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August 6, 2020

Mr. Ken DaRosa, Acting Director
California Department of Resources Recycling and Recovery
P.O. Box 4025
Sacramento, CA 95812

Dear Ken DaRosa:

RECOMMENDATIONS TO MAXIMIZE IMMEDIATE REDUCTIONS IN SHORT-LIVED CLIMATE POLLUTANTS BY UTILIZING ALTERNATIVE TECHNOLOGIES

The Los Angeles County Integrated Waste Management Task Force (Task Force) would like to respectfully offer CalRecycle for consideration its recommendations to maximize reductions in short-lived climate pollutants and help achieve California's climate change objectives. The Task Force believes implementation of the recommendations outlined herein will also help California achieve maximum short-term reductions in landfill-generated methane gas, a powerful short-lived climate pollutant.

Methane's Contribution to Climate Change

Methane is a potent greenhouse gas (GHG) and a short-lived climate pollutant, which has been found to be 86 times more effective than carbon dioxide (CO2) in trapping infrared radiation over a 20-year time frame. Because of its short lifetime (about 10 years) as compared to that of CO2 (about 100 years), expedited methane emission reduction efforts can provide substantial near-term climate benefits.

For this reason, a key strategy in California's efforts to fight climate change is focused on reducing landfill-generated methane emissions by reducing the landfill disposal of organic waste. However, the infrastructure that is necessary to recycle or otherwise manage organic waste currently going to landfills is yet to be developed and it will take years to plan, permit, and develop. Therefore, processes and technologies that maximize near-term reductions in landfill disposal of organic waste, including existing infrastructure, need to be incorporated as part of a comprehensive climate change strategy.

The GHG Emissions Reduction Advantage of Composting May be Limited

It is widely assumed that composting is superior to most other organic waste management processes and technologies in reducing GHG emissions from landfills. However, this

may not be the case. A recent study (Assessment of Regional Methane Emission Inventories through Airborne Quantification in the San Francisco Bay Area) deriving methane emission rates from 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 (SFBA) revealed that composting facilities generate significant methane emissions (see enclosure). The study concluded that anaerobic pathways are likely dominant at these composting facilities (thereby generating methane), whereas it is generally assumed that an aerobic process occurs converting carbon in the organic matter to carbon dioxide (CO_2).

The study, which was published on July 7, 2020, by ACS, Environmental Science & Technology (see enclosure), concludes that *"**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." (emphasis added)*

The study further notes that, *"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 that these facilities are operated with adequate performance standards such that CH4 emissions from anaerobic decomposition pathways are properly controlled**. The regional emission 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…shows CH4 emission hotspots originating from various processes within a SFBA composting facility. Remote sensing imagery and measurement-based CH4 estimates indicate a need for improved management practices to ensure composting facilities to operate under aerobic conditions. **Only with improved practices can this sector play a critical role in California's CH4 emission reduction strategy**." (emphasis added)*

The Need for a Comprehensive State Strategy

California needs a comprehensive strategy that does not rely almost exclusively on composting. In addition to the challenge of controlling methane emissions at composting facilities (and methane's powerful capacity to trap infrared radiation), California will face a great challenge in developing markets for the massive quantities of compost that will be produced if California continues in its current compost-at-any-cost path. A market saturated with an oversupply of compost and mulch will likely result in the disposal of the compost/mulch in rural areas as "land application" or "erosion control," further generating methane emissions and impacting the natural landscapes.

These situations may be avoided if California adopts a diversified GHG reduction strategy that, in addition to composting, takes full advantage of available alternative technologies and processes, such as non-combustion thermal technologies, that are extremely

effective in reducing methane emissions compared to landfilling and are capable of producing renewable energy and clean fuels. A diversified strategy can also maximize short-term reductions in methane generation by diverting solid waste from landfills to existing waste-to-energy facilities until adequate organic waste recycling infrastructure is developed.

Recommendations

Therefore, the Task Force urges CalRecycle to consider alternative technologies such as waste-to-energy (WTE) and non-combustion thermal conversion technologies (CTs) that process organic waste as reductions in landfill disposal under SB 1383. These technologies can help divert hard-to-process organic waste such as paper (including food-soiled paper), cardboard, textiles, and carpets. Unlike mulching or chipping and grinding, these technologies also provide a way of managing organic waste without opening a potential pathway for the spread of COVID-19 and other contaminants. Furthermore, these technologies are being used in Europe and Asia to effectively process organics and mitigate GHG emissions.

The draft SB 1383 regulations propose to allow new technologies to be considered as reductions in landfill disposal through the "Determination of Technologies That Constitute a Reduction in Landfill Disposal" process. However, CalRecycle should consider the permanent lifecycle of GHG emissions reduced by these technologies in comparison to the GHG emissions produced by landfills, instead of composting facilities, in making its determination.

WTE and CTs are capable of converting organic waste to renewable, negative-carbon electricity and/or fuels that reduce emissions of methane and GHGs. Considering these methane-reducing technologies as reductions in landfill disposal will have immediate short-term methane reduction benefits and significantly assist California in achieving its GHG emissions reduction goals. Implementing these changes would also help local jurisdictions divert more organic waste from landfills to meet the state's goal to reduce organic waste disposal 75 percent by the year 2025.

Pursuant to Chapter 3.67 of the Los Angeles County Code and the California Integrated Waste Management Act of 1989 (Assembly Bill 939), the Task Force is responsible for coordinating the development of all major solid waste planning documents prepared for the County of Los Angeles and the 88 cities in Los Angeles County with a combined population in excess of ten million. Consistent with these responsibilities and to ensure a coordinated and cost-effective and environmentally sound solid waste management system in Los Angeles County, the Task Force also addresses issues impacting the system on a countywide basis. The Task Force membership includes representatives of the League of California Cities-Los Angeles County Division, County of Los Angeles Board of Supervisors, City of Los Angeles, waste management industry, environmental groups, the public, and a number of other governmental agencies.

We thank you in advance for your consideration of these recommendations. If you have any questions, please contact Mr. Mike Mohajer, a member of the Task Force, at MikeMohajer@yahoo.com or at (909) 592-1147.

Sincerely,

Margaret Clark

Margaret Clark, Vice-Chair
Los Angeles County Solid Waste Management Committee/
Integrated Waste Management Task Force and
Mayor, City of Rosemead

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Enc.

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California Air Resources Board (Mary Nichols and David Mallory)
California Department of Fish and Wildlife (Chuck Bonham)
California Department of Food and Agriculture (Secretary Karen Ross)
California Department of Public Health (Director Karen Smith)
League of California Cities
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Los Angeles County Agricultural Commission
Each City Mayor/Manager in the County of Los Angeles
South Coast Air Quality Management District
South Bay Cities Council of Governments
San Gabriel Valley Council of Governments
Gateway Cities Counsel of Governments
Each City Recycling Coordinator in Los Angeles County
Each Member of the Los Angeles County Solid Waste Management Committee/Integrated Waste Management Task Force
Each Member of the Task Force Alternative Technology Advisory Subcommittee
Each Member of the Task Force Facility and Plan Review Subcommittee

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Article

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

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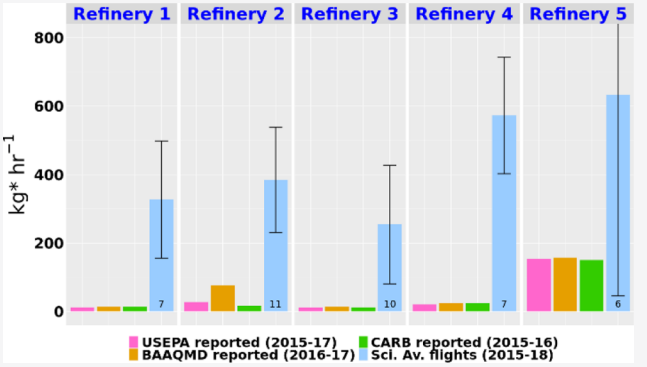
ABSTRACT: This study derives methane emission rates from 92 airborne observations collected over 23 facilities including 5 refineries, 10 landfills, 4 wastewater treatment plants (POTWs), 2 composting operations, and 2 dairies in the San Francisco Bay Area. Emission rates are measured using an airborne mass-balance technique from a low-flying aircraft. Annual measurement-based sectorwide methane emissions are 19,000 ± 2300 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 the 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 subfacility scales to guide and verify effective mitigation policy and action.



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 preindustrial times.[1] Because of its short lifetime (~10 years) as compared to that of CO_2 (~100 years),[2] expedited CH_4 emission 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 emission 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, as per 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;

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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 multipollutant emission 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 emission inventory of GHGs.

Emission inventories at the regional, state, and federal levels are typically generated using simple combinations of emission factors and activity data, and the evaluation of inventory emission estimates requires a comparison against top-down measurement-based estimates. At the state level, several recent top-down emission 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 the underestimation of CH_4 emissions[14,16−19] albeit with a decreasing trend[20] and misallocation of emissions within sectors.[21] Recent top-down emission 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 emission estimates to derive a regional mean CH_4 emission estimate of 240 ± 60 Gg CH_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 to 289 Gg CH_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 multispecies hierarchical Bayesian method to conduct source apportionment analysis combining concurrently measured volatile organic compound 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 that inventories undercount CH_4 emissions by approximately a factor of 1.5−2.

These discrepancies, combined with the need for representative emission 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 emission 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 the Supporting 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 Mehrotra et al.'s 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 undersampled sources such as refineries and previously unsampled sources including composting. Regional sector-specific CH_4 emission 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 emission 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 with 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 Supporting Information. Many sites were sampled multiple times. The aircraft typically spent 20−60 min 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 summarized in the Supporting Information (Appendix 1).

Sector-Specific Emission 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 emission data, day-to-day variations at refineries were statistically significant (see Appendix 2 in the Supporting Information) and indicate that there are process-level activity variations that result in varying emissions (see the "Source Attribution and Discussion" section). We estimated annual emissions at each facility from multiple measurements over a 4 year period. Main assumptions for the estimates include that CH_4 emissions across diurnal cycles (daytime vs nighttime) do not vary a lot and that the multiyear record of individual observations capture a representative sampling of the variability in the

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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 emission inventories, and this leads to our major conclusions.

The airborne measurement approach presented in this study is focused on the 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, such as 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 on 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. As 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 nonsampled facilities and thereby scaled up to the entire sector emission 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 emission estimates, throughput or activity data were used as covariates: for POTWs, throughput of the 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 Supporting Information demonstrates that there were significant day-to-day variations in the refinery emission data above the measurement uncertainty. The individual uncertainties of airplane measurements varied as well. To test how incorporating these uncertainties would affect the emission 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 measurements and uncertainties (see the Supporting 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 arithmetic means approach. Table 1 summarizes the measurement-derived CH_4 emission totals, summing the contributions from sampled and nonsampled facilities for source sectors with airborne measurements, including estimates of uncertainties.

Refineries. A total of 41 measurements were made over 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 the observed CH_4 emission 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 $\overline{y}_i$. Total emissions are computed as $T = \overline{y}_1 + \overline{y}_2 + \ldots + \overline{y}_S$, while the variance of the total emissions is estimated as $s_T^2 = s^2 \sum_{i=1}^{5} 1/n_i$, where $s^2 = \frac{\sum_{i=1}^{5}\sum_{j=1}^{n_i}(y_{ij}-\overline{y}_i)^2}{41-5}$. 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 (multiyear averaged) CH_4 emissions, emphasizing that the inventory for individual refineries is underestimating averaged emissions 4- to 23-fold.

Table 1. Measurement-Based Sector-Specific Methane Emission Estimates Compared to BAAQMD's Annual Prior Bottom-Up Inventory Estimates (1 Gg = 1000 metric tons)

		BAAQMD Prior	airplane mass balance	
source category		estimate (Gg)	estimate (Gg)	standard error (Gg)
refineries	total	1.9	19.0	2.3
landfills	sampled	47	110.0	8.9
	non-sampled[a]	11	26.7	17.4
	total	58	136.7	25.9
wastewater treatment plants (POTWs)	sampled	NA	5.7	1.0
	non-sampled[a]	NA	6.3	0.8
	total	5	11.9	1.5
composting operations	sampled	NA	4.4	0.9
	non-sampled[a]	NA	6.7	2.0
	total	NA	11.1	3.4
dairy operations	sampled	NA	0.4	0.2
	non-sampled[a]	NA	4.6	2.6
	total	9.2	5.0	2.9

[a]Nonsampled 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.

Landfills. BAAQMD's 2016−2018 GHG Emission 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 emission 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 $T_S = \sum_{i \in S} \overline{y}_i$, where $\overline{y}_i$ is the mean airplane-estimated methane from landfill i and **S** is the set of sampled landfills. A regression of facility averages of airborne measurements against inventory estimates is used to estimate the annual emissions from nonsampled landfills. Therefore, a simple linear regression is done with the 10 landfill means, $\overline{y}_i$, as the dependent variable and x_i, the prior estimate as the independent variable. The intercept is nonsignificant (−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 $T_U = \sum_{i \in U} 2.4 x_i = 2.4 \sum_{i \in U} x_i$, where **U** is the set of unsampled landfills. The uncertainty is the slope uncertainty times the sum of x_i: standard error of $T_U = 0.36 \sum_{i \in U} x_i$. The total emissions combining the estimates from the sampled and nonsampled landfills are $T = T_S + T_U$. A derivation of the variance of T is presented in Appendix 4 in the Supporting Information. This

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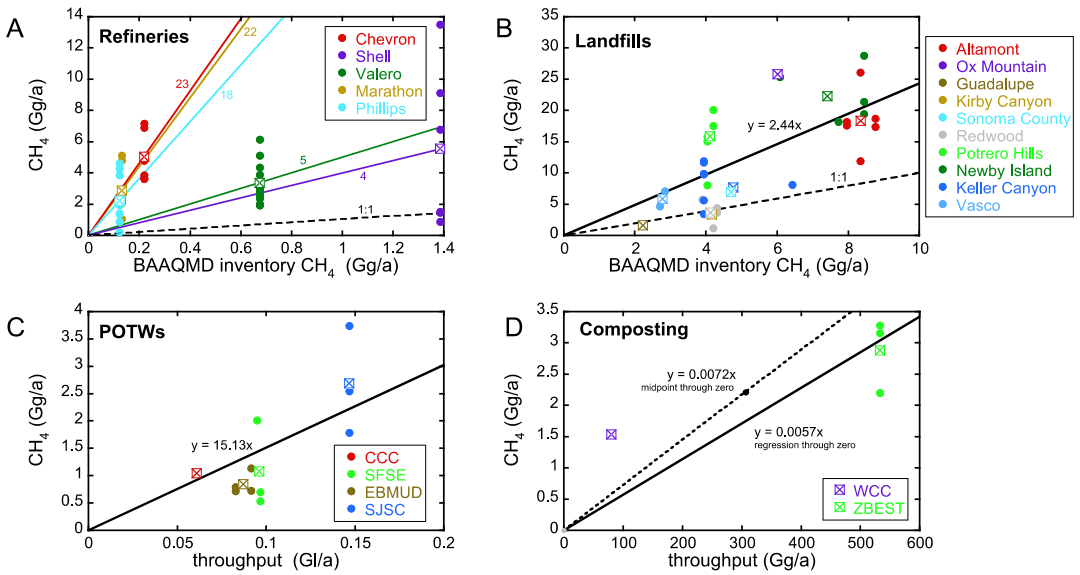

Figure 1. Annual CH_4 emission inventories from (A) refineries, (B) landfills, (C) wastewater treatment plants (POTWs), and (D) composting operations (shown in Gg CH_4) measured by the aerial mass-balance method vs prior inventory estimates (A,B) or throughputs (C,D). 1 Gg = 1000 metric tons; 1 Gl = 10^9 L; and 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 the ratios of measured to inventory CH_4 for different refineries. The dashed line in (A,B) is the 1:1 line, shown for refineries and landfills to emphasize the underestimation in the prior emission inventory. The dotted line for composting is the line between the midpoint of the two facilities and zero.

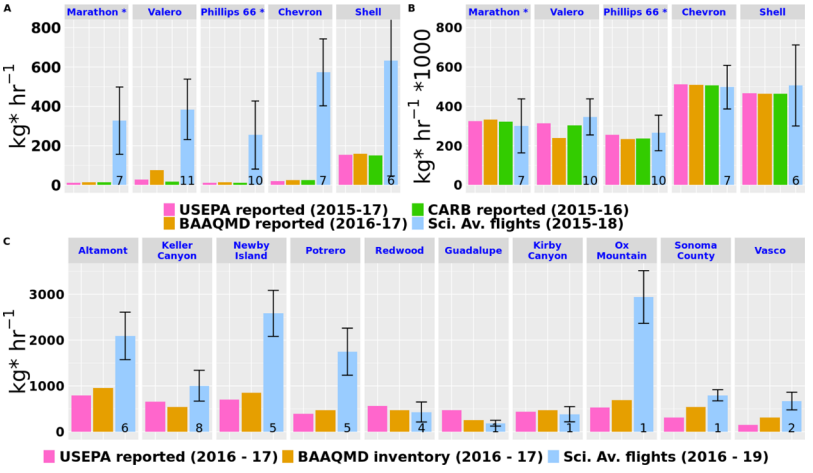

Figure 2. Bar charts comparing (A) refinery CH_4 emission rates (kg h^{-1}), (B) refinery CO_2 emission rates (10^3 kg h^{-1}), and (C) landfill CH_4 emission rates (kg h^{-1}) derived from three self-reported inventories—BAAQMD (orange), CARB (green), and US EPA (pink)—to those derived from airplane 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 emission 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 the case of facilities with a single measurement). The number (within the blue bar) represents the total number of flights over each facility.

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. Airplane measurements were made over 4 of SFBA's ~50 publicly owned sewage treatment works (POTWs). The sampled facilities are among the largest POTWs in the region by throughput of the 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 emission inventory for the POTW sector is not sufficiently quantified at the facility level to include all possible sources. As bottom-up emission inventories for POTWs are based on the 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 as an assumption is made

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Table 2. Comparison of Current Annual Bottom-Up Bay Area Methane Emission Inventory[6] to Revised Estimates Derived from Airborne Mass-Balance Measurements (Gg)

sector	subsector	current inventory(Gg CH_4/a)	% of total emissions	revised inventory(Gg CH_4/a)	% of total emissions
livestock	dairy	9.2		5.0	
	major nondairy[a]	8.5		8.5	
	poultry[a]	0.06		0.06	
	other animals[a]	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[b]	60
-natural gas	distribution[a]	17.3		17.29	
	domestic[a]	0.05		0.05	
	other combustion[a]	0.16		0.16	
	sector total	17.5	15	17.5	8
on-road mobile	on-road mobile[a]	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[a]	2		2.0	
	sector total	7	6	13.9	6
composting	sector total	[e]	[e]	11.1	5
others[c]		8.1	7	8.1	4
anthropogenic total		116.4		222.6	
wetlands[d]		3.7	3	3.7	2
total		120.1		226.3	

[a]No airborne measurements remain the same for the current and revised estimates. [b]Airplane measurements represent the sum of all sources in a facility including composting. [c]Includes emissions from other stationary combustions, aircraft, off-road emissions, etc. [d]SFBA wetland emission estimate based on the study by Potter et al., 2006.[36] [e]Not available.

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 that 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 emission estimates versus the annual throughput of waste feedstock processed. As 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 Supporting 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 as 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 the 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 the 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), respectively, 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 sectorwide 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. As 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 sectorwide emission

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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 subfacilities are minor CH_4 sources (<5%) in reported inventories (although major sources of CO_2). The CH_4 emissions from these subfacilities are included in the refinery totals (for all three reported inventories) to ensure a direct comparison with emissions obtained from airplane measurements. The measured average CH_4 emission rates in Figure 2A range from 4 to 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 10-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 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 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 vs 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 indicate that the 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] Figure 3A,B shows 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

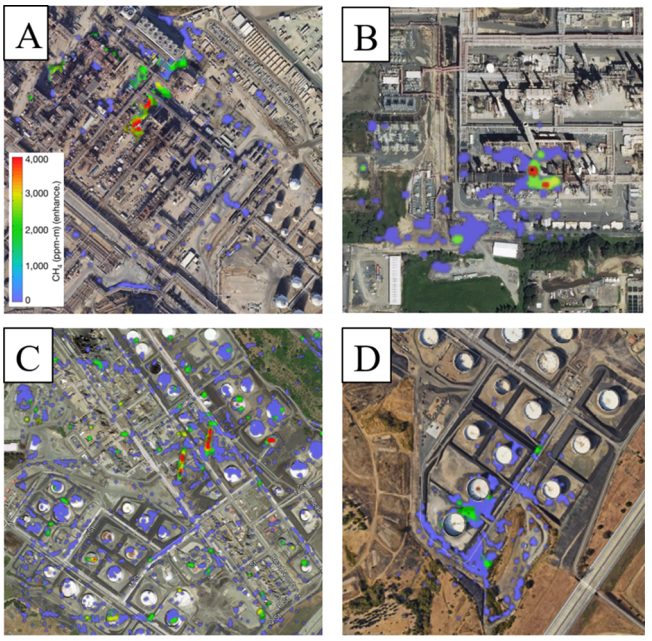

Figure 3. Remote sensing imagery collected using NASA-JPL's Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG) over refineries in the Bay Area showing CH_4 plume hotspots coincident with the location of hydrogen production plants (A,B), refinery flares (C), and organics storage tanks (D). AVIRIS-NG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

the integrated vertical column of the atmosphere between the remote sensing airplane (typically flown 3000 m above the 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 vs <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 the PSA technology. We then plotted the hourly CH_4 emission rates derived from daily averages as a function of the 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 4 times the detection limit of the airborne mass-balance approach,[25] ranges between 94 and 98% for two refineries and 54% for the third refinery. This suggests that CH_4 emissions from H_2 venting are a regular and even continuous 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 et al.[33] can also manifest temporally, at least among refineries. We note that 10% of the days

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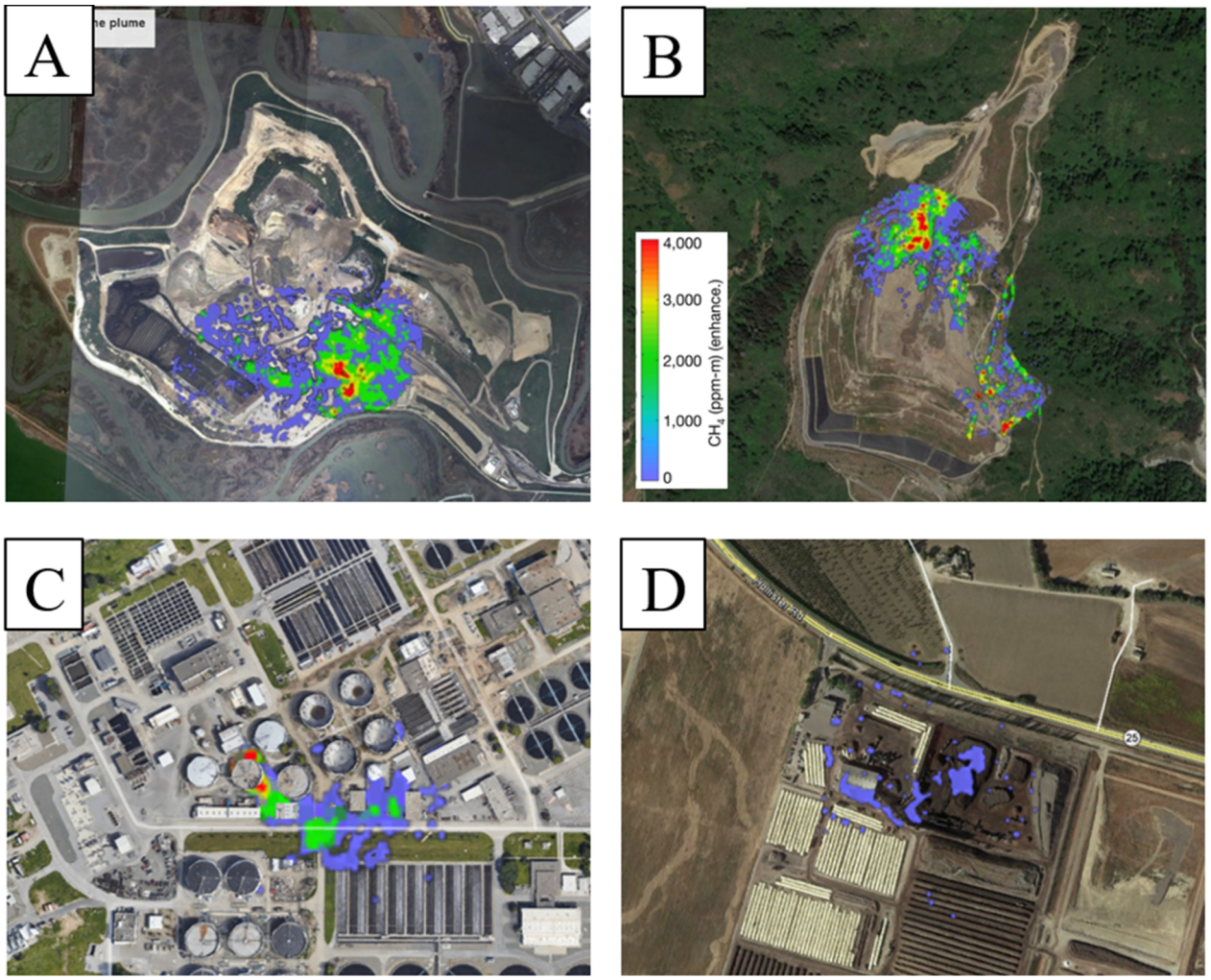

Figure 4. Remote sensing imagery collected in October 2018 using the NASA-JPL's Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG) over known and potential biological CH_4 source facilities in the Bay Area, showing CH_4 plume hotspots over the working surface of two landfills (A,B), anaerobic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

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 emission 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 emission inventory.

The statewide AVIRIS-NG remote sensing imagery indicates that CH_4 plumes are observed over a multitude of sources at complex facilities such as refineries, and plumes are often episodic in nature.[32] Figure 3C,D shows CH_4 plumes observed at locations coincident with those of refinery flares and organic 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 2016−2018 (orange) and (ii) GHG emission 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. As landfills are the largest CH_4 emission source

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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 total (Table 2), making landfills the largest underreported methane source (by emission magnitude) in the SFBA.

Recent AVIRIS-NG imagery across California (including SFBA) has shown that 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 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 the current state and local landfill rules.

Theoretically, fresh waste deposited on 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 waste is a potential source for emissions from the active face as municipal solid waste is a relatively permeable material. Figure 4A,B shows 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 likely originates 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 the estimated and reported inventories. There are other additional sources of unaccounted-for CH_4 emissions including leaks at and hotspots surrounding the gas extraction wells, prolonged use of thin and highly porous, low-fine content daily or intermediate covers over large surface areas, etc., some of which may be exhibited in the CH_4 plume hotspots in the nonactive face regions of Figure 4A,B.

Wastewater Treatment, Composting, and Dairy Operations. Figure 4C,D shows 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 (ADs) 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 organic 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 that these facilities are operated with adequate performance standards such that CH_4 emissions from anaerobic decomposition pathways are properly controlled. The regional emission 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 emission 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 to operate under aerobic conditions. Only with improved practices can this sector play a critical role in California's CH_4 emission reduction strategy.

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 and 2), though large uncertainties remain because of the 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 sectorwide CH_4 emission 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 emission total of 226 ± 40 Gg (including wetland contributions), which represents nearly a 2-fold increase compared to BAAQMD's current estimates that use bottom-up methods only. This revised regional CH_4 emission estimate agrees well with two top-down Bay Area regional CH_4 emission assessments conducted in the 2014–2016 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

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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. Coapplication of airborne flux measurements with remote sensing capabilities such as AVIRIS-NG will be an effective approach for facility-scale emission quantification along with identification and attribution of emission sources from concurrent subfacility-scale emission hotspots.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c01212.

Description of airborne flux estimation method and uncertainty estimation; estimation of actual day-to-day variations at refineries; a two-method analysis of refinery emission data to calculate total emissions and uncertainties; statistical estimation of uncertainty in sectorwide emissions; an alternate emission-to-throughput ratio analysis for the composting sector; significance of methane venting at refinery hydrogen production plants; summary of 92 individual methane flux measurements collected over 5 refineries, 10 landfills, 4 wastewater treatment plants (POTWs), 2 composting operations, and 2 dairies; and facility lists containing inventory landfill methane emissions and material throughputs for composting and POTWs (PDF)

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Notes

The authors declare no competing financial interest.

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