Exhibit A

Article

California's methane super-emitters

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Methane is a powerful greenhouse gas and is targeted for emissions mitigation by the US state of California and other jurisdictions worldwide[1,2]. Unique opportunities for mitigation are presented by point-source emitters—surface features or infrastructure components that are typically less than 10 metres in diameter and emit plumes of highly concentrated methane[3]. However, data on point-source emissions are sparse and typically lack sufficient spatial and temporal resolution to guide their mitigation and to accurately assess their magnitude[4]. Here we survey more than 272,000 infrastructure elements in California using an airborne imaging spectrometer that can rapidly map methane plumes[5–7]. We conduct five campaigns over several months from 2016 to 2018, spanning the oil and gas, manure-management and waste-management sectors, resulting in the detection, geolocation and quantification of emissions from 564 strong methane point sources. Our remote sensing approach enables the rapid and repeated assessment of large areas at high spatial resolution for a poorly characterized population of methane emitters that often appear intermittently and stochastically. We estimate net methane point-source emissions in California to be 0.618 teragrams per year (95 per cent confidence interval 0.523–0.725), equivalent to 34–46 per cent of the state's methane inventory[8] for 2016. Methane 'super-emitter' activity occurs in every sector surveyed, with 10 per cent of point sources contributing roughly 60 per cent of point-source emissions—consistent with a study of the US Four Corners region that had a different sectoral mix[9]. The largest methane emitters in California are a subset of landfills, which exhibit persistent anomalous activity. Methane point-source emissions in California are dominated by landfills (41 per cent), followed by dairies (26 per cent) and the oil and gas sector (26 per cent). Our data have enabled the identification of the 0.2 per cent of California's infrastructure that is responsible for these emissions. Sharing these data with collaborating infrastructure operators has led to the mitigation of anomalous methane-emission activity[10].

Methane (CH_4) is being increasingly prioritized for near-term climate action, given its relatively short atmospheric lifetime and the potential for rapid, focused mitigation that can complement economy-wide efforts to reduce carbon dioxide emissions. In California, efforts to mitigate methane emissions are complicated by large inconsistencies between estimates of emissions derived from atmospheric measurements and from greenhouse-gas inventories: past studies using atmospheric measurements report methane emissions that are higher than those from inventories, both statewide[11–13] and in key regions and sectors[14,15]. Other studies indicate that methane emissions from the oil and gas supply chain are about 60% higher than those reported in the national greenhouse-gas inventory[16] and that there is a heavy-tail distribution of methane-emission sources in the US natural gas supply chain, where typically fewer than 20% of sources (so-called super-emitters) contribute more than 60% of total emissions from that sector[17]. Scientists and policymakers have emphasized the rapid identification and mitigation of methane super-emitters, particularly those due to leaks and abnormal operating conditions[18].

In addition to California, there remain large uncertainties regarding the distribution of methane emissions in other key regions and emission sectors globally[19]. There is a dearth of available observational studies of sectors such as livestock manure management and landfills, both of which are predicted to be larger contributors to California's methane budget than the oil and gas sector[8]. In addition, spatially sparse and infrequent field studies can overestimate or underestimate important methane sources that are intermittent or highly unpredictable. Finally, the relative contributions of methane point sources and area sources have not been well studied in California. We define 'point source' as a

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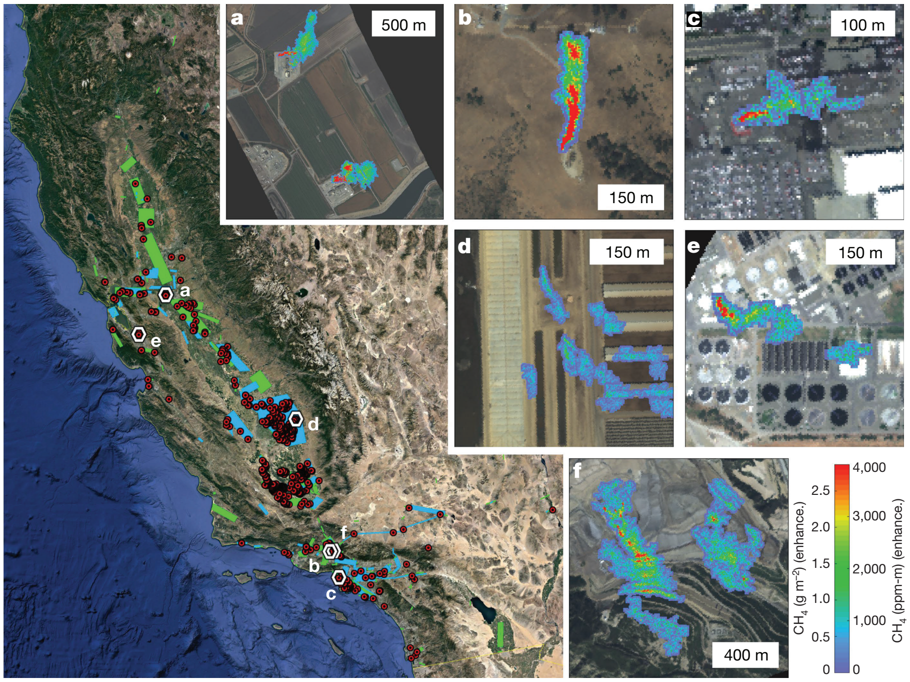

Fig. 1 | Images from our survey of methane point emissions in California. Main image, approximately 2,000 individual AVIRIS-NG flight lines from 2016 (blue) and 2017 (green) covered more than 272,000 individual facilities and infrastructure elements. Detected sources are indicated by red points, with the densest clusters seen in the San Joaquin Valley (dairies and oil fields). The inset images show examples of representative methane plumes from different sectors: **a**, compressor stations at a natural gas storage facility; **b**, oil well; **c**, tank of liquefied natural gas; **d**, dairy manure management; **e**, wastewater-treatment plant; **f**, landfill. The colour scales indicate the methane concentration enhancement (the mass of methane in a plume relative to background air) in each pixel in units of parts per million-metre (ppm-m). Inset images are from AVIRIS-NG. The basemap image is from Google Earth, Lamont-Doherty Earth Observatory (LDEO)-Columbia, National Science Foundation (NSF), National Oceanic and and Atmospheric Administration (NOAA), Landsat/Copernicus, Scripps Institution of Oceanography (SIO), US Navy, General Bathymetric Chart of the Oceans (GEBCO).

condensed surface feature or infrastructure component of less than 10 m in diameter that emits plumes of highly concentrated methane. This contrasts with an ‘area source’, or the combined effect of many small emitters distributed over a large area (typically 1–100 km across) that releases methane in a more diffuse fashion; area sources include anaerobic decomposition from rice cultivation and enteric fermentation from ruminant animals, both of which are better addressed with other measurement methods and are not included here.

The California Methane Survey was designed to provide the first systematic survey of methane point sources across the state, with a focus on detecting, geolocating and quantifying super-emitters. This survey fills an important gap in scale, and complements other observational systems that provide aggregate constraints on emissions from regions and area sources[20–22] and short-duration field campaigns that are limited to a small number of facilities[23,24]. The survey was conducted with the Next Generation Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG). AVIRIS-NG measures ground-reflected solar radiation at wavelengths from 380 nm to 2,510 nm with 5-nm spectral sampling, and has a 1.8-km field of view and 3-m pixel resolution at typical survey altitudes of 3 km (ref. [5]). This class of instrument is unique in terms of its high signal-to-noise ratio, calibration accuracy and response uniformity[25]. The methane retrieval is based on absorption spectroscopy[6,7,26] and can reliably detect and quantify methane point sources with emissions typically as small as 2–10 kg CH_4 h^{-1} for typical surface winds of 5 m s^{-1}, depending on surface brightness and aircraft altitude and ground speed. See the Supplementary Information for a detailed description of datasets, estimation methods and validation.

The spatial and sectoral scope of this survey comprised key methane point-source emission sectors in California, including: oil and gas production, processing, transmission, storage and distribution; refineries; dairy manure management; landfills and composting facilities; wastewater-treatment plants; gas-fired power plants; and liquified and compressed natural gas facilities. Multiple overflights were conducted for the same infrastructure over several years to assess source persistence.

AVIRIS-NG flights for this study were conducted during five campaigns: August to November 2016, March 2017, June 2017, August to November 2017, and September to October 2018. The survey imaged approximately 59,000 km^2, including revisits (Fig. 1). The survey was designed to cover at least 60% of methane point-source infrastructure in California, guided by a Geographic Information System (GIS) dataset known as Vista-CA (see Supplementary Information). Approximately 272,000 infrastructure elements were covered by the survey, most of which were observed multiple times. The survey included more than 200,000 oil and gas wells and related production infrastructure, representing a sample size more than 500 times larger than previous point-source persistence studies[27].

The AVIRIS-NG flights conducted during this survey detected 1,181 individual methane plumes; for each plume we estimated the enhancement (the mass of methane in the plume relative to background air) and attributed it to a Vista-CA infrastructure element (Fig. 1). Average emission rates and 1σ uncertainties were estimated for 564 distinct sources at 250 facilities, using observed methane enhancements and surface wind speed data from weather reanalysis products. The sum of our measured source emissions is 0.511 Tg CH_4 yr^{-1} and we apply a non-parametric bootstrap analysis to the population of observed sources to calculate a 95% confidence interval of 0.433–0.601 Tg CH_4 yr^{-1}. The population has a heavy-tail distribution, indicating that 10% of the point sources are responsible for 60% of the detected point-source emissions (Fig. 2 and Supplementary Information), spanning every sector surveyed.

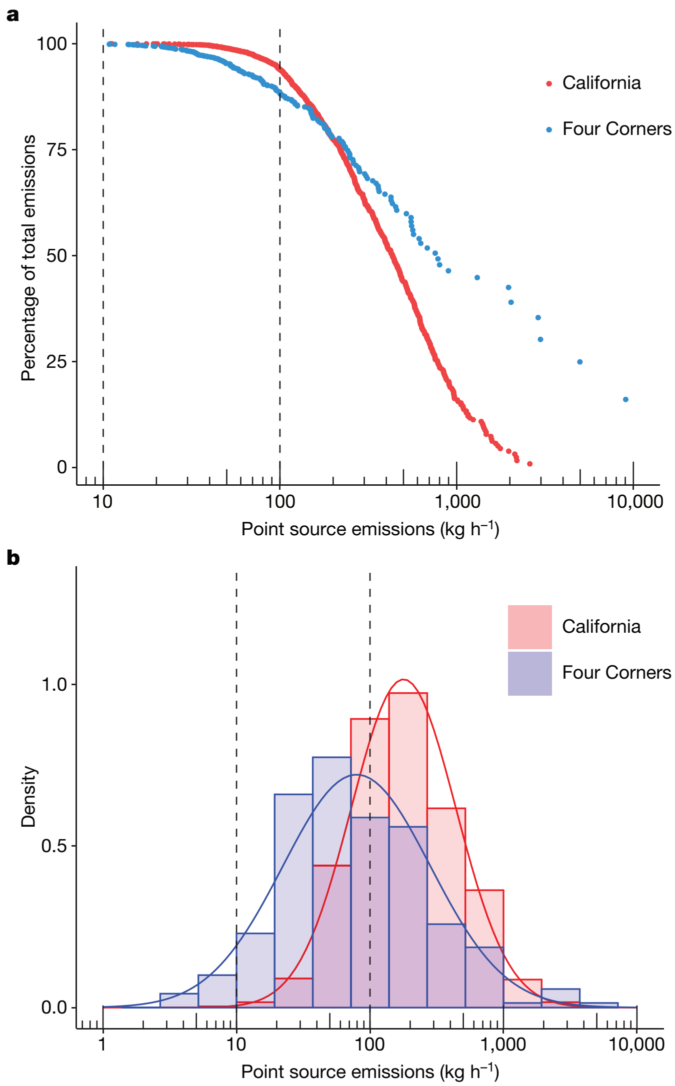

Fig. 2 | The distribution of point-source emissions is consistent between two different regions. a, Data from 564 methane point sources for all sectors in California (red; this study) and from 250 coal, oil and gas sources from the Four Corners region (blue[9]). The numbers for California have not been adjusted for persistence here, as this was not possible for the brief Four Corners study. The heavy-tail distribution indicates that 10% of the point sources are responsible for 60% of the detected point-source emissions. **b**, Histogram showing the density of point-source emissions with lognormal fits. Note that the Four Corners region includes some large emitters associated with coal production that do not occur in California. The vertical dashed lines indicate typical detection limits for this class of infrared imaging spectrometer, ranging from 2–10 kg CH_4 h^{-1} for the typical 3-km flight altitudes used in this study to 100 kg CH_4 h^{-1} for an equivalent satellite in low Earth orbit.

The repetitive, high-spatial-resolution plume imagery enabled us to characterize point-source behaviour and controlling processes, particularly for sectors that have not been as well studied as the oil and gas production sector. Many of the sources were highly intermittent, with a median persistence of 0.20 for the entire population (mean 0.33, range 0.02–1.0). In some cases, the intermittent emissions can be explained by normal operations (for example, periodic waste flushing at large dairies). In other cases, more persistent activity is apparently due to sustained venting at a small number of anaerobic digesters at dairies and wastewater-treatment plants, or to leaking bypass valves at natural gas compressor stations. We find a similar distribution of persistence (20–35% on average) and emissions in the manure-management, wastewater-treatment and oil and gas sectors. Solid-waste management is the largest methane point-source emission sector in California (Table 1), with persistent plumes observed at only 32 of 436 surveyed landfills and composting facilities. Our imaging of landfills identified methane plumes associated with construction, gaps in intermediate cover and leaking gas-capture wells—indicating a subpopulation of anomalous emitters (see Supplementary Information). The fact that we did not detect a larger population of smaller methane point sources across the landfill sector suggests that most of those facilities emit methane as area sources that cannot be detected with this method.

Given that we surveyed a large fraction (32–100%) of every point-source emission sector in California, we can upscale our measurements to estimate statewide point-source emissions, resulting in a total of 0.618 (95% confidence interval 0.523–0.725) Tg CH_4 yr^{-1}—equivalent to 34–46% of the California Air Resources Board (CARB) methane inventory[8] for 2016. We find that solid-waste management contributes 41% of observed point-source emissions, followed by 26% from manure management and 26% from oil and gas (contrasting with the 32%, 39% and 25% of total methane emissions found for these sectors in the CARB inventory[8]). We estimate that upstream oil and gas production contributes about 79% of the total oil and gas methane point-source emissions in California. Spatially, 85% of point-source emissions from upstream production are concentrated in the southern San Joaquin Valley (the highest oil- and associated-gas-producing region in the state), 14% in Los Angeles and Ventura counties, and 1% in the Sacramento Valley. We emphasize that the relative contribution of emission sectors probably varies in other regions around the world owing to regional differences in economic activity, age of infrastructure, and regulation. We also highlight that there are no doubt regional differences in the relative sectoral contributions of area sources (such as urban gas-distribution systems) that are beyond the scope of this study.

In addition to solid-waste management, other emission sectors may be greatly underestimated in the CARB inventory. When comparing our estimates of point-source emissions for those sectors in the CARB inventory most likely to include methane point sources, our sectoral estimates account for about 38% of the CARB inventory's emissions from the wastewater-treatment sector, about 42% of emissions from the manure-management sector, and about 366% of the CARB inventory for the energy industries sector. The latter is probably associated with most refineries and a small number of high-emitting power plants (see Supplementary Information). Large discrepancies are observed between many of the self-reported emissions from participating facilities and the AVIRIS-NG and independent airborne estimates (Fig. 3 and Supplementary Information). Moreover, our survey of point-source emissions in California and the US Environmental Protection Agency (EPA)'s Greenhouse Gas Reporting Program (GHGRP) for the entire US[28] are in agreement that 99% of point-source emissions come from facilities that emit at least 25 kg h^{-1} (see Supplementary Information). This is notable given that manure management and oil and gas production contribute more than half of the point-source emissions in our study, but are mostly not included in the GHGRP for California and are only partially represented in the total US GHGRP.

We shared preliminary findings from our surveys—including images of methane plumes—with collaborating facility operators, who provided verification with surface observations and/or explained the mechanisms underlying the observed emissions and persistence. Many of these collaborative efforts led directly to mitigation of the methane sources detected in the survey. For example, we discovered four cases of leaking natural gas distribution lines and one leaking liquified natural gas storage tank (Fig. 1), which the operators confirmed, repaired, and requested verification of repair by follow-up AVIRIS-NG flights[10].

The prevalence of methane super-emitter activity in multiple sectors in California suggests substantial potential for mitigation. We have found that 30 facilities could be responsible for around 20% of the 2016 CARB methane inventory, including many that exhibit large

Table 1 | Point-source emissions by sector

IPCC source category	Vista-CA infrastructure element	Number of Vista-CA infrastructure elements	Number of surveyed elements	Percentage surveyed	Sectoral scalar	Number of sources detected	Measured emissions ($Tg\ CH_4\ yr^{-1}$)	State total emissions ($Tg\ CH_4\ yr^{-1}$)	State total 95% confidence intervals ($Tg\ CH_4\ yr^{-1}$)	Percentage of total emissions
1A1 Energy industries	Gas-fired power plants	435	238	55	1.83	7	0.007	0.013	0.007, 0.021	2.1
	Refineries	26	26	100	1.00	37	0.015	0.015	0.008, 0.023	2.4
	Subtotals	**461**	**264**	**57**	**1.27**	**44**	**0.022**	**0.028**	**0.015, 0.044**	**4.6**
1B2 Oil and natural gas	CNG/LNG fuelling stations	208	132	63	1.58	6	0.002	0.003	0.003, 0.004	0.5
	Natural gas stations (non-storage compressor, metering, etc)	1,131	538	48	2.10	5	0.005	0.010	0.009, 0.012	1.6
	Natural gas pipeline (transmission, distribution)	216,774	68,548	32	3.16	5	0.004	0.012	0.010, 0.014	1.9
	Natural gas processing plants	26	23	88	1.13	5	0.004	0.004	0.004, 0.005	0.7
	Natural gas storage fields	12	12	100	1.00	11	0.009	0.009	0.008, 0.010	1.4
	Oil and gas: wells	225,766	198,231	88	1.14	107	0.048	0.054	0.046, 0.063	8.8
	Oil and gas: other production equipment	3,356	2,872	86	1.00	120	0.066	0.066	0.056, 0.076	10.7
	Subtotals	**447,273**	**270,356**	**60**	**1.16**	**259**	**0.137**	**0.158**	**0.135, 0.184**	**25.6**
3A2 Manure management	Dairy confined animal feeding operations	620	443	71	1.40	215	0.115	0.161	0.137, 0.187	26.1
4A1 Managed waste disposal	Landfills and composting facilities	1,146	436	38	1.11	32	0.229	0.255	0.175, 0.345	41.3
4D1, 4D2 Wastewater treatment and discharge	Domestic and industrial wastewater treatment	148	57	39	2.60	12	0.004	0.012	0.005, 0.020	1.9
	Industrial wastewater treatment: beef processing	NA	NA	NA	1.00	2	0.004	0.004	0.004, 0.005	0.6
	Totals	**449,648**	**271,556**	**60**	**1.21**	**564**	**0.511**	**0.618**	**0.523, 0.725**	**100.0**

The table summarizes the persistence (frequency)-adjusted point-source emissions found in this study according to sectors identified by the Intergovernmental Panel on Climate Change (IPCC), as well as estimated total emissions derived with population scalars. Most of the scalars are simply the ratio of the number of infrastructure elements identified by Vista-CA to the number of surveyed elements, with three exceptions (oil and gas: other production equipment; landfills and composting facilities; and industrial wastewater treatment), for which we further constrain or eliminate scaling. See Supplementary Information section 2 for details.

discrepancies between reported and measured emissions (see Fig. 3 and Supplementary). Our survey in California and a previous study of the Four Corners region in the US[9] exhibit consistent heavy-tail distributions of methane point-source emissions (Fig. 2) despite the different sectoral mixes for the two regions (the Four Corners emissions are associated primarily with oil, gas and coal production[9]). If similar distributions of methane point-source emissions occur in other key regions around the world, this could translate to as much as 8–11% of global greenhouse-gas forcing, assuming a 100-year warming potential of 32 and 350 $Tg\ CH_4\ yr^{-1}$ of total anthropogenic methane emissions for 2016 (refs. [19,29]). Testing this hypothesis would require additional aircraft surveys and satellite observations that can provide the necessary combination of high spatial resolution, sensitivity and wide area coverage for other key regions globally. Those broader studies would also improve our understanding of waste and manure-management emissions, which, as in California, might dominate the emission budgets of other regions[19].

Detection limits for methane point sources could be relaxed by a factor of ten compared with the survey described here and still identify 90% of super-emitters if applied frequently over large areas that have emission distributions similar to those of California (Fig. 2). Because detection scales linearly with spatial resolution[30], mature technologies such as that used here could be deployed for more efficient point-source monitoring across larger regions on high-altitude aircraft and satellites. Our high-performance infrared imaging spectroscopy would translate to a robust detection limit of 100 $kg\ CH_4\ h^{-1}$ for a satellite in low Earth orbit, depending on spatial resolution (assuming a wind speed of 5 $m\ s^{-1}$). Widespread and sustained deployment of point-source remote sensing methods such as ours, when combined with near-continuous regional monitoring of distributed area sources by surface observations and other satellites, could greatly advance scientific understanding of methane budgets and efforts to manage them. Complete closure of the methane budget and effective mitigation will no doubt require a multi-tiered observational strategy, in which the methods demonstrated here could play a key part.

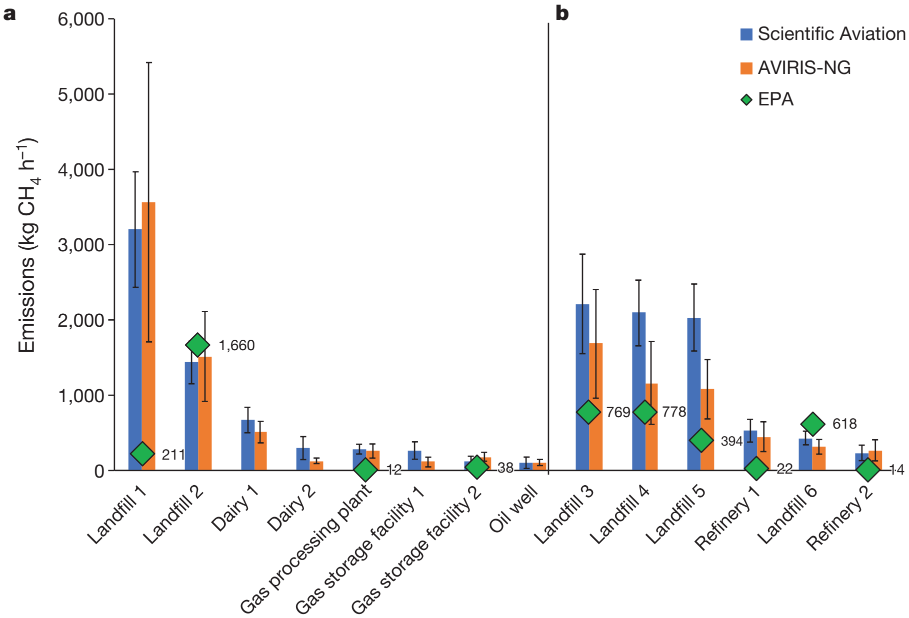

Fig. 3 | Independent airborne measurements of emissions from representative facilities on the basis of simultaneous flights or several visits. a, Simultaneous flights; **b**, average emissions from multiple non-simultaneous flights over several months. Orange bars show AVIRIS-NG estimates of point-source emissions, and blue bars show estimates by Scientific Aviation (Boulder, CO, USA) of facility net emissions[31]. Error bars indicate one standard deviation. AVIRIS-NG estimates are lower than Scientific Aviation estimates for facilities that have some non-point-source activity. The 14 estimates here correlate with an R^2 of 0.86 (see Supplementary Information). The R^2 for the eight facilities in **a** is 0.99. The estimated total emissions here are 11,228 ± 4,981 kg h^{-1} (AVIRIS-NG) and 13,900 ± 3,593 kg h^{-1} (Scientific Aviation). Green diamonds indicate available self-reported emissions[28].

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-019-1720-3.

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Data availability

Radiance and reflectance products calibrated by AVIRIS-NG can be ordered from the AVIRIS-NG data portal at https://avirisng.jpl.nasa.gov/alt_locator/. Retrieved methane images from flight lines in this study are available for download at https://doi.org/10.3334/ORNLDAAC/1727. Vista-CA infrastructure spatial layers are available for download at https://doi.org/10.3334/ORNLDAAC/1726. Images of methane plumes, Vista-CA layers and regional-scale methane-emission products for California can be viewed at https://methane.jpl.nasa.gov/. Tables of methane plume and source characteristics are provided in the Supplementary Information.

Code availability

The custom computer code or algorithms used to generate the results in this study can be made available to researchers upon request.

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Author contributions R.M.D., A.K.T., F.M.H. and C.E.M conceived the study. R.M.D., A.K.T., F.M.H., T.R., I.B.M., M.L.E. and S.C. conducted flight planning. Each author contributed to the collection, analysis or assessment of one or more datasets necessary to perform this study. R.M.D., A.K.T., K.T.F., F.M.H. and T.R performed the analysis with contributions from B.D.B., D.R.T., C.F., N.K.C., M.F., J.D.H, B.E.C., R.O.G. and V.Y. R.M.D., A.K.T., K.T.F. and F.M.H. wrote the manuscript with input from all authors.

Competing interests The authors declare no competing interests.

Additional information

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