Quantifying Methane Emissions from United States Landfills

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Abstract: Methane emissions from solid waste may represent a significant fraction of the global anthropogenic budget, but few comprehensive studies exist to assess inventory assumptions. We quantified emissions at hundreds of large landfills across 18 states in the United States (U.S.) between 2016-2022 using airborne imaging spectrometers. Spanning 20% of open U.S. landfills, this represents the most systematic measurement-based study of methane point sources of the waste sector. We detected significant point source emissions at a majority (52%) of these sites, many with emissions persisting over multiple revisits (weeks to years). We compared against independent contemporaneous *in situ* airborne observations at 15 landfills and established good agreement. Our findings indicate a need for long-term, synoptic-scale monitoring of landfill emissions in context of climate change mitigation policy.

One-Sentence Summary: Comprehensive methane airborne surveys of hundreds of landfills in the U.S. show persistent point source emission behavior.

Main Text:

Introduction:

Landfill methane (CH_4) emissions are estimated to make up nearly 20% of global anthropogenic CH_4 emissions (years 2000-2017) (*1*), and 17% of U.S. anthropogenic CH_4 emissions (years 1990-2020) (*2*). Those estimates are almost entirely driven by bottom-up process models that have not been comprehensively validated by direct measurement across a broad population of global landfills and dumpsites. Landfill gas emission models generally rely on waste tonnage, decay parameterizations, and some estimate of gas capture, if applicable (75% is commonly used in U.S.) in order to estimate annual CH_4 emissions (*3, 4*) These parameters are difficult to generalize, as they rely on factors intrinsic to a particular landfill, regional waste stream (e.g., proportion of organic material), operator practices, and jurisdictional oversight. To the best of our knowledge, direct measurements of methane emissions at landfills to date using surface or aircraft instruments have largely been limited to a small number of facilities due primarily to cost, which has resulted in incomplete spatial and temporal sampling. Given the diversity of operational and environmental factors driving landfill emissions, these observational limitations lead to continued uncertainty in this sector's contribution to regional, national and global CH_4 emission inventories, which can complicate assessing the efficacy of emission mitigation efforts.

Methane emissions at solid waste sites result from several processes. Nearly every location with buried organic waste will generate CH_4 gas at some timescale (*5, 6)*. A fraction of that generated gas may escape to the atmosphere through transport or diffusion through soil layers taking the path of least resistance (*7*). As such, changes in barometric pressure have been shown to influence emission variability (*8-10*). However, current datasets are insufficient to represent pressure-emission relationships at typical landfills with variable topography, landfill design and operation, waste composition and quantity, gas capture and collection, water management, and daily working face design and operation (*10*). Fugitive emissions of landfill gas can occur due to under sizing air pollution control equipment, cracks in cover due to drought, side slope erosion, and how the working face is operated. Emissions can also result from extreme precipitation events, as landfill gas wells can be disconnected from landfill gas header pipes due to high level of liquids. These emissions may manifest as a distributed diffuse "area source" over a wide area of the waste site, or as a "point source" localized to a certain region or

hotspot of the site. Waste sites may also contain multiple point or area sources (or both) at any given time. Area source emissions are constrained by the CH_4 generation potential at a landfill, but point sources are more likely related to the dynamic operational nature of a landfill. For example, planned maintenance or construction at a landfill or equipment failures can result in highly concentrated CH_4 point sources that can persist for periods ranging from hours to months. For safety reasons, operators may also "under-pull" or apply less vacuum to a gas collection system to avoid excess oxygen from entering the soil.

In the U.S., CH_4 is most frequently measured at landfills through surface emission monitoring (SEM) walking surveys. These generally involve a human equipped with a low sensitivity methane detector (e.g., flame ionization detector) walking along a serpentine path across portions of the landfill and logging the coordinates of any exceptionally high detected surface concentrations greater than 500 ppm [40 CFR 63.1958(d); 40 CFR 63.1960(c) & (d)]. Walking surveys are complicated by the fact that many locations on an active landfill are unsafe to measure (e.g., working face–area where new trash is deposited, or steep side slopes). SEM surveys are federally required only four times per year for most landfills, which limits their ability to capture any dynamics in emissions. SEM survey accuracy is also highly dependent on the human operator and exact choice of measurement locations, with the result that high emission locations potentially can be missed entirely. Additionally, SEM measurements do not explicitly represent an emission rate, but instead are designed to flag CH_4 concentration "hot spots" that may indicate a potential regulatory exceedance. Actual quantification of landfill emission fluxes requires the concurrent observation of CH_4 concentration fields and surface wind speed which often requires the use of sophisticated atmospheric transport modeling. Other studies have leveraged various ground and aerial based technologies to measure landfill gas emissions, using technologies like eddy-covariance, radial plume mapping, tracer correlation, and flux chambers among others (*11, 12*). However, due to the complexity operating these measurement systems, these studies are often limited to a small sample of landfills, making extrapolation to larger waste sector dynamics difficult.

Although strategies to compare, design, and scale emission quantification technologies tailored for landfill CH_4 quantification continue to develop, there is an immediate need to make a baseline observational assessment of CH_4 emissions across a large swath of waste sites. Remote sensing offers an efficient method for surveying widely dispersed waste sites without costly and

time-consuming efforts to gain access to facilities with surface-based observations. In 2016 and 2017, airborne imaging spectroscopy was used to observe more than 400 active and closed landfills and waste diversion sites in California as part of the California Methane Survey (*13*). This observational approach is sensitive to high emission CH_4 point sources (typically greater than 10 kg h^{-1} for typical wind speeds and surface albedo), produces high spatial resolution plume maps of emission hotspots, and can quantify emissions of those hotspots under adequate observing conditions. Here "plume" refers to a region of contiguous pixels of elevated CH_4 concentrations that is observed by an imaging spectrometer and attributable to landfill gas emissions. These plume maps have, in turn, been used to guide operators in locating emission sources at landfills and prompted mitigation (*14*).

Patterns of high emission point sources at landfills revealed by the California Methane Survey suggest that persistent super-emitter activity could be prevalent more broadly across the solid waste management sector in the U.S. To test this hypothesis, we generated an observationally based CH_4 dataset spanning a diversity of U.S. climate zones and jurisdictions including repeat observations over multiple seasons and in some cases years. The sites surveyed in this study represent the largest airborne or ground-based survey of U.S. landfills to date, totaling 250 Greenhouse Gas Reporting Program (GHGRP) landfills in 18 U.S. states, surveyed between 2018-2022. We analyzed this statistically robust data set to assess at what rate point source emissions are prevalent at large (i.e., GHGRP-reporting) managed landfills, how long they persist, and whether the magnitude of quantified emission rates are consistent with reported values. This dataset is key for future comparison to other non-U.S. jurisdictions, especially for regions who lack waste management but are looking to incorporate more recommended practices for emission mitigation and public health improvement. In this study, significant point source emissions were detected at a majority of landfills, many with emissions persisting over multiple revisits spanning several months, and in some cases, over multiple years. These results show the need for sustained measurements at landfills to provide operator guidance and better constrain emission variability.

Results:

Landfills surveyed between 2018-2022 across the U.S., as well as landfills surveyed between 2016-2017 during the California Methane Survey (*6*), are summarized in Figure 1a.

These surveys deployed either the Next-Generation Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG), operated by NASA-JPL, or the equivalent imaging spectrometer onboard the Global Airborne Observatory (GAO), operated by Arizona State University. Both spectrometers measure solar backscattered radiance from 380 - 2500 nm with 5 nm spectral sampling, enabling the estimation of atmospheric column CH_4 concentrations using a retrieval algorithm. We used the columnwise matched filter retrieval tuned to the CH_4 absorbing wavelengths between 2200-2400 nm (*15*), consistent with the California Methane Survey. Typical flight altitudes ranged between 3-5 km above ground level, resulting in CH_4 plume concentration maps with 3-5 m spatial resolution. Surveys were designed to require a minimum of three overpasses on different days (average 5.5) in order to provide a basic constraint on emission persistence (number of detections / number of overpasses) and variability. Information on emission quantification can be found in the Supporting Information (SI) *Materials and Methods* section.

The California Methane Survey performed a landscape assessment of 436 facilities across the waste sector in California, including active landfills, closed landfills, dry digestion facilities, and composting facilities (*13*). The study only detected large point source emissions at 32 of those facilities, though emissions from just those facilities made up a disproportionate sector contribution when compared to all aggregated sector emissions quantified in the survey (oil&gas, livestock, energy industries, wastewater treatment). Of those 32 facilities, 21 (66%) were open landfills that reported CH_4 emissions of at least 50,000 MtCO2e to the EPA Greenhouse Gas GHGRP and 12 (38%) reported at least 100,000 $MtCO_2e$ to EPA GHGRP. Given the observed prevalence of high point-source emission rates at large open landfills in California (i.e., landfills that accept large quantities of waste on an annual basis (*4*)) and in an effort to expand coverage across diverse climatic zones and jurisdictions, this study focused on similarly classified facilities based on emissions reported to the GHGRP, generally greater than 50,000 $MtCO_2e$. Although only a fraction of each state's waste facilities, together these facilities represent on average 36% (3.8-81% range) of each state's anticipated landfill emissions, according to GHGRP. The 18 states in this survey made up 67% of the U.S. municipal landfill emissions in the GHGRP (2019 reporting year). To our knowledge, this study represents the most systematic measurement-based study to date of methane point sources from the high emission solid waste sector, spanning 20% of approximately 1200 reported open landfills in the U.S.

We detected plumes at 52% of the landfills we surveyed (Figure 1b), far exceeding the point-source detection rate in other methane emission sectors. For example, airborne surveys in the California and the Permian Basin showed that around 0.2% and 1% of infrastructure had detectable plumes, respectively (*13, 16*). However, landfills are complex facilities with anticipated continuous emissions, and are fundamentally different than other anthropogenic emission sectors. For example, in the oil & gas sector, point sources detected by imaging spectrometers are usually clearly associated with intermittent but expected operations (maintenance, venting, and flaring) or fugitive emissions (leaks). The higher detection rate of point sources at large landfills confounds any clear separation between operational and anomalous emission behavior as some continuous CH_4 emissions are always to be expected at landfills. To underscore this point, Figure 1c shows the persistence (i.e., CH_4 detection frequency; persistence equals number of detections divided by number of overflights) of CH_4 at surveyed landfills compared to the persistence of oil & gas infrastructure in the Permian Basin (*16*). As evidenced by the US Environmental Protection Agency's recently finalized Super Emitter Program for oil and gas production and proposed changes to the Greenhouse Gas Reporting Program, high emission point sources (> 100 kg h^{-1}), regardless of persistence, are important to identify given their contribution to net emissions. Additionally, any such sources that persist over time may indicate anomalous behavior (e.g., leaks, malfunction) that warrant expedited repair. For the Permian, the average persistence for facilities with at least three overflights is 0.26, while for landfills, the persistence is a higher 0.60.

Related to persistence, we calculate the timescale or duration of point source activity for each landfill during its period of observation (*17*). This metric is calculated as the length of time that point sources were detected at a landfill divided by the length of time the landfill was observed. We find a bimodal distribution across landfills (Section S2), meaning there exists a population of landfills where point source activity only was observed for a short period of time and another distinct population of landfills where point source activity apparently persists across ~~the~~ nearly the entire observing record. This long-duration population represents more than 60% of all landfills, and 87% of all quantified emissions. These results highlight the distinct nature of point sources at landfills compared to other sectors. Within the oil & gas sector, point source duration timescales are also bimodally distributed, but long-duration sources make up a smaller fraction of all point sources (*17*), whereas the majority of landfills with point source detections show more persistent and long-lasting emission activity, highlighting the fundamentally different

activities, equipment, and dynamics of these sectors. In particular, mitigating persistent landfill sources potentially poses a greater climate benefit as they make up an outsized contribution of total emissions from that sector, and are more readily attributable and verifiable with on-site leak detection and repair protocols.

Point source CH_4 emissions at landfills may result from complex operational dynamics, including the constant movement of the active or working face, maintenance of the gas capture system, delays between waste burial and gas collection installation, construction of new waste cells, etc. There may also exist operational inefficiencies or exceptional circumstances that lead to emissions (e.g., poor maintenance of cover material, insufficient vacuum applied to wells, flooded wells, droughts creating cracks in cover). This dynamic environment where multiple factors could lead to point source emissions may explain the higher detection rate and persistence of CH_4 point sources. High resolution plume maps can aid in uncovering information about processes that lead to point sources. However, ground information from landfill inspections is also vital to connect observations to processes, as causes of emissions may be due to subsurface processes or small surface features that are difficult to discern even with high resolution aerial imagery. During these surveys, data was shared with several operators and feedback solicited regarding causes of detected point sources. Although we only received limited responses, many plumes were confirmed to be near the active working face, near compromised wellheads, or in areas where wells were being drilled. Sustained efforts are needed to connect detections and quantification to specific practices so that a better database of emission factors can be developed to help improve management practices and understand causes of landfill CH_4 emissions.

We observed many plumes where source attribution was much clearer when compared with high resolution visible imagery. These cases usually corresponded to easily distinguishable gas capture infrastructure. For example, we surveyed a landfill in the southern U.S. in May 2021, October 2021, May 2022, and June 2022 (Figure 2a-e). In this case, we observed exceptionally large (2,000-6,000 kg CH_4 h^{-1}) plumes emanating from multiple points across the face of the landfill at every airborne overpass. However, to the east of these massive plumes is a smaller, though still significant plume emanating from gas capture infrastructure. Figure 2b shows a closeup of this facility. The origin of the plume appears to be from an unlit flare or vent stack, and plumes are detected at every overpass between 2021-2022. Although the massive landfill plumes to the east are larger in magnitude, the persistent emissions from the unlit flare are still

concerning as these are emissions that are not expected with a functioning gas capture and control system – at minimum these excess emissions are normally expected to be flared instead of vented. To check if any flaring occurred at this site in between our overflights, we queried satellite fire detections using MODIS and VIIRS day and nighttime overpasses (*18*). The satellites did not discover any thermal signatures indicative of flaring in the vicinity of this site. When we take the average emission rate from all overpasses of this flare stack (1470 ± 720 kg CH_4 h^{-1}) and integrate across the approximate 12 months observations, the total emissions are 12,900 metric tons CH_4 or 322,000 MtCO2e. For reference, EPA GHGRP reports CH_4 emissions of 2,920,000 MtCO2e for this state's total landfill sector, so this single large point source is equivalent to 11% of that portion of the state's inventory. Therefore, as we continue to use observations to uncover process-level complexities at landfills, there exist a population of CH_4 mitigation candidates where timely repairs could have significant impact.

Emission estimates derived from imaging spectrometers via the Integrated Mass Enhancement (IME) method have been evaluated in multiple controlled release experiments and independent measurements (*19, 20*). However, given the aforementioned complexities with landfills, including topography, meteorology, and multiple plume origin locations, we performed extensive intercomparison with contemporaneous airborne surveys using Scientific Aviation (SA)'s mass balance approach (*20*). This measurement technique uses low altitude aircraft equipped with cavity-ring down spectrometers and a wind measurement system to conduct spiral surveys around a facility at various altitudes (generally 500-1500 m). The emission rate is calculated by applying Gauss's Theorem to observed concentrations and wind speeds. Unlike the imaging spectrometers used in this study that only detect strong point sources, SA measures the net emission flux from a landfill including the sum of diffuse area source fluxes and point source fluxes. Therefore, in comparing imaging spectrometer derived emissions to SA, we would expect the former to produce lower emission estimates than SA if at the time of overpass, there are significant contributions from area sources. However, if the net landfill flux at the time of overpass was dominated by strong point source emissions, we would expect SA and imaging spectrometer derived emissions to be comparable. SA generally requires a 30-40 minutes of spiral observations to quantify emissions from a facility the size of a landfill. This enables as many as 3-6 overpasses with the airborne imaging spectrometer. This approach was initially demonstrated with landfills in California in 2017 but only a small number involved simultaneous

over-flights and intercomparison of measurements separated by days to months were impacted by source variability (*13*). In this study, most of the intercomparison flights were conducted simultaneously as well as a few flights that occurred on the same day but separated by up to 2 hours.

Figure 3 shows a comparison between SA and GAO derived emission rates that passed quality control protocols at 15 landfill overpasses in several U.S. Midwest and Southern states (landfill names redacted). GAO emissions represent the average of all imaging spectrometer observations acquired during an SA observation window. The results are generally consistent (R^2 = 0.69; Figure S7). Figures 4b-c show a visual example of a landfill whose GAO derived emission rate was smaller than SA (Landfill 06), and an example where GAO and SA showed comparable emission rates (Landfill 12). In both cases, the CH_4 plumes observed by GAO corresponds closely with observed downwind high CH_4 concentrations observed by SA. The generally good agreement between GAO and SA builds confidence in the broader application of the remote sensing method to the larger population of landfills. As remote observations are increasingly being considered as contributors to routine CH_4 emission monitoring over large areas, confidence in emission quantification is essential for their adoption.

Figure 4a shows a comparison between the imaging spectrometer quantified average emission rates against emissions reported to the GHGRP for those facilities. In the U.S., landfills report emissions according to Code of Federal Regulations (40 CFR Part 98 Subpart HH); either by modeling generated emission from reported annual waste disposed (HH1/HH6), or, for landfills with gas capture and collection systems, from back-calculation based on reported annual gas captured and assumed collection efficiency (HH7/HH8). For landfills where multiple years of observations are available through Carbon Mapper flights, we take the average GHGRP across those years. Poor correlation exists between aerial emission rates and GHGRP (R^2 = 0.07), which could be expected under sparse sampling. However, even for landfills where we surveyed 10+ times (20 landfills total), we still find little agreement between emission estimates (R^2 = 0.02). This discrepancy appears equally in both directions – there exist a population of landfills (47% of all sites) whose aerial emissions are higher than GHGRP, and a population (53%) whose emissions are lower or did not show evidence of any point source emissions. On average, aerial emission rates were a factor 2.7 higher than GHGRP for all landfills, and a factor 1.4 higher for landfills with 10+ unique overpasses. Consistent with this study, independent assessments of U.S

emission inventories have indicated a needed 1.25-1.5 scaling of waste emissions to reconcile inventories with *in situ* ground-based measurements and coarse resolution satellite observations (*21, 22*). Furthermore, in some cases coarse resolution satellite instruments (e.g., TROPOMI) can quantify annualized emissions from individual landfills that are isolated from other emission sources (*22*). These annualized satellite observations show better correlation with our airborne datasets (15 landfills total) than GHGRP (details in SI Section S2). We also find no significant aggregate bias in airborne results from seasonal/diurnal barometric pressure variability (details in SI Section S3). Therefore, our airborne data, though not continuous are still in aggregate likely indicative of general trends of discrepancy with national inventories.

Figure 4b shows yearly averaged aerial emission estimates from two landfills with 5+ years of aerial sampling and whose trends in emission rates are significant ($p < 0.05$) based on an ordinary least squares fit to the data. For both of these landfills, EPA GHGRP indicates an insignificant trend in emission rates. In the case of Landfill 17 (LF 17), airborne observations also suggest at least a factor of 2.5 underestimate compared to GHGRP. There could be some significant operational issues at that landfill leading to larger than predicted emissions. Figure 4b therefore shows an example of how one could use top-down information as a check against reporting or enforcement protocols. When a significant number of atmospheric observations sustained over many months to years shows persistent discrepancies and diverging trends with bottom-up process-based emission estimates, it highlights areas for attention and action. Additionally, when coupled with nimble application of emerging onsite emission assessment approaches, a single methane plume image with sufficient clarity could trigger expedited response to guide follow-up root-cause analysis, improve general practices, and potentially reduce emissions.

Landfill emissions are comprised of some fraction of spatially dispersed area and localized point sources, but a typical ratio of area to point source emissions for a given landfill in most cases remains unknown and may vary with site operations and environmental conditions. These conditions may affect emission pathways and gas collection system efficacy in complex ways. The comparison of GAO and SA suggests that for landfills with detectable point sources, these emissions may make up an outsized contribution against the total CH_4 contribution. Airborne and satellite remote sensing observations can provide some initial indications regarding the distribution of physical emission types for landfills through relatively frequent, wide area

monitoring of high emission point sources. In a tiered observing strategy, the remote sensing data can be combined with net facility emission estimates from mass balance aircraft (Figure 3) and more wide-spread deployment of continuous surface monitoring to quantify the contribution of point sources relative to the net landfill emission flux and also better understand variability.

Discussion:

There exist at least two use-cases for plume-scale remote sensing of landfill CH_4 point sources. The first is quick detection and precise geolocation of emission hotspots at a landfill. After communicating with some facility operators, we attributed a handful of our detected plumes to specific operations (e.g., working face, well drilling, construction). More effort is needed to connect the detected emission hotspots to operations to better understand the carbon impact of certain management practices and to help guide operators to areas on landfills where remediation may be needed. The EPA Inspector General issued a report in 2020 finding that EPA needs to improve oversight of how states implement air emissions regulations for municipal solid waste landfills (*14*). Much more attention towards Clean Air Act Compliance is anticipated due to ongoing landfill inspections by both federal and state government and the interest in methane reduction.

The second use-case is quantifying emission rates to support evaluation of emission factors used in reporting programs and inventories. At this stage, we find a large discrepancy and generally poor correlation between EPA GHGRP bottom-up emission estimates and what we observed from airborne platforms. This discrepancy may be partially explained by sampling, however there could also be systematic issues with the models that underpin reporting programs. Ultimately, informed comparison of emission rates derived from atmospheric measurements with bottom-up calculations requires an improved understanding of the site processes that these airborne platforms detect. However, regardless of root cause, the detection rate and persistence of point source emissions at landfills and the large magnitude of aerial emission rates found in this study points to potential gaps in landfill models and/or calculation of emissions reported to the GHGRP.

Reconciling top-down and bottom-up estimates requires improved accounting for potential point source emissions in inventories in the context of current regulatory structures. Unplanned emissions such as unlit flares or large leaks from gas collection fields (particularly ones detectable by SEM) may not be reflected in inventory estimates. Planned maintenance

activities of limited temporal extent could be represented at some level. Working face emission potential is largely unknown. In the future, *in situ* measurements and new site metadata, such as changes in time-resolved gas collection and maintenance event temporal tracking, would complement the top-down data, improving inventories and reducing unnecessary emissions. Additional multi-sensor field studies are needed to conclusively determine whether there are systematic biases in landfill emission models used in methane inventories.

Airborne remote sensing platforms have proven extremely valuable for initial surveys and baseline assessments of CH_4 emissions across multiple sectors. However, the sustained sampling of landfills recommended by this study require systems where routine observation is logistically more feasible. Especially outside the U.S., where many waste sites in developing countries lack any form of management or monitoring, emissions may be disproportionately large when compared to other sectors and bottom-up models. As countries move towards incorporating best management practices for waste, using atmospheric measurements that can be deployed at scale to verify emission reductions will be critical. Satellites could provide a solution to sampling when they are configured or capable of scanning large areas frequently and have sufficient sensitivity to point source emissions (*24*). Preliminary studies with the TROPOMI, GHGSat, and EMIT satellite instruments have identified large point sources at a small subset of global waste sites, many which lack management practices geared towards reducing CH_4 emissions (*25, 26*). The Carbon Mapper Coalition plans to launch two Planet Tanager satellites in 2024, optimized for CH_4 and carbon dioxide point source monitoring from space and builds on advances from NASA's EMIT mission (*27*). This system will provide wide area coverage and frequent sampling to quantify methane emissions from a large population of managed and unmanaged waste sites around the world. Satellites offer the ability to monitor methane emissions from landfills and unmanaged dumps across regions that are largely inaccessible due to workforce and resource limitations. Satellites also offer more complete coverage than aircraft in many regions given high costs, logistics and airspace restrictions. Although not a complete solution to waste emission quantification, the ability to quantify and precisely geolocate point source emissions routinely at global scale with a combination of these remote sensing platforms represents an important contribution to this sector. This information – combined with multi-scale information from a tiered observing system - can be effective in accelerating mitigation if efforts to connect observations with operators and regulators are sustained.

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Supplementary Materials

Materials and Methods
Supplementary Text
Figs. S1-S8
Carbon Mapper Emission Data File (XLSX)

Scientific Aviation Data File (ZIP Archive)

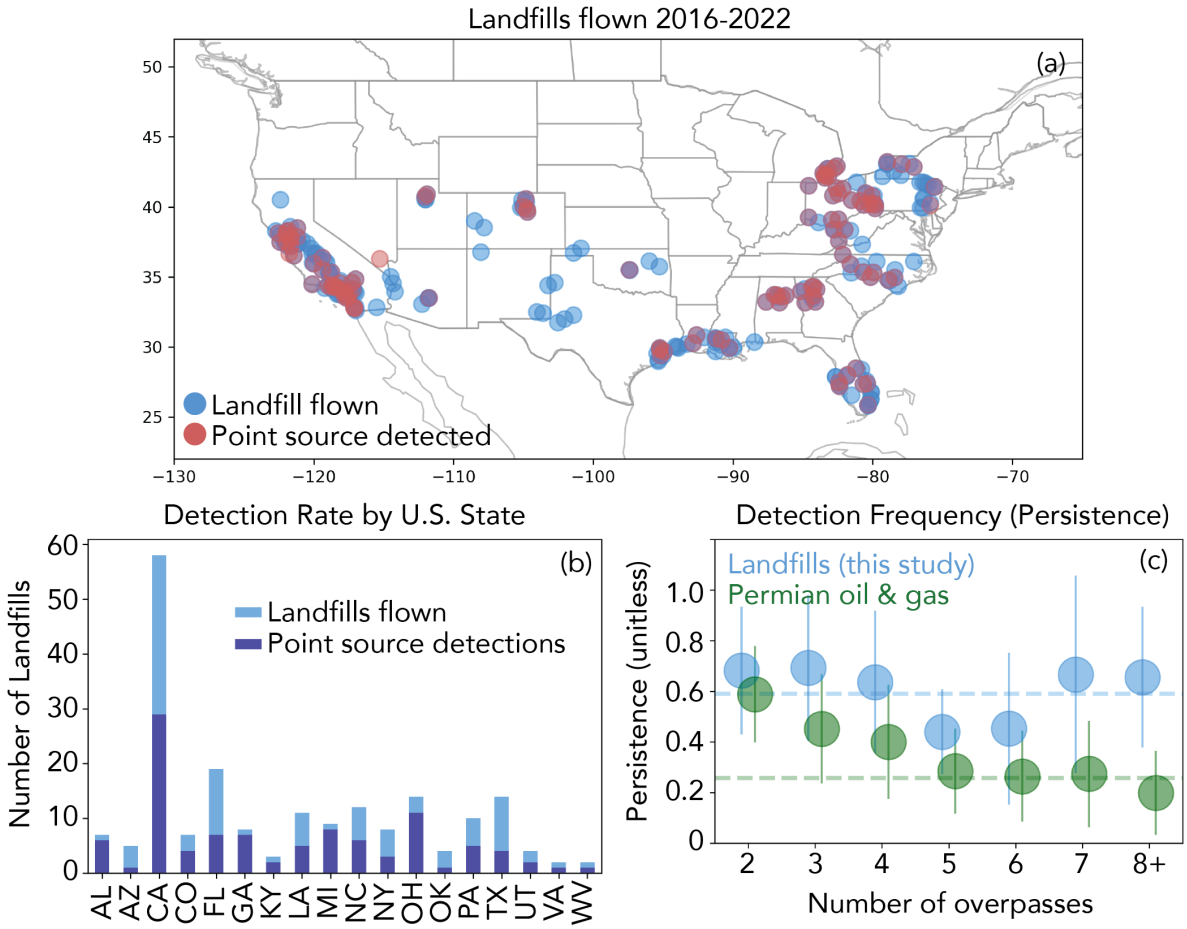

Fig. 1. Landfills flown between 2016-2022 using the Next-Generation Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG) or the Global Airborne Observatory (GAO). Panel (a) shows the spatial extent of U.S. surveys: blue dots represent all landfills with fly overs and red dots represent landfills where point sources were detected on at least one overpass. Panel (b) shows the total number of large (>20,000 MtCO2e reported to GHGRP) landfills surveyed by state (light blue) and the number of landfills where we detected point sources in at least one overpass (dark blue). Panel (c) shows average and standard deviation of detection frequency, also known as persistence (number of detections / number of overpasses), across all surveyed landfills as a function of number of overpasses. Dashed lines represent the average persistence for facilities flown at least three times on three different days.

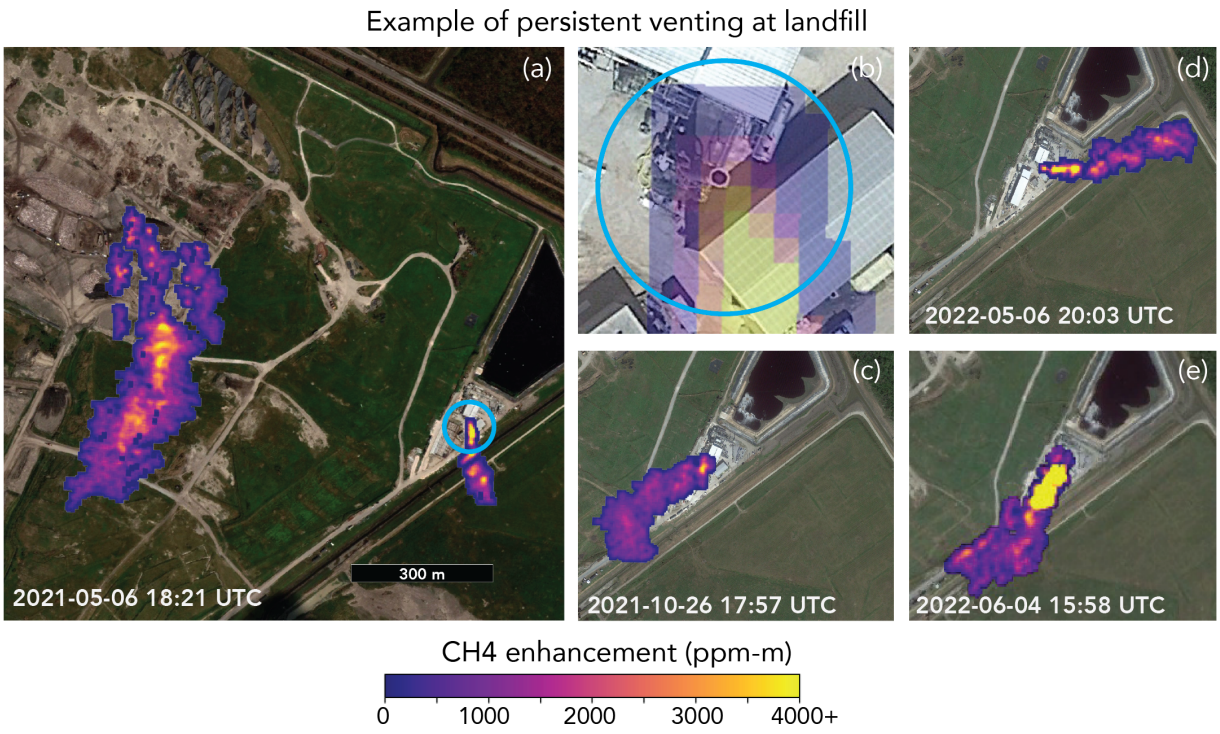

Fig. 2. Example of multiple persistent point sources at a landfill. Panel (a) shows at least two plumes detected from a single overflight in May 2021. The plume emanating from the blue circled region potentially corresponds to a vent or unlit flare (Panel (b)). Emission from this vent persisted across all other overflights between May 2021 – June 2022 (Panels (c-e)). Visible basemaps are provided by Google Earth.

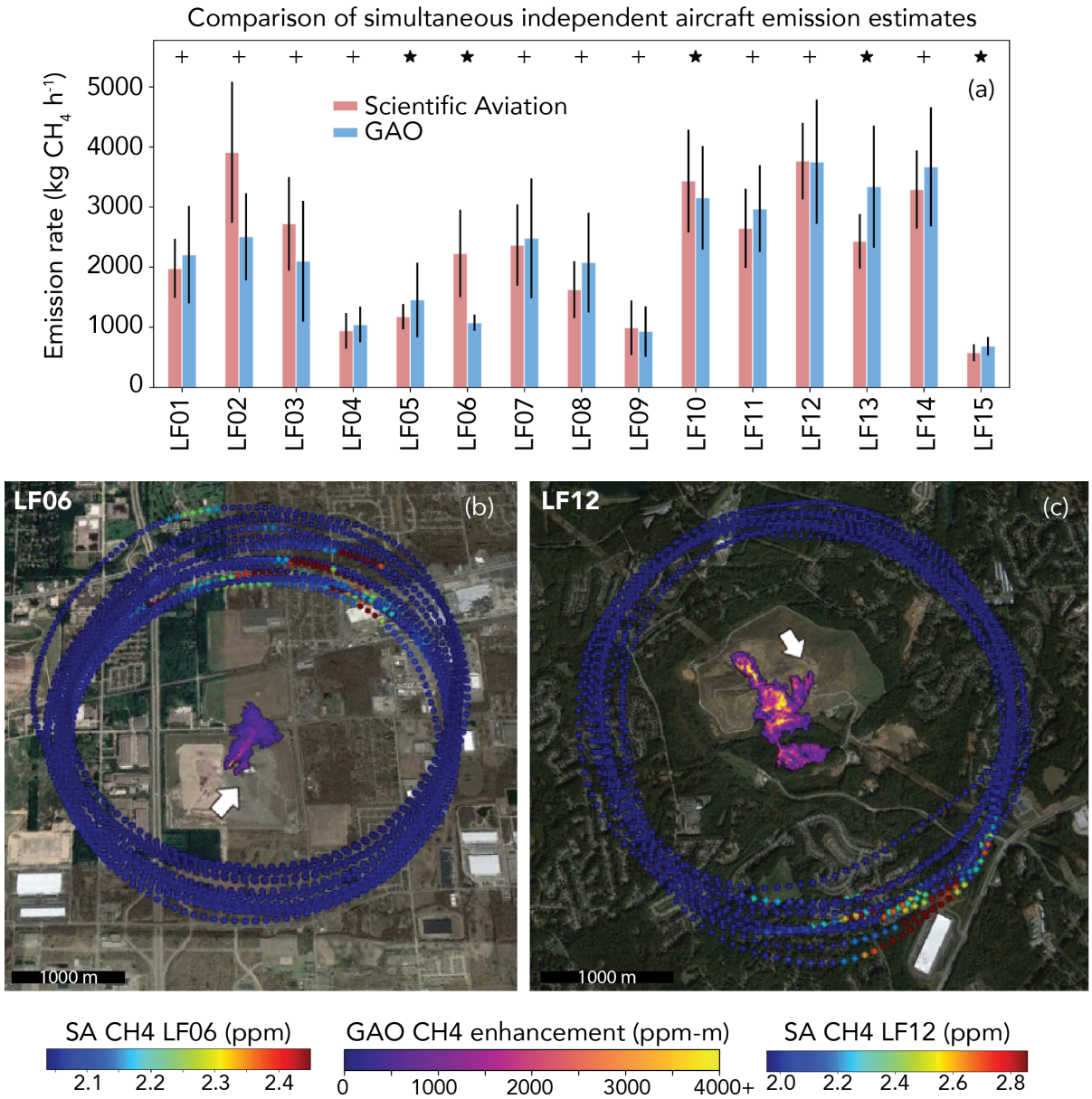

Fig. 3. Comparison of emission rates at landfills derived using the Scientific Aviation mass-balance approach and the GAO imaging spectrometer. Panel (a) shows 15 landfill overpasses where comparison between emissions were possible. The (+) symbol indicates simultaneous data acquisition. The (*) symbol indicates asynchronous, but same day observations within 2 hours of one another. Error bars represent 1 standard deviation uncertainties on emission rates. Panels (b) and (c) show methane observations at LF06 and LF12, respectively, from Panel (a). The white arrow indicates wind direction.

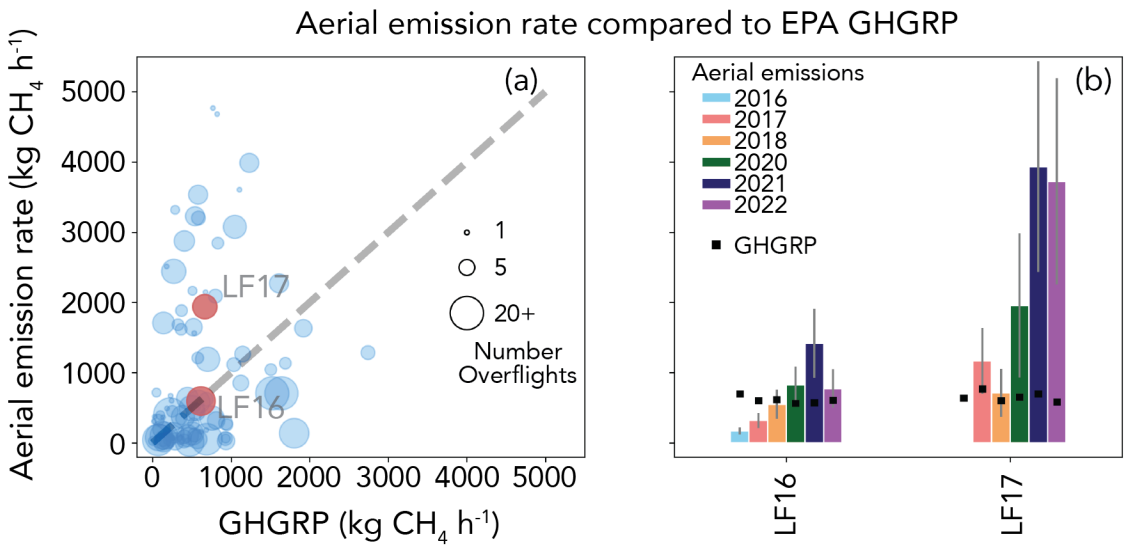

Fig. 4. Comparison of aerial emission rates to EPA GHGRP for landfills where point sources were detected at least once. Panel A shows mean CH_4 emission rates across all aerial overpasses compared to average GHGRP emissions. The grey line represents the one-to-one line. The size of the dots corresponds to number of overflights. Two red colored dots in Panel (a) correspond to landfills with 5+ years of observations whose observed emission trends are significant ($p < 0.05$). The trends for these landfills are shown in Panel (b). Black squares in Panel (b) represent GHGRP reporting for that year.. Error bars represent 1 standard deviation uncertainties on emission rates.

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Supplementary Materials for

Quantifying Methane Emissions from United States Landfills

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The PDF file includes:

Materials and Methods
Supplementary Text
Figs. S1-S8

Other Supplementary Materials for this manuscript include the following:

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Materials and Methods

With plume-mapping imaging spectrometers like AVIRIS-NG and GAO, detection of CH_4 plumes and quantification require separate quality control protocols. Figure S1 shows three landfill examples with CH_4 concentrations overlay high resolution non-contemporaneous visible imagery from Google Earth. Landfill A shows a clear CH_4 plume that travels along mean wind flow, according to the High-Resolution Rapid Refresh (HRRR; *30*) hourly reanalysis. The strong CH_4 gradient between plume and background and the clear plume shape makes the scene a strong candidate for both plume detection and quantification. Landfill B also shows strong CH_4 enhancements above the background, but no clear plume origin is discernible, and likely many pixels are corrupted by retrieval artifacts or false positives due to confusing surface materials (*31*). In this case, enough credible CH_4 enhancements exist to be confident in strong CH_4 activity at that landfill, but the unstable plume shape and prevalence of noise preclude reliable emission estimations. Landfill C shows a third landfill where no strong CH_4 enhancements are present: enhanced pixels are likely the result of very small CH_4 sources or artifacts, so this landfill would be deemed not suitable for point source detection or quantification. For each identified CH_4 plume, an analyst consults CH_4 concentration imagery and high-resolution imagery to mark their best approximation of a plume's origin. Plume imagery and concentration maps are available for surveyed landfills (*28, 29*).

For landfill plumes that pass quality control screening for emission estimation, we implement the integrated methane enhancement (IME) method that was deployed during the California Methane Survey (*13*). This approach quantifies the excess CH_4 mass that was generated by a plume (kg CH_4), then normalizes by the length of the plume (m), and scales by the HRRR 10-m reanalysis wind speed (m h^{-1}) to estimate an emission rate (kg CH_4 h^{-1}). Uncertainty in emission rates is calculated in quadrature by quantifying the spatiotemporal variability in HRRR 10-m winds and by quantifying variability in IME by varying the maximum length of the plume (*13*). Emission rates (Q) are therefore calculated using the following equation:

$$Q = \left(\frac{\overline{IME}}{r_c}\right) u_{10} \quad (S1)$$

Where r_c is the average radial spatial extent of the plume and u_{10}, 10-m wind speed, comes from the NOAA High-Resolution Rapid Refresh (HRRR) meteorological product, which in forecast mode is available hourly at 3 km resolution.

Uncertainty quantification for emission rates derived from imaging spectrometers follows approaches described previous surveys (*13, 16*), and is driven by uncertainty in wind speeds and IME. For an uncertainty assessment, we compute the average wind speed from 27 HRRR grid cells – the 9 grid cells closest to each plume, for 3 time steps (plume detection time plus or minus 1 hour). The uncertainty σ_U is then quantified as the standard deviation among these 27 grid cells. The uncertainty in IME ($\sigma_{IME/rc}$) comes from calculating the standard deviation of the $\frac{\overline{IME}}{r_c}$ ratio for several radial extents (maximum 300-m from origin of the plume). For an uncertainty assessment on emission rate (σ_Q), we combine the uncertainty from wind speed and IME to r_c ratio in quadrature:

$$\sigma_Q = Q\left(\left(\frac{\sigma_{IME/rc}}{IME/r_c}\right)^2 + \left(\frac{\sigma_U}{u_{10}}\right)^2\right)^{1/2} \quad (S2)$$

There can be cases where more than two plumes may be identified at different points on a landfill in a single airborne overpass. In these cases, it may not be clear if these multiple plumes emanate from a single emission event or multiple events. To estimate total landfill point source emissions, we average emission rates of multiple plumes whose estimated origin coordinates are within 500-m of one other. For plumes that exceed this distance threshold, we sum plume emission rates.

Emission rate quantification has been validated against several blinded and unblinded controlled release studies. These studies have shown the airborne imaging spectrometer technology presented here to in aggregate represent unbiased emission rates and detect plumes as low as 10 kg CH_4 h^{-1} (*32, 33*).

Supplementary Text

Timescales of point source emissions

Timescale, or duration, of a point source emissions was quantified in previous analysis of airborne data (*17*), and is defined for a facility as the number of days elapsed between the first and last point source detection divided by the number of days elapsed between the first and last date of observation of the facility (i.e., observation period). Figure S2 shows the distribution of the observation period for all landfills in this study where there were at least 2 days of observation. 51% of all landfills were observed with at least 180 days of spacing. Figure S3 shows the distribution of timescales for all landfills with at least two days of observation. 62% of landfills exceed timescales of over 50% of its observation period, and constitute 87% of all quantified emissions. Figure S4 shows the timescale distribution for landfills whose observation period was at least 100 days. Consistent with Figure S3, 66% of landfill exceed timescales of 50% of its observation period, and constitute 85% of total emissions.

Comparison with TROPOMI Inversions

Nesser et al. (*22*) estimated the U.S. national CH4 budget for 2019 through a flux inversion from TROPOMI satellite observations. This was done by inverting observed CH4 concentrations to emission fluxes through the use of an atmospheric transport model and a regularizing prior. The nadir pixel resolution of TROPOMI is 5.5×7 km^2 and the resolution of flux estimates provided by (*22*) is $0.25° \times 0.3125°$. Given the spatial resolution of TROPOMI and the inverse fluxes estimated by the study, generally emissions from individual waste sites can not readily obtained or isolated from other emission sources. However, in cases where landfills are spatially isolated from other emission sectors and the information content of the inversion (called degrees of freedom for signal) suggests strong information coming from the satellite observations as opposed to the prior, (*22*) reported facility-scale landfill emissions. Of these landfills, 15 overlap spatially with the landfills we observed in this study, however none overlap with the time period of the inversion (2019). Still, we perform a comparison between emissions reported by (*22*), and our airborne campaigns (Figure S5). Given uncertainties in both estimates, we report parity using a reduced major axis regression fit. We find correlation between the two estimates with only some small bias ($y = 1.2x - 290$; $R = 0.40$). However, when we compare (*22*) TROPOMI flux estimates against GHGRP reported values for 2019 (Figure S6), we find less correlation ($y = 0.79 – 228$; $R = 0.12$) . Therefore, we find that although airborne and TROPOMI estimates were derived by totally independent observing and quantification systems and performed during different time periods, airborne emissions (even under more sparse temporal sampling) are more predictive of annualized emission estimates than GHGRP.

Barometric pressure sensitivity

Studies have shown some landfill CH_4 emission rates to be sensitive to fluctuations in barometric pressure. In comparing aerially derived emission rates to annualized emission rates, like GHGRP, we test whether we see significant bias between annually averaged barometric pressure compared to the time of aerial sampling. To do this, we query HRRR sea level pressure at each landfill site at the time of aerial plume detection for landfills with 5+ days of observation. We then randomly sample hundreds of HRRR sea level pressure values for the same landfills, but across the full seasonal and diurnal time range for the same year the landfill was surveyed. We then compare the mean sea level pressure at the time of observation to the mean barometric pressure from this annualized sample. Figure S8 show the results, summarized in terms of relative difference and absolute difference. We find the bias to be near zero (0.08 hPa), though absolute bias ranges from -5.7 hPa to 5.4 hPa. In context of previous study, Czepiel et al. (*10*) constrained an emissions-pressure relationship at a landfill in Nashua, New Hampshire, USA and found an empirical relationship of Emissions = -1.2(Pressure) + 1231.9, with emission units being in m^3 CH_4 min^{-1} and pressure in hPa. Though emissions-pressure relationships are landfill dependent, taking this relationship for example, a 6 hPa sampling bias (e.g., aircraft flew when surface pressure was 6 hPa lower than its annual average, and average pressure at time of observation was 1010 hPa), could result in 36% emission bias. However, given that Figures S8 show even distribution centered around zero, we find that likely the results from these surveys are not overly biased in aggregate due to barometric pressure. Future study with more in situ barometric pressure sampling will be useful to continue to probe the effect of pressure on emissions across the broad diversity of landfills.

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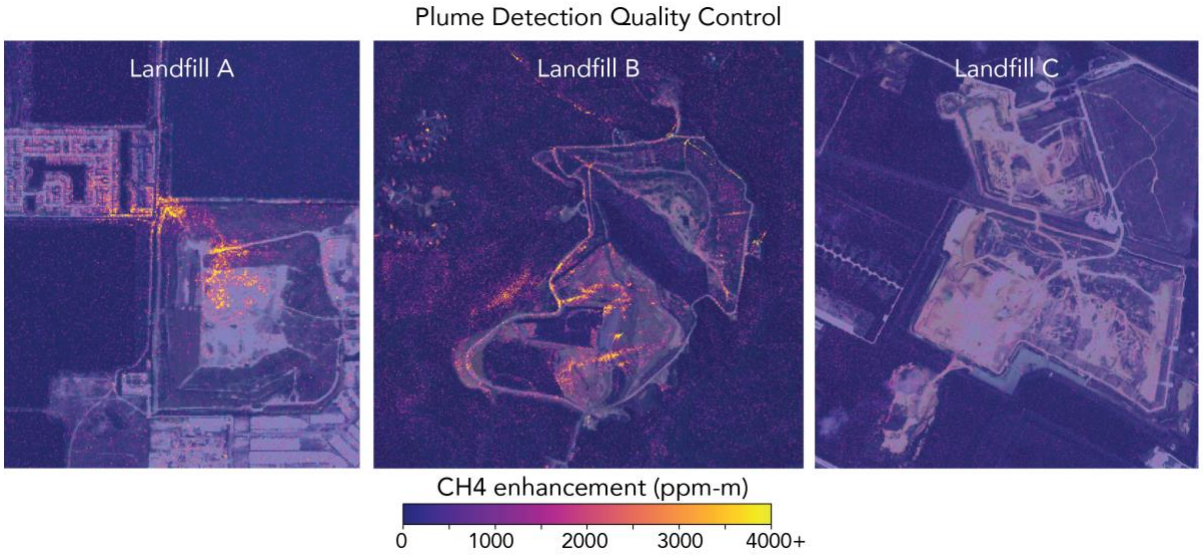

Fig. S1. **Three example landfill observations to illustrate the quality control process.** Landfill A shows a landfill where strong point source behavior is detected with little noise corruption and clear plume delineation (passes detection and quantification quality control). Landfill B shows an example where credible CH_4 behavior is observed along with severe noise artifacts, and no clear plume morphology is observed (passes detection but fails quantification quality control). Landfill C shows an example where no credible CH_4 point source behavior is detected.

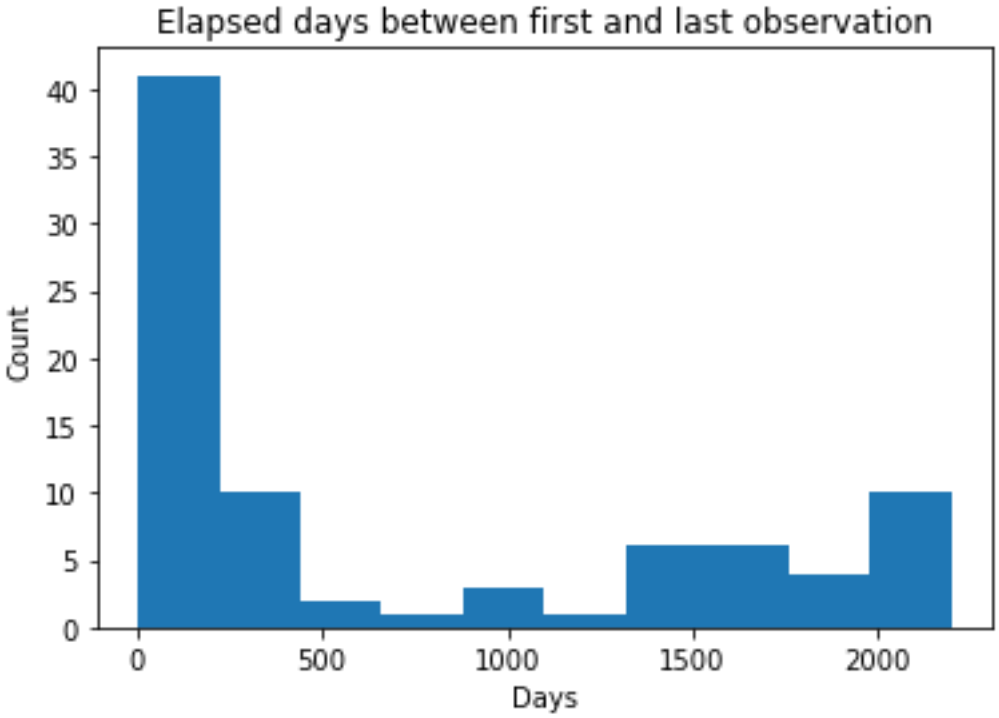

Fig. S2. Distribution of observation periods (number of days in between first landfill observation and last landfill observation) for GAO/AVIRIS-NG airborne campaigns.

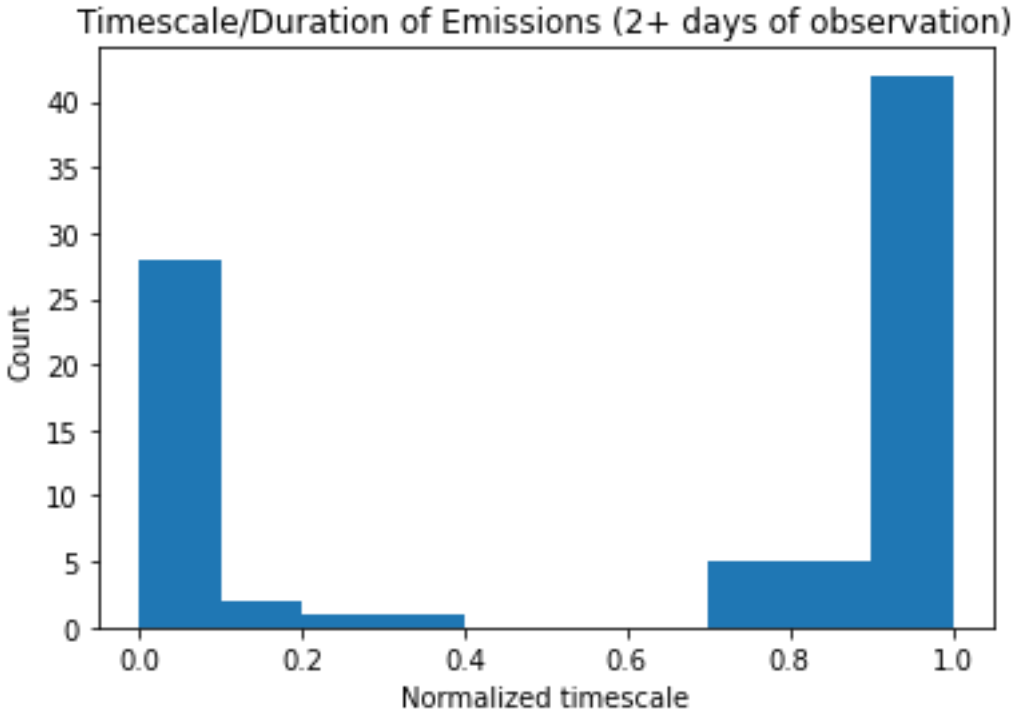

Fig. S3. **Normalized timescale or duration of landfill point source emissions.** This is defined as the time elapsed between the first and last point source detection at a landfill divided by the observation period (number of days in between first landfill observation and last landfill observation).

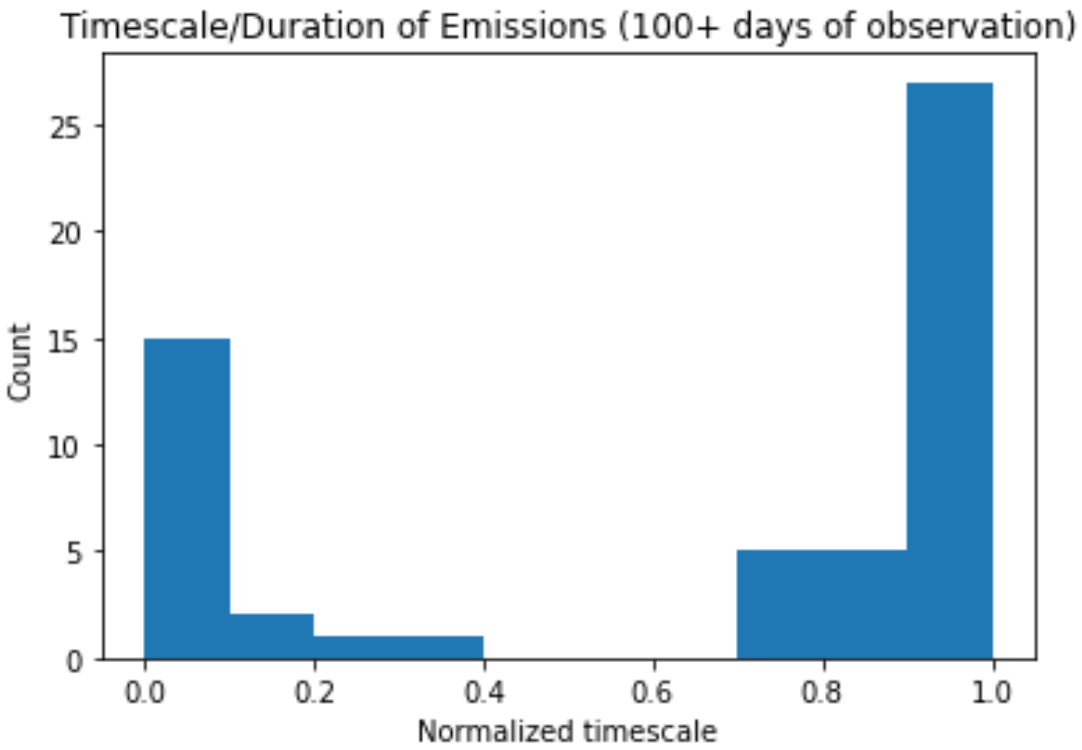

Fig. S4. **Normalized timescale or duration of landfill point source emissions with observation periods of at least 100 days.**

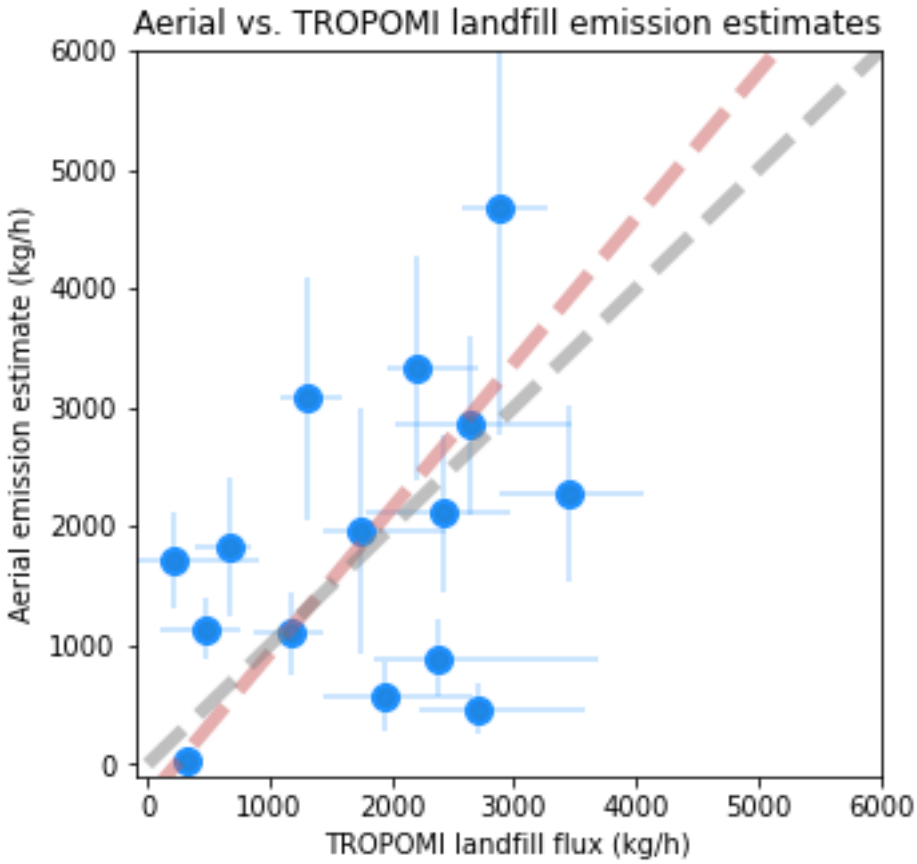

Fig. S5. Comparison between spatially overlapping aerial and TROPOMI-dervied emission estimates at select landfills.

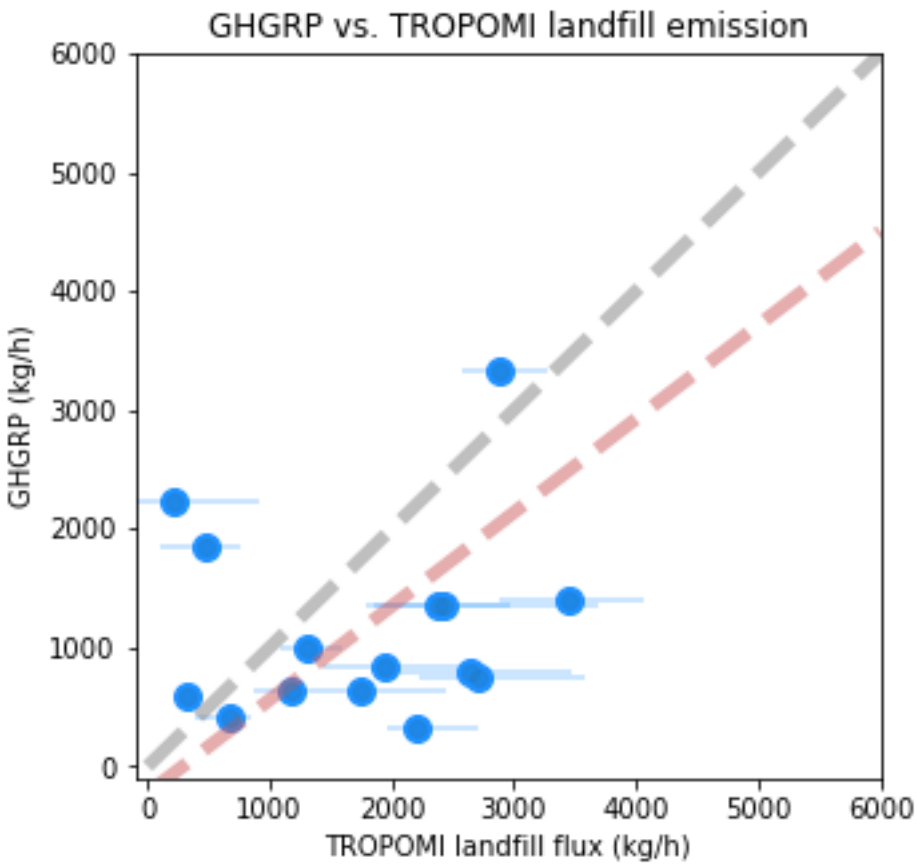

Fig. S6. Comparison between GHGRP reported and TROPOMI-dervied emission estimates at select landfills.

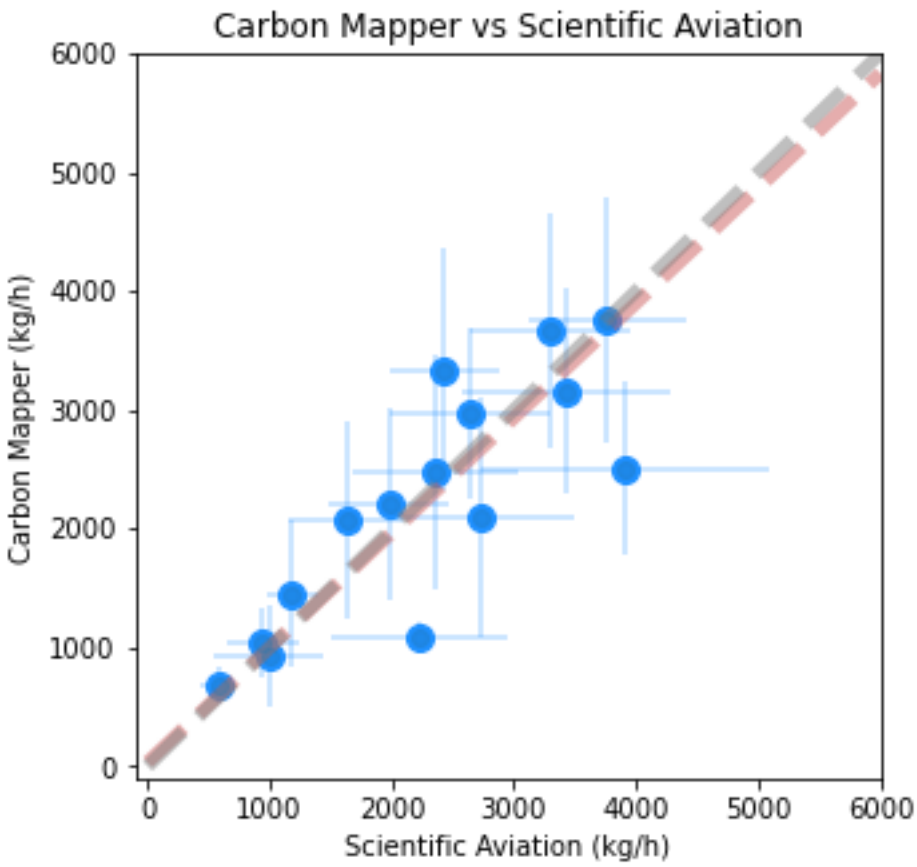

Fig. S7. **Parity plot of Carbon Mapper and Scientific Aviation simultaneous overflights as described in the main manuscript.** The grey dashed line is the 1-1 line, and the red dashed line represents a reduced major axis (RMA) regression fit. The RMA fit produces the following fit: y= 0.96x + 34; R = 0.83.

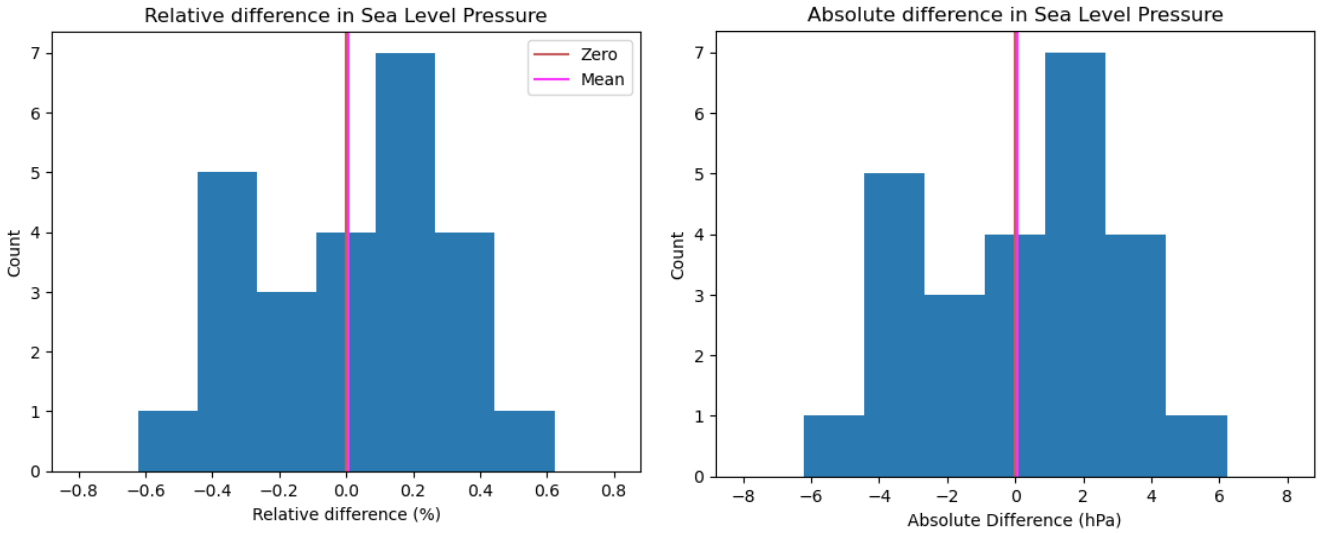

Fig. S8. **Difference in sea level pressure at time of observation compared to annual average for landfills with 5+ days of observation.** The left panel shows the relative difference and the right panels shows the absolute difference. Here, the HRRR reanalysis was sampled and averaged at the time of airborne overpass and compared to average sea level pressure for that year of observation.