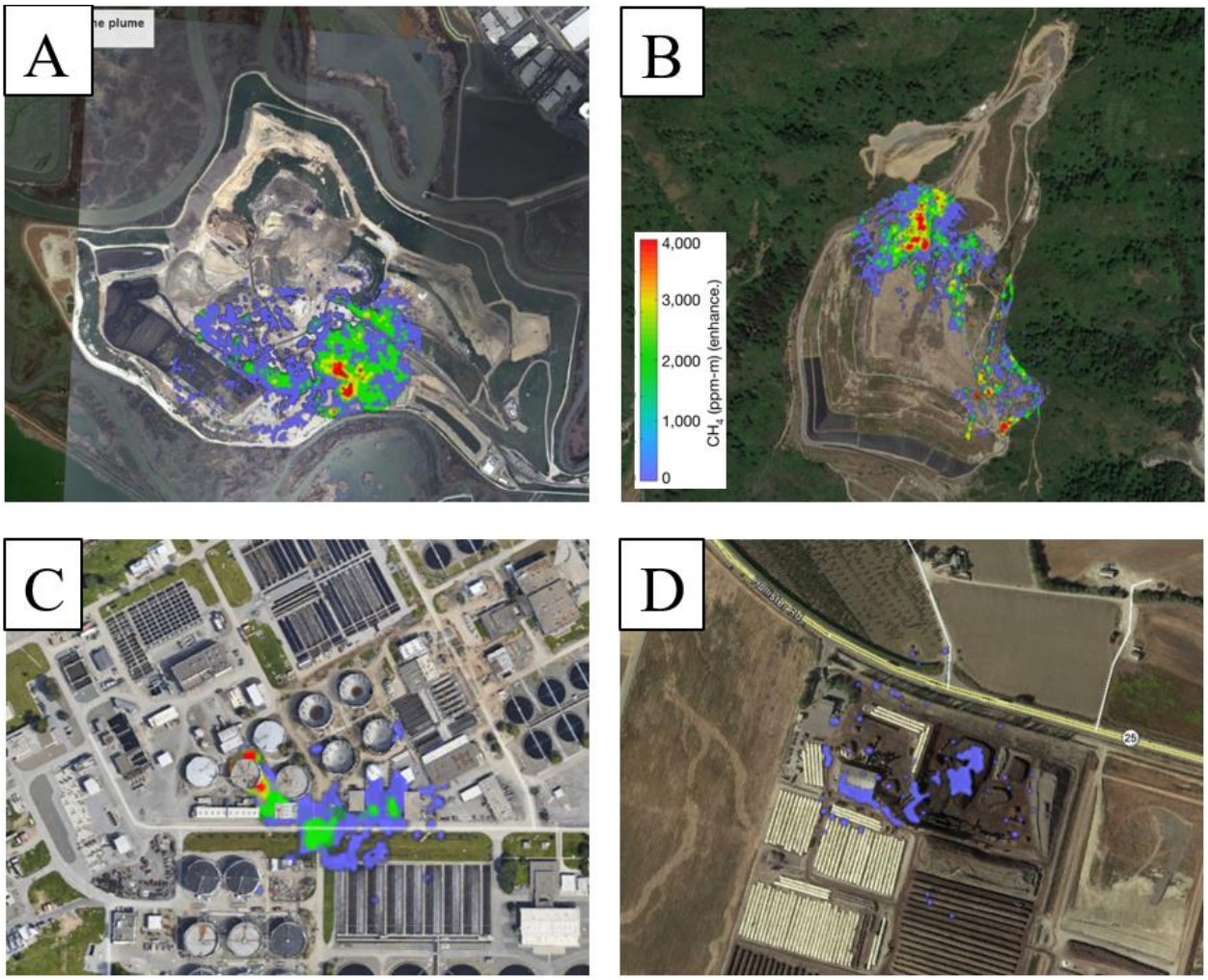

Figure 4. Remote sensing imagery collected in October 2018 using the NASA-JPL's Airborne Visible / Infrared Imaging Spectrometer (AVIRIS-NG) over known and potential biological CH_4 source facilities in the Bay Area showing CH_4 plumes hotspots over the working surface of two landfills (A and B), anaerobic digester at a publicly-owned wastewater treatment plant (C), and an organics composting facility (D). AVIRIS-NG CH_4 plume imagery has been overlaid on true color imagery.

atmosphere. After each day, this area is covered with a thin cover material that mostly serves as a deterrent to wind-blown debris, odor, and wildlife nuisance prior to commencing waste

placement the following day. Working surfaces at landfills are unsafe for conducting surface-level regulatory air monitoring and thus exempted from current state and local landfill rules. Theoretically, fresh waste deposited in the active face decomposes aerobically and should not be a source of CH_4, which is predominantly emitted through anaerobic decomposition pathways. Hence, most landfill models (and inventories) are not designed to account for CH_4 emitted at the active face. However, methane generated in the layers underlying the fresh wastes are potential sources for emissions from the active face as municipal solid waste is a relatively permeable material. Figures 4A and 4B show AVIRIS-NG generated localized CH_4 plume hotspots that broadly coincide with active face locations of two SFBA landfills. This trend is also observed in the plume imagery collected at other landfills in the SFBA (and others in California)[35] indicating that some of the missing methane observed in airborne-mass balance estimates is likely originating at the active face. This CH_4 likely originates from deeper layers of recently buried waste that have started to decompose anaerobically. The active face of a landfill, typically, lacks a landfill gas collection system to collect and divert the produced gas, and hence most of this CH_4 is likely to escape into the atmosphere. The active face CH_4 emissions need to be quantified and included in estimated and reported inventories. There are other additional sources of unaccounted for CH_4 emissions including leaks at and hotspots surrounding gas extraction wells, prolonged use of thin and highly porous, low fines content daily or intermediate covers over large surface areas, etc., some of which may be exhibited in the CH_4 plume hotspots in non-active face regions of Figures 4A and 4B.

Wastewater Treatment, Composting and Dairy Operations

Figures 4C and 4D show CH_4 plume hotspots over a SFBA POTW and a stand-alone composting facility, respectively, in October 2018. For POTWs, the regional emission rates from

airplane measurements yield a SFBA CH_4 inventory that is more than twice (Table 2) that estimated by BAAQMD using bottom-up activity data.[6] A potential cause of this discrepancy could be large observed CH_4 releases from anaerobic digesters (AD) at POTWs (Figure 4C) that are not adequately accounted for in bottom-up inventories; AD-related CH_4 emissions represent <10% of the regional, bottom-up estimates of emissions from this sector.[6]

To date, the organics diversion and composting sector has not had many permitting requirements from local air districts. Prior CH_4 inventories in the SFBA have not included GHG emissions from composting operations. This omission is due in part to a lack of representative emission factors and facility-specific activity data and in part to the unevaluated assumption that composting is an aerobic process that does not produce CH_4. With increased emphasis on composting as an important emission reduction measure in California's SB1383 regulation, and a corresponding increase in permit requests to operate such facilities in the SFBA, there is a growing need to ensure these facilities are operated with adequate performance standards such that CH_4 emissions from anaerobic decomposition pathways are properly controlled. The regional emissions estimates derived here for the largest SFBA composting facilities suggest that this sector is an important contributor to regional methane emissions (>5%). AVIRIS-NG imagery in Figure 4D shows CH_4 emissions hotspots originating from various processes within a SFBA composting facility. Remote sensing imagery and measurement-based CH_4 estimates indicate a need for improved management practices to ensure composting facilities operate under aerobic conditions. Only with improved practices can this sector play a critical role in California's CH_4 emissions reduction strategy.

Table 2. Comparison of the current, annual, bottom-up Bay Area methane emissions inventory[6] to revised estimates derived from airborne mass-balance measurements (Gg).

Sector	Subsector	Current Inventory (Gg CH4/a)	% of Total Emissions	Revised Inventory (Gg CH4/a)	% of Total Emissions
Livestock	Dairy	9.2		5.0	
	Major non-dairy^	8.5		8.5	
	Poultry^	0.06		0.06	
	Other animals^	0.5		0.5	
	Sector Total	**18.3**	**15**	**14.1**	**6**
Landfill	Point Source	56.9			
	Fugitive Area Source	4.6			
	Sector Total	**61.5**	**51**	**136.7[$]**	**60**
Natural gas	Distribution^	17.3		17.29	
	Domestic Natural Gas^	0.05		0.05	
	Other combustion^	0.16		0.16	
	Sector Total	**17.5**	**15**	**17.5**	**8**
On-road mobile	On-road mobile^	2.2		2.2	
	Sector Total	**2.2**	**2**	**2.2**	**1**
Refinery	Refinery	1.9			
	Sector Total	**1.9**	**2**	**19.0**	**8**
Wastewater	Domestic wastewater	5		11.9	
	Industrial wastewater^	2		2.0	
	Sector Total	**7**	**6**	**13.9**	**6**
Composting	**Sector Total**	**--**	**--**	**11.1**	**5**
Others*		8.1	7	8.1	4
Anthropogenic total		**116.4**		**222.6**	
Wetlands[#]		3.7	3	3.7	2
Total		**120.1**		**226.3**	

^ No airborne measurements therefore current and revised estimates remain the same

$ Airplane measurements represent the sum of all sources in a facility including composting

* Includes emissions from other stationary combustions, aircraft, off-road emissions, etc.

SFBA wetland emission estimate based on study by Potter et al., 2006.[36]

--: not available

Most of SFBA's dairies are located in three counties: Marin, Sonoma, and Solano. These dairies typically operate differently from the industrial-scale dairies in California's Central

Valley. Unlike their Central Valley counterparts, most have <1000 milk cows that graze and deposit manure on grass pastures rather than being fed from troughs and having manure flushed to anaerobic waste treatment lagoons (a significant CH_4 source). Regional CH_4 estimates derived from limited airplane measurements suggest that the bottom-up techniques may be overestimating SFBA's dairy operations CH_4 inventory (Tables 1 & 2), though large uncertainties remain due to limited number of observations. The overestimation in the inventory is possible because bottom-up estimates for dairies in Table 2 are derived from California-specific enteric fermentation and manure management emission factors that are based on data collected at large-scale, commercial dairy operations in the Central Valley, which are not representative of the open pasture-styled dairy operations in the SFBA.

SFBA Regional Methane Emissions

Table 2 lists SFBA sector-wide CH_4 emissions estimates generated using a combination of measurement-based and current bottom-up estimates. Although, the airplane measurement-based assessment is limited to those source categories that have been sampled, it includes those that have significant contributions to the bottom-up CH_4 inventory. Using this hybrid approach, we estimate a revised Bay Area CH_4 emissions total of 226 ±40 Gg (including wetland contributions) which represents nearly a two-fold increase compared to BAAQMD's current estimates that use bottom-up methods only. This revised regional CH_4 emissions estimate agrees well with two top-down Bay Area regional CH_4 emission assessments conducted in the 2014-16 period by Fairley and Fischer[22] (240 ±60 Gg) and Jeong et al.[23] (166-289 Gg).

Future Inventory Assessment

An assessment of CH_4 emissions using an airborne mass-balance technique suggests that this can be a useful tool for further evaluation of bottom-up inventories and can uncover both

underestimation and overestimation in such inventories. In this study, we extend our understanding of sector-level CH_4 apportionment (using airborne mass balance) and CH_4 source attribution (using remote sensing) and provide key insights into the missing methane question. These measurements need to be continued and extended to more facilities (and categories) to improve statistical confidence and reduce uncertainties in our estimates. Repeat sampling over previously measured facilities will provide additional information on the persistence of high-emitting but episodic methane sources. Co-application of airborne flux measurements with remote sensing capabilities such as AVIRIS-NG will be an effective approach for facility-scale emissions quantification along with identification and attribution of emission sources from concurrent sub-facility-scale emission hotspots.

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Supporting Information – Description of airborne flux estimation method and uncertainty estimation; estimation of actual day to day variations at refineries; a two-method analysis of refinery emissions data to calculate total emissions and uncertainties, statistical estimation of uncertainty in sector-wide emissions; an alternate emissions-to-throughputs ratio analysis for composting sector; a figure demonstrating significance of methane venting at refinery hydrogen production plants; five tables summarizing 92 individual methane flux measurements collected over 5 refineries, 10 landfills, 4 wastewater treatment plants (POTWs), 2 composting operations, and 2 dairies; and three tables with facility lists containing inventory landfill methane emissions and material throughputs for composting and POTWs.

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