Comprehensive evaluation of aircraft-based methane sensing for greenhouse gas mitigation

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This manuscript is a non-peer reviewed preprint submitted to EarthArXiv

Abstract

Methane is a major contributor to anthropogenic greenhouse gas emissions. Identifying large sources of methane, particularly from the oil and gas sector, will be essential for mitigating climate change. Aircraft-based methane sensing platforms can rapidly detect and quantify methane point-source emissions across large geographic regions, playing an increasingly important role in industrial methane management and greenhouse gas inventory. We independently evaluate the performance of five major methane-sensing aircraft platforms: Carbon Mapper, GHGSat-AV, Kairos Aerospace, MethaneAIR, and Scientific Aviation. Over a 6-week period, we released metered gas for over 700 single-blind measurements across all five platforms to evaluate their ability to detect and quantify emissions that range from 1 kg(CH_4)/hr to over 1,500 kg(CH₄)/hr. Aircraft consistently quantified releases above 10 kg(CH₄)/hr, and GHGSat-AV and Kairos Aerospace detected emissions below 5 kg(CH₄)/hr. Fully-blinded quantification estimates for platforms using spectroscopy-based measurements have parity slopes ranging from 0.76 to 1.13, with R² values of 0.61 to 0.93; the platform using an in situ measurement approach has a parity slope of 0.5 ($R^2 = 0.93$). Results demonstrate aircraft-based methane sensing has matured since previous studies and is ready for an increasingly important role in environmental policy and regulation.

1 1 Introduction

2

Methane is a potent greenhouse gas with over 80 times the global warming potential of carbon
dioxide over a 20-year timespan¹. With a short atmospheric lifetime, methane shapes near-term
climate outcomes, making it a priority for climate change mitigation efforts. Top anthropogenic
methane sources and targets for emissions reductions are the oil and gas sector, waste

- 7 management, and agriculture 2 .
- 8

9 Aircraft-based methane sensing enables rapid and widespread assessment of methane emissions.

10 In the last several years, aerial surveys have identified methane leaks several-fold larger than

11 those reported in greenhouse gas inventories or found using conventional ground-based surveys

12 ^{3–8}. Sherwin et al. find that in multiple oil and gas producing regions across the United States,

aerially detected emissions from roughly 1% of sites constitute 50-80% of total methane

14 emissions from oil and gas production, processing, and transportation infrastructure, highlighting

15 the prospect of massive emissions reductions through aerial surveys ⁸. Following these technical

advances, US Environmental Protection Agency has proposed new rules that, if adopted, would
 allow companies to use remote sensing technologies, including aircraft, to comply with

allow companies to use remote sensing technologies, including aircraft, to comply
 emissions monitoring and reduction efforts at oil and gas production sites ⁹.

19

20 Methane-sensing aircraft typically use one of two approaches for quantifying methane emissions:

21 infrared spectroscopy and in situ methods. Spectroscopy uses the differential absorption of

22 infrared (IR) light by methane compared to other atmospheric gases. Imaging is most commonly

23 passive, relying on reflected sunlight as a radiation source, and thus requiring favorable weather

- 24 conditions. An alternative approach is active spectroscopy LiDAR system, in which a laser
- 25 mounted within the aircraft sends a radiation signal that is reflected and used in analysis ¹⁰. For

- 26 the in-situ approaches, an aircraft measures atmospheric concentrations of methane in real time
- 27 during the flight, and emission magnitude is quantified using models that combine multiple
- 28 concentration measurements with flight altitude and distance from the target ¹¹. While time-
- 29 intensive compared to imaging, in situ approaches allow for analysis of other air pollutants
- 30 alongside methane, including carbon dioxide, nitric oxide, and nitrogen dioxide ¹².
- 31
- 32 As companies and governments increasingly rely on aircraft methane management, accurately
- assessing these technologies' capabilities becomes increasingly important. Here, we report
- 34 independent, single-blind evaluation of five different aircraft operators. We examine their ability
- to identify high-volume methane emissions from a point source. Four operators use passive IR
- 36 spectroscopy: Carbon Mapper, GHGSat-AV, Kairos Aerospace, and MethaneAIR. We also test
- 37 Scientific Aviation, which uses an in situ measurement approach.
- 38
- 39 Prior studies have evaluated the performance of aircraft-based methane detection and
- 40 quantification. Carbon Mapper, GHGSat-AV, Kairos Aerospace, and MethaneAIR participated
- 41 in previous Stanford led singe-blind controlled release experiments ^{10,13,14}. These operators
- 42 sought additional validation based for new testing configurations or modifications informed by
- 43 their previous results. While not included in the present study, Bridger Photonics' Gas Mapping
- LiDAR has been independently tested elsewhere in single-blind and location-blind studies ^{10,15,16}.
- 46 This study fills important gaps in the previous literature. In particular, this is the first
- 47 independent single-blind test of Scientific Aviation and MethaneAIR (Chulakadabba et al., 2023
- 48 ¹⁴ used a collaborative technology validation experimental design). In addition, the Kairos
- 49 Aerospace and GHGSat-AV systems presented here represent a significant advance over those
- 50 tested previously. Finally, this is the first single-blind evaluation of a field-realistic deployment
- of the Carbon Mapper system, as the previous Stanford test was conducted with shorter
- 52 flightlines than used in field deployment, resulting in artificially low quantification estimates
- 10,17 . As a result, this work provides the most definitive assessment to date of the five tested
- 54 airborne methane sensing systems, which represent the majority of currently deployed
- 55 technology systems in this space.

56 2 Methods

57

We conducted aircraft testing from October 10th through November 11th, 2022 in Casa Grande 58 59 (Arizona) as part of a 2-month experiment that also tested satellites and ground sensors. For inter-comparison purposes, we use established experimental and data reporting protocols ^{10,13}. 60 Briefly, the Stanford field team releases a fixed stream of methane at a constant rate while an 61 aircraft operator conducts measurements. We maintain strict blinding protocols: operators are not 62 63 informed whether a release is being conducted or not. Participants are provided the coordinates 64 of gas release in advance, and asked to mimic standard field operations as closely as possible in both data collection and analysis. Additional information describing data collection is provided 65 66 in Supplementary Information Section S1.1. 67

68 2.1 Methane Controlled Releases Equipment

- Gas is released from a trailer parked at a fixed location [32.8218489°, -111.7857599°]. The 70
- 71 trailer is equipped with high-precision meters and two stacks that release gas at 7.3 meters (24
- 72 feet) and 3.0 meters (10 feet) above ground level. We refer to these as the tall and short stacks,
- 73 respectively. The methane source for all experiments was compressed natural gas (CNG), stored onsite in two trailers provided by Rawhide Leasing and refilled from Arizona-based CNG 74
- 75 providers as needed. Gas was transferred from the CNG trailers to a pressure regulation trailer
- 76 (Rawhide Leasing, RT-30), and then to the gas metering trailer, as depicted in Figure 1.
- 77



Figure 1: Experimental field setup top view (left) and on-the-ground (right). Methane supply is from compressed natural gas trailers (depicted in the left image only). Gas pressure is reduced in a pressure regulation trailer, then delivered to a metering 81 and release trailer. Wind data is collected using a 3-D sonic anemometer mounted on a 10-meter wind tower. Stanford set 82 desired flow rates from the workstation. Also visible in the image but not labelled are ground sensor that were deployed during 83 testing.

84 Upon entering the metering and release trailer, gas is diverted through one of three parallel flow paths based on the desired release rate. The three flow paths are designed to release flow rates of 85 1-30 kg gas/hour (kg/h), 30-300 kg/h, and 300-2.000 kg/h, and are each fitted with an 86 Emerson Micromotion Coriolis meter sized accordingly. The Stanford team used a laptop to 87 remotely set the flow rate from the field workstation (additional details on flow control in SI 88

- 89 Section S1.1.3.1).
- 90
- 91 2.1.1 Safety
- 92

We established a 45-meter (150 ft) safety perimeter around the gas release point, and no Stanford 93 94 personnel were allowed within this perimeter while gas was flowing. Experienced and safety-95 certified gas contractors (Rawhide Leasing) operated the gas release equipment, and Stanford 96 team regularly monitored the plume with an infrared camera (FLIR GF320) to ensure methane 97 remained far from all onsite personnel. The team also remained vigilant for olfactory signals of methane.

98 99

Description of Aircraft-Based Technologies Tested 100 2.2

- 101
- We tested five different aircraft-based methane-measurement technologies: Carbon Mapper, 102
- 103 GHGSat-AV, Kairos Aerospace, MethaneAIR, and Scientific Aviation. Details of each platform

- are included in the supplementary material. Briefly, Carbon Mapper, GHGSat-AV, Kairos
- 105 Aerospace and MethaneAIR all use passive infrared spectroscopy. Carbon Mapper, GHGSat-
- 106 AV, and Kairos Aerospace conduct surveys that identify and quantify large-scale methane point
- source emissions, particularly from oil and gas (examples include but are not limited to: ^{3,4,18}).
 MethaneAIR, the aircraft pre-cursor to MethaneSAT, is designed for wider spatial coverage and
- MethaneAIR, the aircraft pre-cursor to MethaneSAT, is designed for wider spatial coverage and measuring diffuse sources in addition to point source ¹⁴. Scientific Aviation uses a in situ
- 10 measurement technique, conducting multiple consecutive loops around the target methane source
- 111 while collecting ambient air samples ¹¹. Methane measurements are conducted onboard using a
- 112 Picarro 2210-m instrument that measures methane, ethane, carbon dioxide and water. All five
- 113 aircraft operate at different altitudes and implemented different flight patterns during testing (see
- 114 Table 1). Hence, the time necessary to conduct a single measurement varies across operators, as
- does the total number of measurements feasible in one day.
- 116 117

	<u>Carbon</u> Manner	<u>GHGSat-AV</u>	<u>Kairos</u> Aerospace	MethaneAIR	<u>Scientific</u> Aviation
Testing dates (Month/Day format)	10/10 - 10/12, 10/28-10/29, 10/31	10/31, 11/02, 11/04, 11/07	10/24 – 10/ 28	10/25, 10/29	11/08, 11/10, 11/11
Range of flight height above target (meters or feet above ground level) ⁱ	3,050 - 3,230 meters (10,000 - 10, 600 ft)	1,930 – 2,080 meters (6,320 – 6,840 ft)	370 - 540 meters (1,210 - 1,770 ft)	12, 690 – 13,610 meters (41, 620 – 44, 670 ft)	N/A
Average measurement frequency ⁱⁱ	12 min	4 min	3 min	22 min	21 min
Wind Reanalysis Data Source for Fully Blinded Submission ⁱⁱⁱ	HRRR	GEOS-FP	Dark Sky	DI method: HRRR; mIME method: HRRR/LES	N/A

118

^{III} Wind reanalysis data source abbreviations: HRRR = High Resolution Rapid Refresh (provided by US National Oceanic & Atmospheric Administration); GEOS-FP = Goddard Earth Observing System Forward Processing (provided by US National Aeronautic and Space Administration); For MethaneAIR, LES refers to 1-way coupled Large Eddy Simulation.

119 120

121 2.3 Field Data Collection Procedures

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124 consistency and comparability with other testing results. Briefly, operators were asked to recreate

 ⁱ Flight altitude for the 1-minute leading up to measurement timestamp. Measurement timestamp refers to the moment when the aircraft distance from the release target was at a minimum, using GPS coordinates.
 ⁱⁱ For imaging technologies, this is the average time between individual measurement timestamps across all flight days for a given aircraft. The measurement time itself is instantaneous, and differences in measurement frequency reflect operator specific flight patterns. For Scientific Aviation, measurement frequency represents the average time for conducting one complete measurement.

¹²³ Field measurement protocols were based on those previously reported ^{10,13,14} to maintain

125 typical flight operations and submitted measurement frequency, planned flight lines, altitude, and

126 predicted lower detection limit in advance. For spectroscopy-based platforms, we held a constant

- 127 release rate while the aircraft passed overhead. The Stanford ground-team tracked the GPS
- 128 location of each aircraft, aiming to change the release rate at least two minutes before the aircraft 129 next passed overhead. For Scientific Aviation, we set a measurement schedule in advance, and
- hext passed overhead. For Scientific Aviation, we set a measurement schedule in advance, and
 held a constant release rate for 35-40 minutes. Details on field data collection are included
- 131 supplementary materials Section 1.3.
- 132
- **133** 2.4 Data collection and filtering

We collected raw 1 Hz flow measurement data from all three Coriolis meters, and data cleaning
is described fully in supplemental materials Section 1.2. To convert whole gas flow rate to
methane, we use gas compositional data provided by the upstream supplier of the CNG station
from which we purchased natural gas (additional details in supplemental materials section

139 1.2.3.). Mean mol% CH₄ over the study period is 94.53% and the standard deviation is 0.62%.140

- 141 Wind conditions varied widely through the testing period. Aircraft operators reported observing 142 stagnant methane from previous releases pooling around the site in some conditions. To ensure each new measurement occurred with a clean background, we developed a wind-based filtering 143 144 criteria for spectroscopy-based operators, which excludes measurements where it is likely that a significant residual signal from the previous measurement might be present. A full description is 145 included in SI section 1.3.5.1. For Scientific Aviation, we excluded any measurements where the 146 147 standard deviation over the measurement period was greater than 10% of the mean flow rate for 148 the same period.
- 149

150 2.5 Operator Data Collection and Reporting

151

152 We use the multi-stage unblinding and data reporting procedures described in Rutherford et al., 2023. In Stage 1 of data reporting, all operators submit fully blinded quantification estimates. 153 These Stage 1 data are therefore most representative of real-world measurement conditions. In 154 Stage 2, we provided operators with 10-m wind data collected onsite. All operators could then 155 reanalyze results and submit modified quantification estimates using the measured wind data. 156 157 The difference between Stage 1 and Stage 2 results therefore represents potential improvement from having access to real-time ground wind data. Finally, in Stage 3 we provided operators with 158 159 metered methane release rates for approximately half of their measurements, and which could be 160 used to inform a final submission based on an updated algorithm. Stage 3 results thus represent potential improvements possible with algorithm tuning. Details on data selection criteria for 161 Stage 3 are included in supplemental materials Section 1.3.6. All operators were provided the 162 163 opportunity to participate in all three stages of analysis, although only Carbon Mapper, GHGSat-AV, and Kairos chose to do so. Also note that Kairos data are the combined results from two 164 measurement units and MethaneAIR reports the average of two different analysis methods (both 165 166 discussed in detail in supplemental materials section 2.1.). 167

168 3 Results

- 170 Over the aircraft testing period, October 10th through November 11th, 2022, we conducted 704
- measurements with the five different aircraft operators. Of these measurements, 189 were
- 172 removed by Stanford for failing to meet quality control criteria designed to ensure clean
- conditions given real-time winds. Stanford exclusion criteria were finalized and applied before
 Stanford personnel viewed any operator results. The remaining 515 releases are included in
- Figure 2. Of total measurements conducted, 63 (8.9%) were intentional zero releases (0 kg/h) to
- serve as negative controls. There were a small number of times when the aircraft flew over the
- 177 field site, but no associated measurement was submitted with the operator report (due to some
- 178 measurement or processing error). These points are classified as "missing data" in Figure 2
- 179 (additional details in supplementary materials section 1.3.3 and Table S3).



Release Rate (kg / h)
 Figure 2: Distribution of releases for each aircraft tested, colors indicate results classification: true positive, true negative, false positive (no teams reported false positives), false negative, operator filtered (measurements for which the operator determined quantification was not possible), and missing data. Note that the three plots on the left have a different y-axis than the two on the right. For all operators, we conducted releases ranging from 0 to 1,500 kg CH₄ / hr. Figures do not include measurements filtered by Stanford, e.g. due to insufficient wind transport.

186 Table 2 summarizes operator-specific parameters for the measurements conducted in this study. 187 For reported metered flow rates, we use significant figures based on level of precision of the measurement and calculated uncertainty. All teams correctly categorized negative controls as 0 188 $kg(CH_4)/hr$, with no teams producing false positives. Additionally, we find no false negatives 189 190 larger than 30 kg(CH₄)/hr, and Kairos, GHGSat and Scientific Aviation quantified plumes 191 smaller than 4 kg(CH₄)/hr. Carbon Mapper, GHGSat-AV, and Kairos consistently quantify 192 releases above 10 kg(CH₄)/hr. For Kairos, 107 of 191 valid measurement were less than 15 193 kg(CH₄)/hr, providing the greatest characterization of minimum detection across all operators. 194 MethaneAIR and Scientific Aviation had a smaller sample size overall, and particularly for releases under 50 kg(CH₄)/hr. GHGSat-AV had three false negatives above 5 kg(CH₄)/hr (16.78 195 196 [16.67, 16.81], 29.01 [28.83, 29.18], and 29.17 [28.99, 29.35] kg(CH₄)/hr), which make up 8% of all measurements conducted in this range between 15 and 30 kg/hr. Additionally, Carbon 197 Mapper detected (but did not quantify) a release at 8.64 [8.45, 8.80] kg(CH₄)/hr 198 199

Carbon MapperGHGSat-AV AerospaceKairos MethaneAIRMethaneAIR Aviation
--

Number of reported	121	192	349	24	18
Number of	8	57	119	4	1
measurements filtered	°			•	-
by Stanford					
Number of	31	1	39	0	7
measurements filtered					
by operator ⁱ					
No. of quantified	82	140	191	20	11
measurements to pass					
all filtering					
Range of non-zero	4.45 [4.30,	1.05 [1.02,	0.64 [0.59,	24.42 [24.31,	3.77 [3.72,
Stanford release	4.59] - 1,440	1.08] - 1,140	0.69] - 1,110	24.53] - 1,290	3.83] - 800
volumes ⁱⁱ	[1,370, 1,520]	[1,110, 1,180]	[1,050, 1,180]	[1,220, 1,360]	[780, 830] kg
	kg CH4 / hr	CH ₄ /hr			
Smallest Quantified	10.92 [10.78,	2.91 [2.86,	3.40 [3.35,	33.61 [33.27,	3.77 [3.71,
Plume (kg CH ₄ / hr)	11.06] kg CH4	2.96] kg CH4/	3.46] kg CH4 /	33.94] kg CH4	3.83] kg CH ₄ /
	/ hr	hr	hr	/ hr	hr
Largest False Negative	6.61 [6.47,	29.17 [28.99,	10.47 [10.40,	24.42 [24.31,	No false
(kg CH ₄ / hr)	6.76] kg CH4/	29.35] kg CH4	10.53] kg CH4	24.53] kg CH4	negatives
	hr	/ hr	/ hr	/ hr	

200

ⁱ Operator filter applied only to measurements that pass Stanford filtering

"Non-zero Stanford releases before operator filtering

201

In Figure 3, we assess quantification accuracy for all correctly identified non-zero releases (true positives). For each stage of unblinding, we compare the metered release rate in kg(CH₄)/hr (xaxis) with the reported estimate (y-axis). Carbon Mapper, GHGSat, and Kairos Aerospace participated in the three stage unblinding process described above, and for these three operators Stage 1 results are in the left column, Stage 2 in the middle column, and Stage 3 in the right column. MethaneAIR and Scientific Aviation only participated in the first stage, submitting fully

208 blinded results. Results for these two operators are in the bottom row.

209

210 For plots in Figure 3, we include all quantified non-zero measurements to determine the linear

equation of best fit using ordinary least squares (OLS) regression, as in Sherwin, Chen et al.

212 2021.¹³ OLS is appropriate here because of the much smaller x-axis errors than y-axis errors

213 (e.g., metered emissions rate has high certainty). For all operators except Kairos, error bars on

both x- and y-axes represent the 95% confidence intervals (CI) of metered and reported results,

215 respectively. Carbon Mapper, GHGSat-AV, and Scientific Aviation reported uncertainty using 1-

- sigma values, which we convert for consistency. MethaneAIR reported uncertainty in 95% CI.
- 217 Kairos did not report uncertainty values for quantification estimates. For Kairos, each point

represents the average of the two measurement units used for collecting data, which vertical error

219 bars depicting reported values of individual units (analysis for each pod included in

220 Supplemental Results).



222 223 224

Figure 3: Quantification accuracy of aircraft platforms. Metered release rate is on the x-axis with error bars representing 95% CI, often not visible due to low values. Operator reported quantification estimates are on the y-axis. The dashed line represents the 225 226 227 x=y parity line. For all operators except Kairos, y-axis error bars represent operator reported uncertainty as 95% CI. Kairos does not report uncertainty, and y-error bars represent the variability in the two wing mounted measurement units flown during testing conditions.

228 For fully blinded result submission (Stage 1), we requested operators submit using analysis

- typical of standard operations. The four spectroscopy-based technologies submitted using the
- wind analysis products listed in Table 1. All three operators who submitted Stage 2 estimates
- used Stanford-provided 10-meter wind data. For Stage 3 partially unblinded submissions, Figure
 3 only includes the quantification estimates for releases that remained blinded, resulting in a
- so only includes the quantification estimates for releases that remained binded, resulting in asmaller sample size. Carbon Mapper requested the ability to re-add measurements they filtered in
- earlier stages as "poor quality" if unblinded information in later stages (wind data or unblinded
- 235 measurements) increased confidence in quantification estimates (discussed more fully in SI
- 236 Section S2.1.1.). Thus, quantification estimates for measurements not in Stages 1 and 2 appear in
- the Stage 3 parity figure.
- 238

239 Of the 71 measurements included in the fully blinded Carbon Mapper report (slope = 0.89, R^2 =

- 240 0.61), 89% have 95% confidence intervals that encompass the metered release rate. When
- provided ground truth wind data, Carbon Mapper reported estimates with reduced scatter (slope
- 242 = 0.82, $R^2 = 0.73$), but only 76% of included measurements have a 95% CI that intersects the true
- 243 metered value, as reflected in the decrease in slope. Both strength of fit and accuracy are highest
- in Carbon Mapper's Stage 3 results (slope = 0.96, $R^2 = 0.89$), where reported quantification
- estimates were informed using a subset of unblinded releases. In this stage, 80% of reported
- 246 measurements have error bars that intersect the parity line. Note that in this stage, Carbon
- Mapper chose to include 2 measurements previously removed by their own internal quality
 control. The percentage of Carbon Mapper measurements within 50% of the metered release rate
- 249 are 68% for Stage 1, 44% for Stage 2, and 62% for Stage 3.
- 250

251 Of the 121 reported measurements included in the fully blinded GHGSat report (slope = 0.76, R² 252 = 0.93), 93% of quantification estimates have 95% confidence intervals that cross the parity line. 253 Ground truth wind data improved slope alignment with the parity line in Stage 2 (slope = 0.93, 254 R² = 0.93). GHGSat-AV quantification uncertainty decreased in Stage 2: on average, the 95% CI

- reported in Stage 2 is 60% that of Stage 1 (range is 10% 110%). However, narrowing of
 confidence intervals resulted in a corresponding decrease in the number of quantification
- estimates with error bars crossing the parity line, despite improvement in slope. In the fully blind
- submission (Stage 1), 93% of quantification estimates have error bars that cross the parity line,
- whereas this is the case for only 84% of estimates when wind data is unblinded (Stage 2).Sixteen quantification estimates switched from crossing the parity line in Stage 1 to not crossing
- it in Stage 2, while only 5 estimates switched in the opposite direction. GHGSat-AV participated
- in Stage 3, but chose to make no adjustments to their Stage 2 submission after viewing the
- unblinded data. The percentage of GHGSat-AV measurements within 50% of the metered
- release rate are 80% for Stage 1 and 88% for Stage 2.
- 265

Kairos Aerospace quantified 124 non-zero releases and showed consistent performance across all three stages. Stage 1 results display a slight upward bias for larger quantification estimates (slope $= 1.13, R^2 = 0.87$), which becomes more pronounced with unblinded wind data in Stage 2 (slope $= 1.30, R^2 = 0.90$) and ground-truth data in Stage 3 (slope $= 1.31, R^2 = 0.90$). Kairos does not report uncertainty for quantification estimates (error bars represent range of the two instruments used in testing). However, we find that 73% of true positive quantification estimates fall within $\pm 50\%$ of the metered flow rate, and 38% are within $\pm 25\%$ of the metered flow rate.

274 Both MethaneAIR and Scientific Aviation only submitted fully blinded results. For

- 275 MethaneAIR, we include 18 non-zero quantification estimates (slope = 1.08, $R^2 = 0.93$). Of these
- 276 quantification estimates, 83% have 95% confidence intervals that cross the parity line and 78%
- of quantification estimates are within 50% of the metered release rate. Results included here are
- the average of two methods, whose individual results are included in the SI, Section S2.2.4. For
- 279 Scientific Aviation, we include 8 non-zero true positive quantification estimates (slope = 0.52, R² 280 = 0.93). Five of the 8 data points have 95% CI values that intersect the parity line, and seven
- (88%) have quantification estimates within 50% of the metered release rate.
- 282
- 283 For all spectroscopy-based technologies (Carbon Mapper, GHGSat-AV, Kairos and
- 284 MethaneAIR), percent error (depicted in supplementary Figure S18 Figure S20) is greatest for
- measurements conducted at rates below 200 kg(CH₄)/hr. For Carbon Mapper, GHGSat-AV and
 Kairos, absolute quantification error increases with increasing release rates while percent error
- 200 Karos, absolute quantification error increases with increasing release rates while percent error 287 decreases. The magnitude of the quantification error does not appear to increase with increasing
- 288 emission rates for MethaneAIR, although the sample size is limited. This result likely reflects the
- high sensitivity of the sensor to differences in CH_4 enhancement, and the application of two
- 205 ingli sensitivity of the sensor to differences in Cri4 emancement, and the application of two 290 quantification methods with complementary error characteristics. The small sample size for
- 250 quantification methods with complementary error characteristics. The small sample size for 291 Scientific Aviation limits our ability to draw conclusions regarding trends in error profile.
- 292 Percent error for Scientific Aviation quantification estimates are within the range of those
- 293 observed for fully blinded estimates by Carbon Mapper, GHGSat and Kairos Aerospace for the
- similar release ranges. A small sample size means the low estimate at 800 kg/h has an outsized
- effect on the linear regression and additional testing is needed for a more complete picture of
 Scientific Aviation's capabilities and error profile.
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Figure 4 illustrates the fraction of releases detected below 30 kgh for Carbon Mapper, GHGSat-

- 299 AV, and Kairos. MethaneAIR and Scientific Aviation are not included due to low sample size in
- this range. Characterizing lower detection limit was not a focus of Carbon Mapper
- 301 measurements, hence the smaller sample included in Figure 4. All operators consistently detected
- 302 releases above $10 \text{ kg}(\text{CH}_4)/\text{hr}$. While we conducted far fewer releases below $10 \text{ kg}(\text{CH}_4)/\text{hr}$ for
- 303 GHGSat-AV,both Kairos and GHGSat-AV detected a small proportion of releases below 5
- kg(CH₄)/hr. Additionally, GHGSat-AV missed 3 non-zero releases above 15 kg(CH₄)/hr. All
 operators detected all releases above 30 kg(CH₄)/hr.





Discussion 310 4

311

In this work, we evaluate performance of five different aircraft-based methane sensing systems. 312

313 This is the first independent single-blind test of Scientific Aviation. Of the four systems

previously tested by Brandt-group researchers at Stanford, all demonstrated improved 314

performance ^{10,13,14}. Note that previous tests with Kairos were conducted at a higher flight 315

altitude (900 meters/3,000 feet above ground level). 316

317

318 Carbon Mapper shows improved detection and quantification performance compared to results reported in Rutherford et al., 2023¹⁰. Previously, Carbon Mapper flew flight lines shorter than 319 typical, which their internal post-facto analysis suggests introduced low bias into quantification 320 321 estimates ¹⁷. For results reported here, Carbon Mapper flew 20 km flight lines, but other technical configurations remained similar to the earlier test. The best-fit slope for fully blinded 322

323 quantification estimates increased from 0.33 ($R^2 = 0.35$) to 0.89 ($R^2 = 0.61$). In the previous

324 study, Carbon Mapper showed a trend of overestimating lower emissions and underestimating

larger emissions, a trend not observed in these results. 325

326

327 GHGSat-AV fully-blinded results in this study show reduced scatter compared to previous testing¹⁰. R² increased from 0.38 to 0.93, indicating much closer agreement with a linear fit. 328 329 While the best-fit slope deviates more from the parity line (current study slope=0.76, previous

330 study slope=1.0), reduced scatter is indicative of overall improved performance: in Rutherford et

al., 2023, GHGSat-AV at times underestimated releases greater than 1,000 kg/h by a factor of 331 two more,¹⁰ while our results show no evidence of biased quantification for large releases.

332

333 GHGSat-AV also demonstrated improved lower detection capabilities. In Rutherford et al., they 334

did not detect any releases below 10 kg(CH₄)/hr, and missed over half of releases between 10 335 and 15 kg(CH₄)/hr.¹⁰ Here, GHGSat-AV detected one release below 5 kg(CH₄)/hr, and all 336 337 releases between 5 and 15 kg(CH₄)/hr. In both studies, GHGSat-AV missed a small number of releases above 25 kg(CH₄)/hr. In Rutherford et al., GHGSat-AV missed 2 of 42 releases between 338 339 25 and 35 kg(CH₄)/hr (release rates: 31.0 kg(CH₄)/hr and 32.4 kg(CH₄)/hr) ¹⁰. In this study,

340 GHSat-AV missed 2 out of 16 releases between 25 and 35 kg(CH₄)/hr, both ~29 kg(CH₄)/hr.

341

342 Kairos Aerospace maintained quantification performance while improving lower detection limit ¹³. In Sherwin, Chen et al., 2021, Kairos had a best-fit slope of 1.19 (with Dark Sky wind 343

reanalysis), compared to our result of 1.13. However, the flight configuration here shows a 344

decrease in detection threshold. Previously, Kairos was able to correctly identify all wind-345

346 normalized release rates 15 kgh/mps or larger ¹³. When normalizing our results by windspeed,

we find Kairos identifies all releases above 5 kgh/mps (see supplemental Figure S25). Sherwin, 347

Chen et al. find a standard deviation of percent error for all releases above the full detection limit 348

 $(41.76 \text{ kg}(\text{CH}_4)/\text{hr})$ to be 30-40% ¹³. Using the same lower limit for comparison purposes, we 349

350 find a similar standard deviation for percent error of 43%. However, we note that the tested configuration with two wing-mounted units may not be representative of field performance and 351

352 different test configurations limit direct comparison.

353

MethaneAIR previous conducted volume-blind controlled releases in collaboration with 354

Stanford, reported in Chulakadabba et al., 2023.¹⁴ Quantification accuracy is similar to the 355

previous study with reduced scatter (current study slope=1.08 with R²=0.93; previous study OLS 356 slope=0.85 and York slope=0.96, $R^2 = 0.83$)¹⁴. However, results are not directly comparable, as 357 the previous study reports quantification estimates using the mIME method, while MethaneAIR 358 359 reported the average of two methods in the current study (results for individual methods in 360 supplementary material Section 2.2.4). 361 362 Conley et al., 2017 report two natural gas controlled release measurements for Scientific Aviation, although these were not part of a single-blind study ¹¹. Both these releases were at rates 363 364 of 14 kg(CH₄)/hr, smaller than all but one of the non-zero releases quantified by Scientific 365 Aviation in the current study. 366 367 The present study has several important limitations. Providing participants with a known source 368 location could artificially inflate detection performance. However, it is unlikely to affect 369 quantification capabilities. We also selected our testing location to minimize confounding 370 sources and provide a uniform, dry terrain as background. Field measurements will often occur 371 over complex terrains with multiple confounding sources within measurement range, thus 372 technology performance may vary in other environments. Furthermore, except for Scientific 373 Aviation, weather conditions during testing were conducive to measurement, with limited cloud 374 cover. Cloudy conditions add challenges for spectroscopy-based detection and quantification. 375 376 This work provides a comprehensive overview of the major methane-sensing aircraft 377 technologies. While we did not test Bridger Photonics, this company has been extensively tested elsewhere^{10,15,16}. We evaluate the state-of-the-art for all systems tested, demonstrating the ability 378 of aircraft-based technologies to produce estimates with limited bias and within reasonable error. 379 380 Our results also underscore the importance of controlled-release testing to allow technology developers to fine-tune their systems. Both Carbon Mapper and GHGSat-AV demonstrated 381 substantial performance advances compared to previous tests ¹⁰, and the multi-stage unblinding 382 within this study allowed Carbon Mapper to rapidly iterate and hone their quantification 383 384 algorithm. 385 386 This study demonstrates aircraft-based methane sensing is posed for an increasingly important 387 role in climate change mitigation efforts and improving accuracy of the global methane budget. The approach outlined here can be used as technologies continue to mature and new methods 388 389 develop, ensuring high quality, accurate measurements underpin environmental regulation and 390 enforcement. 5 Data and Code Availability 391 392

All data and code required to reproduce the figures and analysis in this paper will be made
available prior to publication. Due to ongoing analysis of other parts of the study, we are
currently refraining from sharing raw data publicly as of June 2023.

396

397 6 Acknowledgements

- 399This research was funded by: Environmental Defense Fund, Global Methane Hub, International
- 400 Methane Emissions Observatory, and Stanford Natural Gas Initiative (an industry consortium401 that supports independent research at Stanford).
- 402

We acknowledge all operational team who supported participation in this test, and provided
logistical and coordination support. Carbon Mapper flight planning and execution: Joseph
Heckler (ASU), Greg Asner (ASU), Andrew Aubrey (Carbon Mapper); Carbon Mapper data
processing and quality control: Daniel Cusworth, Alana Ayasse, Riley Duren, Kate Howell,
We acknowledge all operational team who supported participation in this test, and provided
Heckler (ASU), Greg Asner (ASU), Andrew Aubrey (Carbon Mapper); Carbon Mapper data
processing and quality control: Daniel Cusworth, Alana Ayasse, Riley Duren, Kate Howell,

- Kelly O'Neill, David Stepp, Ralph Jiorle; GHGSat: Marianne Girard, Jason McKeever, Warren
 Shaw, Jordan Deboer, Rafael Del Bello, Gillian Rowan, Ángel Esparza, Charlott Reed; Kairos
- 409 Aerospace: Belinda Chin, Matt Cocca, Sheamus Flanagan, Amy Giver, Harshil Kamdar, Patrick
- 410 Steele, Michael Swope, Erin Wetherley. MethaneAIR: Apisada Chulakadabba, Maryann Sargent,
- 411 Jenna Samra, Jacob Hawthorne, Bruce Daube, Steven Wofsy. Scientific Aviation: Mackenzie
- 412 Smith, David Carroll.
- 413
- 414 Rawhide Leasing and Volta Fabrication personnel provided essential operational, logistical,
- 415 planning, and technical support for the experiment: Mike Brandon, Walt Godsil, S.M., Merritt
- 416 Norton, Dana Walker. C. Kocurek provided helpful input on experimental design. Thuy Nguyen
- 417 and Cerise Burns provided invaluable administrative support. We also thank Natalie Schauser for
- 418 technical advising on Git and version control, and the Creative Café for accommodating the419 dietary restrictions of the Stanford field team.
- 420

421 7 Author Contributions

422

423 Conceptualization – S.H.E., E.D.S., A.R.B. Methods – S.H.E., J.S.R., Y.C., E.D.S., A.R.B.

424 Software – S.H.E., P.M.B., Z.C. Validation – S.H.E. Formal analysis – S.H.E. Investigation –

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429 8 Statement of Competing Interest

430

431 A.R.B. is a member of the Advisory Committee (Science and Measurement Committee) for

- 432 Carbon Mapper. Y.C. and Z.Z. were research interns at Carbon Mapper in Summer 2022, and
- 433 Z.Z. received academic funding from Carbon Mapper in Fall 2022 for a project unrelated to the
- 434 current work. J.S.R. is currently employed by Highwood Emissions Management but was an
- 435 affiliate of Stanford University when contributing to the current study.
- 436

437 9 References

- 438
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- 518

⁵¹⁹ Supplementary Information

520 1 Supplemental Methods

521 1.1 Experimental Field Setup

We conducted controlled releases from October 10th, 2022 through November 30th, 2022 near 522 Casa Grande, Arizona at coordinates [32.8218489, -111.7857599]. We evaluated aircraft, 523 satellite, drone, and ground-based technologies. Natural gas trailers and pressure regulation 524 525 trailers were provided by Rawhide Leasing (https://www.rawhideleasing.com/), and Rawhide 526 personnel operated this equipment (Mike Brandon, Walt Godsil, and S.M.). The gas metering 527 trailer was designed by the Stanford team in collaboration with Volta Fabrication, who constructed the trailer. The Stanford team controlled gas flow rates using a WiFi-enabled laptop 528 529 connected the flow control system on the metering trailer. Gas was released from two stacks,

- each with 6-inch diameter and release heights of 24 and 10 feet.
- 531



Figure S1: Overhead view of Stanford field site, with key components labelled. Also visible but not labelled here are individual ground sensors deployed for the duration of the experiment.

The testing configuration included the following key components, also depicted in Figure S1,described below in full:

537

532 533

- 538 1. Two compressed natural gas trailers
- 539 2. Pressure regulation trailer
- 540 3. Flow metering trailer fitted with three Emerson Micromotion Coriolis meters measuring
 541 gas flow rate in kg(CH₄)/hr. Two release stacks allow for vertical gas release at 7.3
 542 meters (24-feet) and 3.0 meters (10-feet) above the ground
- 543
 4. Three-dimensional sonic anemometer (Campbell Scientific, CSAT 3B) mounted at 10544 meter height on an aluminum trailer tower (Aluma Towers)
- 545 5. Two-dimensional ultra-sonic anemometer, mounted at 2 meters
- 546 6. Workstation with computers for controlling flow rates and logging data, located over 45
 547 meters (150 feet) from all gas flow equipment (49 meters (160 feet) from metering trailer, 45 meters (150 feet) from Rawhide equipment).
 - 7. Infrared camera (FLIR GF320) focused on stack and used for real-time plume observations and recording
- 552 1.1.1 Compressed natural gas trailers
- 553

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551

As previously described, we used compressed natural gas (CNG) as the source of methane for controlled releases (Ravikumar et al., 2019; Sherwin, Chen et al., 2021; Rutherford et al., 2022). CNG was purchased from local filling stations and stored onsite in two contracted CNG storage trailers. Capacity of CNG trailers is described in Table S1. Pressure in the CNG trailers ranged from 3.5 – 17.3 MPa (500 psig to 2500 psig), varying with ambient temperature and gas fill level.

560

Table S1 Compressed natural gas trailer specifications. Trailer IDs are assigned by Rawhide Leasing. Water volume in cubic feet
 refers to the total volume of water that can be held in the tank. Full capacity and working capacity refer to gas capacity at max
 pressure. Working volume accounts for the pressure differential needed to maintain gas delivery to the pressure regulation
 trailer.

Trailer	Water Volume (ft ³)	Max Pressure	Full Capacity (Mscf at max pressure)	Working Capacity (Mscf)
911-49	19.81 m ³ (699.5 ft ³)	16.6 MPa (2400 psig)	106	90
911-2	9.63 m ³ (342 ft ³)	17.3 MPa (2500 psig)	76	63

565

566

567 1.1.2 Pressure regulation trailer

568

569 When releasing gas, one or both of the CNG trailers is connected to a pressure regulation trailer (Rawhide Leasing, RT-30), which reduces pressure from that of the CNG trailer to the pressure 570 571 rating of the gas metering trailer. Gas is transferred from trailers to the pressure regulation trailer using 13mm Parflex CNG hose, rated to withstand 34.5 MPa (5,000 psi). Depending on the 572 573 amount of gas remaining in the CNG trailer, the inlet pressure to pressure regulation trailer 574 changes. Detailed descriptions of the pressure regulators on the RT-30 are provided below. Gas 575 leaves the pressure regulation trailer at 1.14 - 1.48 MPa (150 - 200 psig), and is delivered via a 576 hose to the gas metering trailer. 577



Figure S2 RT-30 pressure regulation trailer. Gas is delivered from CNG trailers via the red hoses on the left, and exits through the blue hose on the right.

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580

Photograph and schematic of the regulation trailer are depicted in Figures S2 and S3. After
entering the trailer inlet, gas is delivered to either one or both of two parallel pressure regulation
lines. Each line is fitted with a microglass 6-micron fuel filter (3B Filters Inc., Model A8579V6MD) followed by two pressure regulation units: a stainless steel Tescom pressure regulator
(Model number 44-1325-2122-011) followed in-series by a Fisher pressure regulator (initially

- 587 model number 627, then model number 627H; discussed further below).
- 588

Tescom regulators, rated for inlet pressures up to 31.13 MPa (4500 psig), decrease pressure from

- the level of the CNG trailer to 2.86 MPa (400 psig). The inlet pressure to the Tescom regulators
- changes with the amount of gas remaining in the CNG trailer, and thus the pressure drop across
- the regulator changes. This affects the cooling generated during gas expansion (discussed
- below). Next follows the Fisher pressure regulators, rated for inlet pressures of 700 psig, further stepping down the pressure to 1.14 - 1.48 MPa (150 - 200 psig). Initially, RT-30 was fitted with
- 595 Fisher 627 pressure regulators, the maximum outlet pressure of which is 1.14 MPa (150 psig).
- 596 On October 22, 2022, Rawhide personnel replaced the Fisher 627 with model 627H, enabling an
- 597 outlet pressure of 1.48 MPa (200 psig). After leaving the Fisher pressure regulators, gas flows
- from the two pressure regulation lines into a multi-port manifold fitted with a pressure gauge to
- 599 measure final outlet pressure from the regulation trailer.
- 600
- Each pressure regulator is fitted with a catalytic heater (Tescom heaters: CATCO 90-66S1G-40;
- 602 Fisher heaters: CATCO 90-612S1G-40), as depicted in Figure S3. Heaters are applied to
- 603 partially compensate for the Joule-Thompson temperature drop resulting from the increase in gas
- 604 pressure. Throughout the experiment, these catalytic heaters were used for gas flow rates
- exceeding ~250 kg/h for over 15 minutes. Gas from the multi-port manifold is used to power the

606 catalytic heaters. An additional Fisher pressure regulator reduces gas pressure from the manifold

level (1.48 MPa or 200 psig) to 0.14 MPa (5 psig) before delivery to the heaters (not depicted in 607 608 Figure S3).

609



610 611

Figure S3 Pressure regulation trailer. Gas enters from the CNG trailers via red hoses on the left of the image, and passes through 612 either one or both of two pressure regulation lines. Pressure is first dropped through the Tescoms pressure regulators to 2.86 613 MPa (400 psig) in the Fisher pressure regulators. A pressure gauge measures final outlet pressure before gas is delivered to a 3-614 inch hose which connects to the metering trailer.

The multi-port manifold on the pressure regulation trailer also connects to a buffer tank fitted 615

with a pressure gauge and gas sampling port. The buffer tank is rated to withstand pressures of 616

617 up to 1.48 MPa (200 psig) and has a safety valve set for 1.31 MPa (175 psig). To collect gas

618 samples for analysis, a gas line connecting the manifold to the buffer tank is opened and pressure

is allowed to reach 0.27 MPa (25 psig) within the buffer tank. Laboratory-supplied collection 619 620

canisters are connected to the sampling port, the line to the port is opened, and gas flows from the tank into the collection canister. Details of gas sampling are described in further detail below 621

622 (see Section S1.2.3 for further discussion of gas composition).

623

624 The pressure regulation trailer also includes the following equipment not depicted in Figure S3: Safety pressure release valves in case of failure in the pressure regulators; safety release valves 625

on the buffer tank; and ambient air heaters (not used during the experiment). Safety pressure 626

- 627 release valves on the pipe connecting the Tescoms to the Fishers are set for 6.65 MPa (950 psig).
- 628

After successive drops in pressure in the RT-30, gas is delivered to the gas metering trailer. The 629

630 gas metering trailer was designed for inlet gas pressure of 1.14 MPa (150 psig), with all

equipment rated for 1.48 MPa (200 psig). To achieve the desired inlet pressure, the metering 631

trailer was originally intended to be coupled with the RT-60 pressure regulation trailer, not the 632

633 RT-30. However, due to supply chain delays, RT-60 construction was not complete in time for testing. While pressure regulators in the RT-30 can drop gas pressure to 1.48 MPa (200 psig), 634

this pressure is not maintained due to a constriction at the point where the RT-30 outlet connects 635

to the hose that transports gas to the metering trailer. This meant that gas at the inlet of the 636

637 metering trailer was typically lower than 1.14 MPa (150 psig) for large release volumes, despite

- the pressure gauge in the RT-30 multi-port manifold reading 1.48 MPa (200 psig). Pressure 638
- 639 limitations impacted the maximum flow rate of the metering trailer. While designed to support
- 640 gas releases up to 2,000 kg gas / hr, the maximum release volume achieved during throughout

- 641 the duration of this experiment was ~1,600 kg gas / hr. At the highest flow rates, the pressure
- 642 drop in the system becomes large and flow becomes erratic.
- 643
- 644 1.1.3 Gas metering trailer
- 645



646 647

Figure S4 Aerial photograph of gas metering trailer

648 Gas from the pressure regulation trailer is transported to the gas metering trailer (photograph in 649 Figure S4), consisting of three flow paths each fitted with a Coriolis gas flow mater. The 650 Stanford team controlled the desired flow rate, diverting gas through one of the three flow paths, before it is released through one of two vertical stacks. In this section, we describe the metering 651 652 and flow control mechanisms in detail.

653

654 1.1.3.1 Gas metering and flow control

A 7.62-cm (3-inch) wire-spring reinforced hose (pressure rating 200 psig) transports gas from the 655 pressure regulation trailer to the gas metering trailer, as depicted in the schematic in Figure S5. 656

657 Gas first passes a Quadratherm thermal mass flow insertion meter (see additional details below),

658 before being diverted to one of three parallel lines, each line is fitted with an Emerson

- 659 MicroMotion Coriolis meter (https://www.emerson.com/en-us/automation/micro-motion). All
- 660 pipes in the Ouadratherm measurement apparatus and the gas metering trailer are Schedule 10 661 stainless steel.
- 662
- 663 Model identification numbers, serial numbers, and calibration dates for each Coriolis meter are
- included in Table S2, and flow ranges for each meter are included in Table S3. The max flow 664
- 665 range for each meter is based on the maximum recommended gas velocity of 140 m/s through
- 666 the sensor (personal communication from Hector Rodriguez of Micro Motion to Jeff Rutherford
- on November 12, 2021). Due to supply chain delays, all testing prior to October 24th was 667
- conducted using the medium-diameter CMF050M meter only; subsequent tests used all three 668
- 669 Coriolis meters. We attempted to conduct large satellite-coincident releases using the

670 Quadratherm 640i, but these measurements were discarded due to system malfunction (discussed

671 below). 672



673 pressure regulation skid
674 Figure S5 Schematic of Quadratherm measurement apparatus and gas metering trailer (not to scale). Gas flows past the
675 Quadratherm insertion meter, before being diverted through one of three parallel lines, each fitted with a Micromotion Coriolis
676 meter. Gas then enters a 15.24 cm (6-inch) pipe connected to two release stacks. Stacks are 6.1 meters (6 feet) and 1.8 meters
677 (20 feet) long, releasing gas at 3.0 meters (10 feet) and 7.3 meters (24 feet) above ground level, respectively.

678 The gas flow into each meter is controlled by a solenoid valve (Magnatrol Valve Corp, Models 679 F42K37-GSW, F31K34-GSW, and F14K32-GSW on the 1¹/₂-inch, 1-inch, and ¹/₂-inch lines, respectively) which can either be in the fully open or fully closed position. When a given 680 681 solenoid valve is open, gas flows through the corresponding pipe and Coriolis meter. Only one solenoid valve was opened at a given time. A downstream flow control gate valve (SVC Flow 682 Controls, Model E400X2EC) is used to set the gas flow rate based on the extent to which it is 683 opened, measured in percent. After passing through flow control valves, gas then enters a 6-inch 684 pipe fitted with two 6-inch butterfly valves (SVF Flow Controls, SLB series) that control flow to 685 686 the release stacks.

- 687
- Each Coriolis meter on the metering trailer is equipped with a field mounted Micro Motion 5700
- transmitter that converts the raw sensor data to a 4-20 milliamp (mA) signal (see Table S3). A
- 690 wired connection delivers the mA output from each transmitter to the metering trailer's
- 691 programmable logic controller (PLC). The mA output signal was transmitted with zero
- dampening applied to the flow reading, dampening effects will be applied in the subsequent data

- analysis pipeline. The PLC (Horner Automation, HE-X5GN) is located in the control panel
- depicted on Figure S5. A WiFi adaptor transmits data to a laptop computer operated at the
- 695 Stanford Work Station. All external wiring from flow meter transmitters, flow control valves,
- and solenoid valves use Class 1 Division 1 hazardous location approved, ruggedized, pre-
- 697 manufactured Mineral Insulated cables that are fire resistant and waterproof. Cables provide
- 698 power to all meters and valves (M.I Cable Company, Part Number 2/16/3/SB6-12/H) and
- transmit the 4-20 mA signal from the meters to the PLC (M.I Cable Company, Part Number2/16/3/SB6-12/H-TD).
- 701

Table S2: Full model number of each Coriolis meter. A meter consists of a sensor, through which the gas flows, and its associated
 transmitter. We include model ID and serial numbers for each sensor / transmitter pair.

Meter (Model	Sensor Model	Transmitter	Sensor Serial	Transmitter	Calibration
Abbreviation)	ID	Model ID	Number	Serial	Date
				Number	
Small	CMFS015H52	5700I12AB2A	12219231	12222349	October 14,
(CMFS015H)	0NFA2ECZZ	ZZXAAAZA_			2022
		40102			
Medium	CMF050M31	5700R12ABA	21175085	12205694	September
(CMF050M)	9N2BAEZZZ	AZZXAAZZZ			21, 2021
		_40102			
Large	CMFS150M	5700I12AB2	12220939	12222533	October
(CMFS150M)	341NFA2EK	AZZXAAAZ			18, 2022
	ZZ	A_40102			

704 705

Table S3 Sizing and specifications for Emerson MicroMotion Coriolis meters. Flow range represents the desired flow range for
 each meter, although rates outside the specified range are possible and require adjustments to meter uncertainty.

Meter	Meter Size	Connecting Pipe	Flow Range	4-20 mA Output Range
		Size	(kgh)	
Small	0.166 inch	0.5 inch	2-30 kgh	0 - 50 kgh
Medium	0.5 inch	1.0 inch	30 – 300 kgh	0-400 kgh
Large	1.5 inch	1.5 inch	300 – 2,000 kgh	0-3,000 kgh

707

Flow rates can be controlled using either an automated or manual control system, using a WiFi connected laptop. The automated feedback system uses a proportional-integral-derivative (PID) controller to adjust the flow control valve to achieve a desired set point, while the manual control system allows the Stanford team to set the degree to which the gate valve opens by specifying a desired percentage.

- 713
- From October $10^{\text{th}} 20^{\text{th}}$, we used an automated feedback system for flow control. However, we

observed flow fluctuations associated with overcorrections in the feedback system. While

accuracy of measurement was not affected, flow rate was more variable (see Figure S6A). On

717 October 20th we switched to manually controlling valve settings via the laptop interface. With

this control mechanism, we set the gate valve position in order to achieve a desired flow rate.

Figure S6B shows the reduction in fluctuations achieved by switching to this manual control

720 system.



723 724

722

Figure S6 Sample flow rates with automated (A) vs manual (B) flow control. Note the y-axis differs in the two plots

725 1.1.3.2 Thermal mass flow metering

726

727 Our gas metering and release trailer included upstream thermal mass flow meters for comparison 728 with the Emerson MicroMotion meters and for potential use prior to the arrival of the large meter 729 (shipment delayed due to supply chain issues). The Sierra instruments are calibrated for a range of 250 to 1,180 kg/hr (when installed in a 3-inch pipe). Thus, we intended to use it alongside the 730 731 Medium Coriolis meter to conduct higher volume releases. However, both meters are calibrated 732 to conduct releases at 300 kg/hr. When we conducted releases at this range, we observed 733 inconsistent discrepancies between the two meter readings, in which they were often offset from each other by 10 - 20%. Due to higher documented level of manufacturer-reported measurement 734 735 certainty with the Coriolis meters, we opted to use only the Coriolis meters for all experimental 736 data. However, here we provide documentation of the thermal mass flow meter configuration.

737

738 Prior to entering the metering trailer, gas passes through a 7.6 cm (3-inch) inlet pipe equipped for

739 installing an insertion flow meter. For all testing, the spool included an installed Sierra

740 Instruments Quadratherm 640i (Figure S7A), connected to the PLC and transmitting meter

readings to the connected laptop computer via WiFi. During testing on October 10, 11th and 12th,

gas was delivered through the Quadratherm 640i and subsequently through the CMF050M, at

flow rates overlapping with the calibration range of the two meters. Due to observed

inconsistencies in meter reading, we made several adjustments to the hose and Quadratherm

- metering configuration, and added installed an additional upstream Quadratherm 780i for crosscomparison.
- 746 747

Throughout all testing, the Quadratherm 640i had an upstream straight run of pipe over 356 cm

749 (140 inches) long, corresponding to >46 upstream diameters (see Figure S7A). This upstream

pipe length exceeds the requirements reported in the Sierra Quadratherm manual, which

recommends 40 upstream diameters of straight pipe after a flow control valve or two elbows in a

- different plane (<u>Sierra Instruments, 2014</u>). Downstream of the Quadratherm 640i were 32 cm
- 753 (12.5 inches) of straight pipe before a 7.6 cm to 5.1 cm (3-inch to 2-inch) pipe flange that marks

- gas entering the gas metering trailer itself. This length corresponds to >4 downstream diameters,
- exceeding the recommended 3 straight-pipe diameters recommended downstream of the
- 756 Quadratherm 640i when downstream pipe size decreases by a factor of 4:1 (Sierra Instruments,
- 757 <u>2014</u>).
- 758



759 bell reducer
 760 Figure S7 A. Upstream pipe connecting 3-inch hose to metering trailer with installed Quadratherm 640i. B. Pipe spool with
 761 Quadratherm 780i installed in series upstream of the Quadratherm 640i. The inline pipe attached to the Quadratherm contains
 762 the flow conditioning unit and is depicted in orange.

On October 13th, the 3-inch hose delivering gas to the Quadratherm 640i spool was straightened and elevated with car jacks to further reduce any potential upstream sources of turbulence to the gas flow. With this configuration, there were 14.5 feet between the Quadratherm 640i and the downward curve of the hose, and 35 feet before the hose curved laterally towards the pressure regulation trailer.

768

From October 17th through October 30th, an additional 2-inch pipe was installed with an inline 769 770 Sierra Instruments Quadratherm 780i meter, provided by Kairos Aerospace for measurement inter-comparison (Figure S7B). This meter was placed upstream of the 640i and allowed us to 771 772 compare the reading from two Quadratherm meters in series. The 780i Quadratherm included a 773 flow conditioning unit within the inline pipe manufactured and attached to the meter itself. The installed piping included 35.56 cm (14 inches) of upstream straight pipe and 38.1 cm (15 inches) 774 775 of downstream straight pipe, corresponding to 7 and 7.5 upstream and downstream diameters, respectively. Upstream and downstream pipe lengths comply manufacturer's recommendations 776 for this instrument and this piping configuration. 777

- 778
- 779 Ultimately, none of the data from Quadratherm thermal mass flow meters are used in generating
- final flow rate measurements for analysis. The calibrated uncertainty on the Coriolis meters is
- much smaller than the Quadratherm meters, and the empirical comparison between the two

782 Ouadratherm meters in series supports this fact. Therefore, the Ouadratherm meters are treated as 783 "backup" meters only and were not required to be used in any testing. Intercomparison between 784 the measurements of the Quadratherm meters and the CMF050M Coriolis meter may be the 785 subject of future analysis.

786

787 1.1.3.3 Gas Release Stacks

788

789 The two gas release stacks are made of 6-inch diameter high-density polyethylene tubing, 6 790 meters (20 feet) and 1.8 meters (6 feet) long, attached to a rotating elbow joint. When stacks are 791 in the vertical position, gas is released 7.3 meters (24 feet) and 3 meters (10 feet) above ground 792 level, respectively. The rotating elbow assembly allows gas to also be released while the stacks 793 are in a horizontal position, with the polyethylene tubing parallel to the ground. In this 794 configuration, gas is released 0.9 meters (3 feet) above ground level. In this study, however, 795 stacks were only used in the vertical position.

796

797 The metering trailer was designed for gas to flow through one open butterfly valve to the desired 798 release stack, while the other butterfly valve shut gas flow to the stack not in use. We conducted initial testing using the 20-ft release stack. On Oct 26th, we observed gas slip from the short stack 799 800 using the infrared camera (FLIR GF320). We reviewed our own internal infrared footage, as well 801 as infrared footage continuously collected by a Kuva Systems unit installed on site for testing as part of the continuous monitoring testing program (Kuva Systems, 2023). We determined gas 802 slip may have begun as early as October 20th. While we did not systematically evaluate when slip 803 804 was occurring, we were able to visualize gas slip using high sensitivity mode on the FLIR camera at whole-system flow rates as low as 300 kg/hr. However, we were only able to 805 806 consistently visualize slip during whole-system release rates exceeding 800 kg/hr. Because we 807 only conducted releases at rates greater than 300 kg/hr after the large Coriolis meter arrived on 808 October 20th, we have high certainty that meaningful gas slip did not occur before this date.

809

To prevent further gas leakage, on November 1st, we removed the short stack and sealed the pipe. 810 On November 14th, we reinstalled the short stack, removing and sealing the tall stack. The 811 timeline associated with stack leak is provided in Table S4. Methane slip occurred during testing 812 813 of Kairos Aerospace, and all continuous monitoring teams deployed for the relevant dates. Kairos Aerospace reported observing leaks from both stacks in imaging, and we provided the

- 814
- 815 information in Table S4 to all continuous monitoring teams.
- 816
- 817

Date	Stack Usage and Slip
October 10 th – October 20 th	Tall Stack, no slip
October 20 th – October 30 th	Tall stack, with slip
October 31 st	Short stack, with tall stack slip
November 1 st – November 14 th	Tall stack, short stack removed (no slip)
November 14 th – November 30 th	Short stack, tall stack removed (no slip)

818 Table S4 Dates indicating usage of tall vs short release stacks, and whether or not methane slip was observed.

820 1.1.4 3-D ultrasonic anemometer

821

A three-dimensional (3-D) ultrasonic anemometer (Campbell Scientific, CSAT 3B) was mounted

823 on a 10-m stainless steel trailer tower (Aluma Towers) at coordinates [32.8220109, -

824 111.7861257], 33 meters (108 feet) from the release point. Coordinates are measured using an
825 iPhone Google Maps pin drop. The anemometer was installed with the prongs oriented towards

the direction of the dominant prevailing wind (NE), per manufacturer recommendations. The

azimuth angle, or the angle of the anemometer orientation relative to Magnetic North, was 45°,

as measured with a magnetic compass. This corresponds to a 35.3° angle relative to True North,

- assuming a declination value of 9.7° for Casa Grande, AZ (National Oceanic and Atmospheric
- 830 Administration Geophysical Data Center, 2023). Orientation of the anemometer relative to True
- 831 North and Magnetic North is depicted in Figure S8. Wind directionality is recorded in degrees
- relative to True North, and reported as a vector indicating the direction from which the wind is
- 833 coming.
- 834

835 Wind speed and direction were recorded at a frequency of 1 Hz using a CR1000X data logger.

836 We collected data daily and processed it using PC400 (version 4.7), software provided by

837 Campbell Scientific. Our script uses all default settings provided by Campbell Scientific, but

adjusted the scan interval to 1 second for 1 Hz data logging. We also programmed the azimuth

- angle (discussed above) of 35.3° .
- 840

841 After collecting data, we combined all files corresponding to a single date (in UTC). During data

cleaning, we removed any gaps or repeats in the dataset. Gaps were 2-5 second in length, and

843 none occurred during aircraft testing periods. We replaced data gaps with NA values. There were

844 also occasional repeated timestamps in the data: a timestamp would appear once with wind data 845 entered in each relevant field, with the same timestamp appearing again with blank entries in

- each field. In each such instance, we deleted the redundant (empty) timestamp.
- 847

On November 4th, 2022 we experienced equipment malfunction with data collection, and no data
are available from the 3-D anemometer for that date.

850



Figure S8 Orientation of CSAT 3B relative to True North and Magnetic North, with a 9.7° declination. Azimuth angle refers to the
angle between the anemometer orientation and North. The azimuth angle relative to Magnetic North was measured to be 45°,

854 855	and this value was adjusted to account for the magnetic declination (9.7°) to determine the azimuth angle of 35° relative to True North.
856	
857 858 859 860 861 862 863 863 864 865 866 867	1.1.5 2-D ultrasonic anemometer A two-dimensional (2-D) ultrasonic anemometer (Gill Instruments, Windsonic 60) was mounted on a tripod at 2-meter height at coordinates [32.8219591, -111.7851434], measured via iPhone Google Maps pin drop. The 2-D anemometer was 59 meters (194 feet) from the release stack. The indicator on the anemometer was oriented towards North, as per manufacturer's instructions. The size of the indicator was such that it was not feasible to reliably differentiate the between True North and Magnetic North given the declination of $< 10^{\circ}$. While we logged 2-D anemometer data daily, it was only used in analysis for November 4 th when 3-D anemometer data was not available (discussed above).
868 869	1.2 Data processing for raw meter data
870 871 872 873 874 875 876 877 878 879 880 881 882 883 884	1.2.1 Metering trailer data log Data from the metering trailer was collected on a Stanford laptop using the program Configuration: Node-Red which outputs a CSV data log with secondly timestamps and corresponding columns for each solenoid valve, gate valve, and flow path. The data log indicates whether or not each solenoid valve was open, the percent to which the gate valve is open, as well as providing the metered flow through each flow path. The format of the data log was programmed by VINCEENGINEERING, PLLC (Salt Lake City, Utah), and modifications were made based on requests by the Stanford team throughout the testing period. Due to limitations in the programming of the software control system, we rely on the metered data collected directly from the Coriolis meters themselves, as opposed to from the data log to determine whether or not the solenoid valves are open, discussed in greater detail below.
884 885 886 887 888 889 890 891 892 893 894 893 894 895 896 897 898	On November 3 rd , 2023 we collected the historical data from all three Coriolis meters. Data recorded on each meter began October 3 rd . Subsequently, for each day of testing, we collected historical data for all meter used. Figure S9 summarizes the data cleaning process for the historical Coriolis meter files. First, we applied necessary timestamp corrections to enable merging the Coriolis meter data file with the flowskid data log. Briefly, the CMF050M internal clock lagged 10 minutes and 10 seconds behind the other meters, likely because of its previous purchase date. The flowskid data log timestamps were adjusted to UTC time using side-by-side photographs of the laptop clock and an iPhone displaying the time from World Clock (timeanddate.com/worldclock/). All adjustments to flowskid data log timestamps are summarized in Table S5. Additionally, for the CMFS015H and CMFS150M meters, we removed all historical data from before the meters arrived onsite. Mass flow units of the meter files were also converted from whole gas kg/s to kg/hr.



Figure S9 Data processing flow chart for determining methane release rate ($kg(CH_4)/hr$), using flowskid data log and Coriolis meter raw data files.

902

Table S5 Summary of timestamp adjustments to flowskid data log. All comparisons are between the data logging laptop and
 World Clock UTC on iPhone. On October 10th, the data logging computer system clock had not been calibrated to UTC time, and
 a 17 second adjustment was made after the first day of testing. Flowskid laptop was then set to British Summer Time (BST), and
 aligned with UTC World Clock time. On October 13th, British Summer Time shifted by 1 hour, resulting in a delay compared to
 UTC. On October 19th, we changed the laptop system clock to UTC time.

Date	Timestamp Correction to Flowskid Data Log
2022-10-10	Flowskid log 17 seconds behind World Clock UTC
2022-10-11	Flowskid log aligned with World Clock UTC (laptop system clock set
	to British Summer Time)
2022-10-13 through	Flowskid log 1-hour behind World Clock UTC (British Summer Time
2022-10-19	time change, resulted in 1-hr offset)
2022-10-22 and after	Flowskid log aligned with World Clock UTC (laptop system clock set
	to UTC)

908

After merging the two file types for each meter, the data is filtered to generate two datasets for

each meter: one in which gas was flowing through the meter, and one in which it was not. Gas

911 was determined to be flowing through the meter if the solenoid valve status was set to open. For

912 a meter reading to qualify for the "no-flow" dataset, it must meet two criteria: the solenoid valve

913 must be closed and the meter reading must be below a noise threshold of 0.2 kg/hr. Both criteria

are required because the field team would close the solenoid valve when flow rate was at 5 to 20

- 815 kg/hr, meaning the solenoid status was not sufficient for determining if no gas was flowing
- 916 through the meter or not. We use this noise threshold instead of setting a required flow rate to 0

- 817 kg/hr because the CMF050 flow readings included noise of up to 0.2 kg/hr when the meter was
- 918 definitively not in use (likely due to random vibration or other noise sources in the system).
- 919
- Gas flow and no-flow data files for each file where then combined to generate flow and no-flow
- 921 datasets across all meters. To generate the dataset of gas flow rates, we use the union of all three
- 922 meter flow files. To generate the dataset of no-flow periods, we determine the intersection of the
- three no-flow meter files: in other words, all three solenoid valves must be closed and gas flow
- rate through each meter must be less than the noise threshold. Meter measurements where the
- solenoid valve is closed but flowrate is greater than the noise threshold are re-added in the final
- data-cleaning stage. During this final cleaning, we also set meter readings outside of testing
- 927 periods to NA. We also set metered values to NA if the measured flow rate resulted in a percent
- 928 error greater than 2% of the flow rate, per manufacturer recommendation. Flow rates for each929 meter corresponding to this error threshold are summarized in Table S6.
- 930
- 931Table S6 Low-bound flow cutoff for accuracy less than 2%, calculated using Emerson MicroMotion online sizing tool. We aimed932to maintain flow rates within target ranges listed in Table 3, whereas these values represent lower bounds of meter accuracy
- 933 and are used for data cleaning only.

<u>Meter</u>	<u>Meter Model</u>	Flow Rate Accuracy Threshold
Small	CMSF015H	0.56 kg/hr
Medium	CMF050M	3.87 kg/hr
Large	CMFS150M	40 kg/hr

934

935 There are periodic gaps in the secondly metered data, which are filled in the final stage of meter 936 data cleaning. Communication with Emerson indicated that data gaps are likely caused by the transmitter power cycling due to unstable power supply the onsite portable generator. Table S7 937 938 summarizes the length of gaps in seconds and total number of occurrences across all metered 939 data, and total number of occurrences during non-zero flow rates. Gaps represent 2.26% of all 940 non-zero secondly measurements conducted during testing days, and all but 14 gaps had a 941 duration of 7 seconds or shorter. We fill gaps in data using a linear interpolation between the two measurements on either side of the gap. 942

943

944Table S7 Summary of gaps in Coriolis meter data across all days of testing. There were a total of 6,520 gaps in the data, of which9454,054 occurred during non-zero releases. These missing data made up 4.13% of all data, and gaps that occurred during non-zero946releases are 2.26 % of total data. Total number of secondly measurements in the dataset is 713,975 and total number of

947 seconds of missing data (including those occurring during zero releases) is 29,543 seconds. The longest gap length is 244

948 seconds, or about 4 minutes, and occurred during a zero-release. The longest gap during a non-zero release was 16 seconds.

Gap Length (seconds)	No. of Occurrences During	Total No. of Occurrences
	Non-Zero Releases	
1	1,444	1,747
2	2	2
3	928	1,538
4	0	2
5	0	1
6	2	2
7	1,664	3,202
8	0	0

9	0	0
$10 \le t < 20$	14	22
$20 \le t < 200$	0	2
$100 \le t < 200$	0	1
$200 \le t < 300$	0	1
Total No. of Gaps	4,054	6,520
Fraction of Total Data (%)	2.26%	4.13%

949

950 Whole gas flow rates are provided for each day of testing. For each secondly timestamp (in UTC), we include the release rate in kg/hr of whole gas, and list the meter that was used for the 951 952 measurement. We also include a data QC column to indicate which data was below the meter 953 accuracy threshold (2%) or an interpolated value as described above. In the raw data files for 954 each date, a QC flag of 1 indicates a non-testing period, 2 indicates non-original interpolated 955 data, and 3 indicates the flow range was below the accuracy level of the meter in use. Note that 956 non-original interpolated data can exist outside of testing periods, in which case the OC flag 957 would still be 1, whereas Table S7 summarizes gaps in data on testing dates during both testing 958 and non-testing periods of testing days.

959

960 1.2.3 Gas compositional analysis

We converted from whole gas flow rate to methane flow rate using gas composition data. Two 961 gas sample canisters were collected and analyzed by an independent laboratory from each tank of 962 963 gas, and gas composition data were also collected from two measurement stations upstream of the CNG fill point (see Supplemental Information Section S1.2.3. for more details). In the field, 964 965 we noted each time the CNG trailers were filled, and the point at which we switched to using gas 966 from a new supply. Trailer fill dates were matched with the gas compositional data. To account 967 for variability in gas composition and latency between the measurement station and the CNG 968 station, we average the data from both measurement stations for the five days leading up to and 969 including the date of a given truck refill. The standard deviation across this period is used for 970 determining uncertainty associated with gas composition. Mean mol% CH₄ over the study period 971 is 94.53% and the standard deviation is 0.62%.

972

We converted from whole gas flow rate to methane flow rate using gas composition for the CNG supply used in this experiment. In the field, we noted each time the CNG trailers were filled, and the point at which we switched to using gas from a new supply. Trailer fill dates and usage are then matched with the corresponding compositional analysis. We then used two methods for determining gas composition: measurement station compositional data and laboratory sample analysis.

979

All primary analysis uses gas compositional data from the two nearest upstream measurementstations on the pipeline that supplied the station at which our gas trailers refilled. We obtained

981 stations on the pipeline that supplied the station at which our gas transfermed, we obtained 982 datafiles for gas composition provided by the gas supplier that serves the CNG station. To

account for variability in gas composition and latency between the measurement station and the

account for variability in gas composition and latency between the measurement station and the

984 CNG station, we average the data from both measurement stations for the five days leading up to

and including the date of a given truck refill. We use gas composition on these days to determine
both mean methane fraction and the standard deviation, as an indicator of variability of gas

both mean methane fraction and the standard deviation, as an indicator of variability of gascomposition. In the field, we documented the start time of each new trailer batch, and thus

988 matched gas composition with specific releases. Table S8 shows the average gas composition

and standard deviation on specific dates of the experiment. These are the values used in the

990 primary analysis and ground truth meter data provided to participants.

991

Table S8 Gas composition for each truck refill, using data from upstream measurement stations. When we used a new truck at
 the start of the day, we set start time to 0:00 for simplicity in coding.

Batch	Batch Start Date	Batch End Date	Average Percent	Standard Deviation
<u>No.</u>	<u>(UTC)</u>	<u>(UTC)</u>	Methane	of Percent Methane
1	10/5/22 0:00	10/12/22 18:12	93.6%	0.162%
2	10/12/22 18:12	10/19/22 0:00	93.9%	0.196%
3	10/19/22 0:00	10/25/22 17:42	94.6%	0.241%
4	10/25/22 17:42	10/28/22 0:00	95.1%	0.137%
5	10/28/22 0:00	10/29/22 16:00	95.0%	0.136%
6	10/29/22 16:00	10/31/22 0:00	95.3%	0.461%
7	10/31/22 0:00	11/1/22 16:00	95.4%	0.365%
8	11/1/22 16:00	11/8/22 0:00	95.0%	0.269%
9	11/8/22 0:00	11/9/22 0:00	95.3%	0.132%
10	11/9/22 0:00	11/11/22 0:00	95.4%	0.166%
11	11/11/22 0:00	11/15/22 16:56	95.4%	0.203%
12	11/15/22 16:56	11/17/22 18:47	95.1%	0.127%
13	11/17/22 18:47	11/21/22 16:00	94.6%	0.528%
14	11/21/22 16:00	11/28/22 0:00	94.1%	0.093%
15	11/28/22 0:00	12/1/22 0:00	94.2%	0.100%

994

995 In the field, we had also collected samples from each CNG refill for analysis at Eurofins Air Toxics Laboratory. We sampled gas from the pressure regulation trailer by connecting laboratory 996 997 supplied canisters to the RT-30 sampling port (described above). Canisters were sent to Eurofins 998 Toxics Laboratory in Folsom, CA for gas composition analysis. Initially, we collected gas using 999 the vacuum gauge supplied by the Eurofins to indicate fill level. Canisters were connected to a 1000 vacuum gauge, and reading prior to fill was typically 25 - 30 Hg of vacuum. We opened 1001 sampling port to begin filling the canister and allowed the vacuum gauge to reach 0 Hg. After receiving laboratory results indicating gas pressure in the canister remained lower than 0 Hg, we 1002 1003 increased fill time to 1 minute and then to 5 minutes, switching methods at the dates listed in 1004 Table S9. Table S9 also summarizes fill method and time for each sample. We discarded all canisters that arrived at Eurofins laboratory with a pressure below 15 Hg vacuum. 1005

1006 1007

1008 Table S9 Fill method or time for each gas sample collected from the pressure regulation skid

Sampling Dates	Sample ID	<u>Fill Method</u>	
Oct 10 – Oct 29	01 through 12	Vacuum gauge reading	
Nov 1 – Nov 14	12 through 17	1-minute fill time	
Nov 14 – Nov 28	18 through 27	5-minute fill time	

- 1011 We requested quantification of the following compounds using Modified Method ASTM D-
- 1012 1945: methane, oxygen, nitrogen, carbon monoxide, ethane, ethene, acetylene, propane,
- 1013 isobutane, butane, neopentane, isopentane, pentane, and hydrogen. However, we ultimately
- 1014 chose not to use the gas compositional data reported by Eurofins for several reasons. First, for 1015 most trailer refills, we collected two samples from the same truck (see Table S10 below).
- 1016 However, Eurofins results for these replicate samples could be offset by up to 4%. Additionally,
- 1017 ten of the samples reported 100% methane, an unrealistic composition for compressed natural
- 1018 gas. For these samples, the sum of all analyzed constituents was greater than 100%. Finally,
- 1019 personal communications with Eurofins Technical Director indicated that the laboratory
- 1020 instruments have an uncertainty of +/-4.5% [95% CI], far greater than the observed variability in
- 1021 the more precise gas measurement station data described above. For these reasons, we consider
- the Eurofins results to be unreliable and opt to use the analysis provided supplier measurementstations.
- 1024

Table S10 compares the average percent methane for each truck refill using raw Eurofins data,
normalized Eurofins data, and measurement station data. Data from the measurement station
were reported with higher levels of precision than Eurofins data (five significant figures vs. two
significant figures). Also, the sum of all components analyzed in the measurement station data is
always within 0.002% of 100% while the sum of all components in the Eurofins data reached
over 105% in some samples.

1031

1032Table S10 Comparison of CNG gas percent methane using three methods: Eurofins raw reported values, Eurofins values1033normalized such that the sum of all constituents is 100%, and mean measurement station values. Where more than one Eurofins1034canister was collected per refill, we report the average of the two values. We did not collect a sample for Truck Refill 10, and the1035reported value for Eurofins data is the average of canisters 16, 17, and 18. Measurement station values are the average of the1036measurements of the two taps for the five days leading up to and including the date of refill (as summarized previously).

Refill	Eurofins	Eurofins Raw	Eurofins Normalized	Measurement
<u>No.</u>	Canister ID	<u>(% CH4)</u>	<u>(% CH4)</u>	Station (% CH ₄₎
1	01	94%	94%	93.636%
2	03	92%	92%	93.909%
3	05	90%	90%	94.602%
4	06	88%	88%	95.064%
5	08, 09	87%	87%	95.029%
6	10, 11	92%	92%	95.332%
7	12, 13	96%	93%	95.429%
8	14, 15	96%	93%	95.034%
9	16, 17	100%	96%	95.337%
10	NA	100%	95%	95.367%
11	18	100%	95%	95.389%
12	20, 21	100%	95%	95.094%
13	22, 23	100%	96%	94.637%
14	24, 25	100%	95%	94.112%
15	26, 27	99%	94%	94.227%

1040 1.3.1 Field testing conditions

1041

We tested five different aircraft technologies from October 10th through November 11th, 2022. 1042 Field measurement protocols were based on those previously reported to maintain consistency 1043 and comparability with other testing results (Rutherford et al., 2023). Operators were asked to 1044 1045 recreate typical or commercial flight operations as closely as possible. Each operator submitted key measurement parameters prior to the start of testing, including time necessary for 1046 measurement, planned flight lines, flight altitude, and predicted lower detection limit. Pre-1047 scheduled testing dates avoided simultaneous tests of technologies. However, due to scheduling 1048 1049 limitations on the operator side, supply-chain delays affecting equipment on the Stanford side, 1050 and flight-prohibitive weather conditions, this was not always possible and real-time adjustments 1051 to flight days were required. The final testing dates and flight altitude are depicted in Table 1 of the main text. 1052

1053

1054 For the spectroscopy-based technologies, we set and then held a release rate while the aircraft 1055 passed overhead, following a pre-planned flight trajectory. We typically held a constant rate for 1056 multiple overpasses of a given aircraft, aiming to change release rates at least two minutes before the next expected overpass, although this was not always possible for aircraft with shorter 1057 measurement times or on days when we tested multiple aircrafts at the same time. The Stanford 1058 1059 ground team tracked the GPS location of each aircraft being tested using the FlightRadar24 1060 mobile app. For Kairos Aerospace, we used a Spidertrack link provided by the company. While 1061 we documented timestamps when the aircraft appeared to directly overhead, all subsequent data processing used timestamps based on digital GPS tracking. For these spectroscopy-based 1062 technologies, a measurement occurs when the aircraft passes over the release site and reports the 1063 1064 overpass to Stanford. We use a measurement timestamp based on the moment when the GPS 1065 coordinates of the aircraft are closest to directly above the release stack.

1066

1067 Scientific Aviation conducts continuous data collection over a 20 - 40 minute time period, and 1068 an individual measurement refers to one such flight period. Because we were conducting multiple 20-minute releases throughout the testing period to test satellite-based methane sensing, 1069 1070 and we created a fixed release schedule to align satellite releases with Scientific Aviation measurements (included in Table S11). To avoid providing Scientific Aviation with any potential 1071 1072 information about release rates, we did not indicate which release periods aligned with those of 1073 the satellites. The Scientific Aviation aircraft arrived onsite for the start of the release period, and 1074 conducted measurements for the length of time determined by the Scientific Aviation scientific 1075 team on board the aircraft. If they completed all necessary measurements before the end of the 1076 release period, they left the site to return for the start of the next release period. Based on input from Scientific Aviation, the initial release duration was set to 40 minutes. On November 11th, 1077 1078 this was reduced to 35 minutes.

1079

For Scientific Aviation, a measurement refers to the period over which the aircraft was in
proximity of the release point and collecting data, with start and end times as submitted in the
Scientific Aviation results report.

1083

1084 Table S11: Coordinated release schedule for Scientific Aviation testing. The Stanford team set a new release rate and the time 1085 indicated on the table, and Scientific Aviation would arrive onsite and begin measurement protocols shortly thereafter. If 1086 Scientific Aviation completed all necessary measurements before the rate change, the plane left the immediate area to return

1087 measurement window in the schedule. The schedule was designed such that the Stanford team could simultaneously conduct

1088 satellite releases (20 minutes long) without providing information on which measurements were coincident with satellites to the 1089 Scientific Aviation team. All times are in local Arizona time.

November 8 th	November 10 th	November 11 th
14:35	11:00	12:00
15:15	11:40	12:35
15:55	12:20	13:10
16:35	13:00	13:45
	14:20	14:20
	14:50	14:55 – Refuel
		15:30
		16:10

1090

1091

1092 For all operators, the Stanford ground-team was in communication with the flight operations team via radio or text message. At the start of each scheduled flight day, the Stanford team 1093 would send an image of the sky overhead to operators to allow them to determine if onsite cloud 1094 conditions were conducive for measurement. Subsequent communication between the ground 1095 1096 team and flight operations team was kept to a minimum, and limited to the following topics: 1097 communication regarding clouds onsite or over the release point, or any local field disturbances and associated deviations from flight patterns. Table S12 documents all notable deviations from 1098 1099 flight patterns or field procedures.

1101 Table S12: Summary of all deviations from typical flight patterns or measurements

Datetime (UTC)	Operator	Notes
2022-10-26 16:58	Kairos Aerospace	Delay between measurements due to hot air balloon taking off within proximity of the field site
2022-10-28 19:04	Carbon Mapper	Aircraft conducted atypical circular flight pattern
2022-11-02 17:28	GHGSat-AV	Aircraft conducted atypical flight pattern; circling above field site due to cloudy conditions
2022-11-02 17:52	GHGSat-AV	Aircraft conducted atypical flight pattern; circling above field site due to cloudy conditions, testing for the rest of the day cancelled shortly after
2022-11-10 21:50	Scientific Aviation	Aircraft ended measurements early upon reaching fuel limit; Scientific Aviation team reported to Stanford team while in the field
2022-11-10 18:15	Scientific Aviation	Stanford team switched CNG trailer, causing an increase in flow rate associated with the sudden increase in trailer pressure; This measurement was removed through Stanford filtering

2022-11-11 23:03:12	Scientific Aviation	Power outage onsite, immediately shutting
		off all gas flow. Stanford field team
		informed Scientific Aviation field team that
		power had been cut, and the next
		measurement started with a delay to allow
		power restart

1.3.2 Description of Technologies Tested

We tested five different aircraft-based methane sensing platforms in this study: Carbon Mapper,
GHGSat-AV, MethaneAIR, Kairos Aerospace, and Scientific Aviation. As discussed in the main
text, all platforms except Scientific Aviation use spectroscopic imaging-based measurement,
while Scientific Aviation uses an in situ measurement approach. We requested sample plume
images from all test participants. At the time of this manuscript submission, only MethaneAIR
provided a plume image.

1.3.2.1 Carbon Mapper

In this study, Carbon Mapper operated the Global Airborne Observatory (GOA). This aircraft is equipped with a visible / infrared imaging spectrometer integrated on a Dornier aircraft. The spectrometer measures reflected solar radiation in the visible-to-shortwave infrared (380 - 2,510 nm) with 5-nm spectral sampling (Duren et al., 2019). At a flight height of 3 km (10,000 feet) above ground level, as in this study, the instrument typically has a 1.8-km field of view and 3-m pixel resolution (Duren et al., 2019). Data processing pipeline for Carbon Mapper is described in the Performer Info tab in the Carbon Mapper data report in the Github repository for this manuscript.

1123 Carbon Mapper, Inc. is a non-profit organization funded by philanthropy with the mission of 1124 identifying and tracking methane and carbon dioxide emissions (Carbon Mapper, 2023).

- *1.3.2.2 GHGSat-AV*

In this study, GHGSat flew a C-GJMT aircraft equipped with the GHGSat-AV2 sensor. The
sensor leverages similar technology to GHGSat's corresponding satellite sensor (GHGSat-Cx), a
wide-angle Fabry-Perot sensor with pixel resolution of <1 m (Esparza et al., 2023). At a flight
altitude of 3,000 meters, slightly above the 2,000 meter above ground level flown in this study,
GHGSat report a swath width of 750 m (Esparza et al., 2023). For details of the flight operations
for this study, see the Performer Info tab on GHGSat report submissions in the Github
repository.

- GHGSat Inc. is a private company that offers commercial methane detection services throughboth satellite and aircraft platforms (Esparza et al., 2023).
- *1.3.2.3 MethaneAIR*

- 1141 MethaneAIR is the aircraft-based precursor to the upcoming satellite mission MethaneSAT,
- 1142 developed by MethaneSAT, LLC (Chulakadabba et al., 2023). The MethaneAIR spectrometer
- measures methane enhancement in the 1,650 nm band and a 10m x 10m spatial resolution
- 1144 (Chulakadabba et al., 2023). Designed for wide spatial coverage, the instrument has a swath
- 1145 width of 4.5 km when flying at 12,960 m above ground level (Chulakadabba et al., 2023), the
- altitude used in this test. Additional details on the MethaneAIR measurement procedures and
 data processing are included in the MethaneAIR operator report in the GitHub repository. Figure
- 1148 S10 provides an example plume image output from this study, provided courtesy of
- 1140 S10 provides an example plume image output from this study, provided courtesy o 1149 MethaneAIR.
- 1150
- 1151 MethaneSAT, LLC is a wholly owned subsidiary of the nonprofit Environmental Defense Fund,
- with the mission of providing accurate and rapid quantification of methane emissions(MethaneSAT, 2023).
- 1154





Figure S10 Example methane enhancement and plume image from this study provided by MethaneAIR

1157



Kairos Aerospace measures methane using LeakSurveyorTM, an infrared imaging spectrometer 1160 with 3 m resolution (Sherwin, Chen et al., 2021). In previous testing, an aircraft flies at an 1161 altitude of 900 m (3,000 feet) above ground level, nearly twice the flight altitude of the current 1162 1163 study (Sherwin, Chen et al., 2021). Typical flight configuration involves a single wing-mounted unit for generating infrared and optical images during survey (Sherwin, Chen et al., 2021). In the 1164 1165 current study, Kairos operated with two measurement units, one mounted on each wing of the 1166 aircraft. Additional details on the quantification algorithm are included in Sherwin, Chen et al., 1167 2021 and further information on the Kairos measurement procedures and data processing for this 1168 study are included in the operator report in the GitHub repository. 1169 1170 Kairos Aerospace is a for-profit company that conducts aircraft surveys to identify oil and gas 1171 methane emissions, and facilitate rapid action and repair (Kairos Aerospace, 2023). 1172 1173 1.3.2.5 Scientific Aviation 1174 1175 Scientific Aviation uses an in situ measurement approached described in Conley et al., 2017. 1176 While conducting laps around an emission source, the aircraft collects ambient air and measures methane concentration using a Picarro cavity ring down spectrometer (Conley et al., 2017). In 1177 1178 this study, methane measurements were conducting using a Picarro 2210-m, and analysis of 1179 other compounds is feasible as well (discussed in the main text). Real-time analysis informs the 1180 number of laps and altitude (Mackenzie Smith, personal communication). 1181 1182 Scientific Aviation is a ChampionX research company that provides commercial methane detection services using aircraft, ground-based and drone platforms (Scientific Aviation, 2023). 1183 1184 1185 1.3.3 Data reporting and unblinding 1186

1187 Operators submitted results using a template provided by Stanford, subject to modifications by 1188 the operator as necessary. Timestamps for each individual measurement were documented by 1189 the Stanford field team and reported by operators. For Carbon Mapper, GHGSat-AV, and 1190 Methane Air, aircraft GPS coordinates and altitude were downloaded from FlightRadar24. 1191 Kairos Aerospace used Spidertrack for flight monitoring during testing, and provided positional 1192 and altitude data after the fact. Because different operators use different methods for reporting 1193 measurement timestamps, we use flight tracking GPS coordinates for consistency. Thus, timestamps of measurements refer to the moment when the distance between aircraft GPS 1194 1195 coordinates and the coordinates of the release stack are at a minimum. For Scientific Aviation, 1196 the Stanford team cannot independently ascertain when data collection occurring in the aircraft 1197 starts and stops. Thus, we used the measurement start and stop time as reported by Scientific 1198 Aviation in their data report.

1199

Overpasses documented by Stanford and on FlightRadar24 or Spidertrack that were not included
in the operator report are classified as "missing data" in figures of the main text. This occurred if
the aircraft flew above the release site, and thus was documented by Stanford ground team and
GPS tracking as an overpass, but no measurement was conducted by the field team. The Stanford

- 1204 ground team documented 711 airplane overpasses, of which 704 were matched with operator
- 1205 reported measurements. The number of missing measurements for each operator are summarized

in Table S13. All GHGSat-AV missing measurements were reported by the operations team to

1207 the Stanford team during testing. Carbon Mapper and MethaneAIR missing measurements were

identified after results were submitted, during data analysis. Kairos Aerospace and Scientific

1209 Aviation did not have any missing measurements. Because we do not have access to sensors or

1210 processing software used by the operators, we cannot determine reasons why any particular 1211 measurement would have failed.

- 1211
- 1212

Table S13 Total number of missing measurements by operator. A missing measurement occurs when an overpass is documented
 by GPS coordinates and Stanford field team observation, but no corresponding measurement is reported in operator results.

<u>Operator</u>	<u>Number of Missing</u> <u>Measurements</u>
Carbon Mapper	3
GHGSat-AV	2
Kairos Aerospace	0
MethaneAIR	2
Scientific Aviation	0

1215

1216

1217 Operators had the option to participate in a multi-stage unblinding process. For Stage 1,

operators submitted fully-blinded results report. Next, the operator was provided with Stanford
measured 10-meter wind data and allowed to submit revised quantification estimates. After
submitting Stage 2 revised results, operators were provided a subset of metered gas flow data. In
Stage 3, operators could use these data to make any additional revisions to their results for final
submission (described below).

1223

1224 1.3.4 Data processing for aircraft testing

1225

1226 For each operator, we set the gas release rate based on our desired sampling strategy. However, 1227 we were unable to set rates with full precision due to the technical limitations of our system. 1228 Thus, we identified a target release range while in the field, and manually documented flow rates 1229 for each aircraft measurement. Ideally, we would have an automated log of all set points, but did 1230 not because of software issues. As a result, we developed an automated method to determine changes in the release rate, which we describe here. Figures S11 and S12 shows outputs of the 1231 1232 plume definition algorithm for two days of testing (outputs for all days are included in the Appendix). One shows the plume definition criteria as applied to a test date when we used the 1233 automated feedback system to set the flow rate, and the other shows a date when we set the flow 1234 1235 rate using the valve position. 1236



Figure S11: Results of plume identification algorithm when system flow rate was set using an automated feedback (PID) system.



Figure S12: Results of plume detection algorithm when system flow rate is set using a fixed valve positions. Note change in y-axis compared to Figure S10.

1242

During aircraft testing, a single release rate was often held for multiple aircraft overpasses. The release rate was then changed directly to a new release rate, without allowing for a pause or zero release in between two different release rates. Here, we use the term "release" to refer to one such constant release rate, during which one or multiple aircraft overpasses and corresponding measurements may have occurred. Defining individual releases was necessary for providing aircraft operators with a subset of their data in Stage 3 of unblinding, described further below.

- 1249
- In order to define an individual release, we identify periods of steady flow rate given allowable
 tolerance for noise in meter measurement and flow variability. There are two sources of noise in
 meter readings which occur when the desired flow rate is set:
- 1253
- Meter noise: noise inherent to the instrument reading of gas flow rate. We quantified
 typical meter noise across each meter's calibrated range by holding steady release rates
 for several minutes and calculating the associated deviation from the mean (see Table

- 1257 S14) 1258
- Overcorrection-associated noise: from October 10th 20th, over-correction by the automated feedback system (Proportional Integral Derivative or PID controller), contributed to increased fluctuations in flow rate (an example of this is shown in Figure S6 above).
- 1263

We observed lower levels of meter-associated noise in the small (CMFS015H) and medium
(CMF050M) meters compared to the large (CMFS150) meter. Meter noise across all three
instruments was lower than the PID-associated noise, which displayed greater amplitude and
variability. Defining releases required us to separate meter and PID-associated noise from the
flow variability that occurs during the transition from one release rate to a new release rate. Thus,
for each meter we define a maximum allowable deviation function (see Table S14).

1270

Maximum deviation equations accounts for both meter and overcorrection-associated noise, and
are a function of the date, flow rate, and meter used for the measurement. For the small
(CMFS015H) and medium (CMF050M) meters, we selected values for maximum deviation that

1274 were three times the typical standard deviation observed over a 3-minute period. For the medium

1275 meter, we added a correction factor to account for the PID-associated variation, for periods when

1276 PID system was used. Variability in the large (CMFS150) meter increases at higher flow rates.

1277 The equation in Table S14 was determined using the standard deviation of flow rates ranging

- 1278 from 180 kg/hr to 1600 kg/hr over 1-3 minute periods.
- 1279

Table S14 Maximum allowable deviation in meter reading for defining individual releases. Equations are based on typical
 standard deviations observed in meters across their calibrated range.

Meter	Empirically Determined Deviation Function				
CMFS015H	0.12 * flow rate				
CMF050M	<i>If date is before 2022-10-20:</i>				
	5 + 0.2 * <i>flow rate</i>				
	<i>If date is after 2022-10-20:</i>				
	0.12 * flow rate				
CMFS150M	If flow rate \leq 700 kg/hr whole gas:				
	65				
	If flow rate >700 kg/hr whole gas:				
	-94 + 0.24 * flow rate				

1282

1283 To identify unique releases, we iterate through meter data. Logic for this function is summarized 1284 in Figure S13. First, we compare the flow rate at a given time point *t* with the mean flow rate 1285 since the end of the previous release. If the difference between the two rates is less than the 1286 maximum allowable deviation, the mean is updated to include time *t*, and we continue to the time 1287 t+1. When the function reaches a timepoint *t* where flow rate is greater than the allowable 1288 deviation, we may have reached the point that marks the end of one release rate and the

- 1289 beginning of another.
- 1290



 1291
 merge_release

 1292
 Figure S13 Flow chart for release categorization algorithm. The variable mean_rate is the average release rate for the current

 1293
 averaging period, rate(t) is the flow rate at time t, and rate_last is the mean flow rate of the last release. The input

 1294
 max_deviation refers to the calculated allowable deviation from mean flow rate, when considering noise from the meter and

 1295
 automated feedback system. Minimum duration (min_duration) is the minimum length of a release, set to 2 minutes. Max

 1296
 duration (max_duration) is the time difference between the end of the last release and the start of the current one, and is set to

 1297
 10 minutes to allow for merging of releases during periods of high variability caused by the PID-associated noise.

To determine if a new release is beginning, first we must check if the current time over which we
are averaging is a period of noise. We define individual releases to be at least 2-minutes in
length. Thus, if the averaging period is less than this minimum duration, we consider it to be
noise occurring in between true releases. The averaging start time is reset to *t*, and we begin
iteration again.

1303

1304 However, if the length of the averaging period is greater than two minutes, we implement one final check to determine if the averaging period is a distinct release. We compare the mean flow 1305 rate of the current averaging period with the mean flow rate of the previous release. If the 1306 1307 difference between the two is greater than the allowable deviation for the meter of the current 1308 release, the averaging period is considered a new release. If the mean flow rate of the current period is close to the mean flow rate of the previous release, as defined by the maximum 1309 deviation function, the two releases are instead merged. However, two releases will not be 1310 merged if the start of the second release occurs over 10 minutes after the end of the previous 1311 release, preventing the merging of plumes with similar release rates but with a time gap between 1312 them. When the current averaging period is determined to be a unique release, it is added to a list 1313 1314 of releases and the function continues to iterate through meter data to identify subsequent plumes. 1315

- 1316
- 1317 1.3.5 Stanford data quality control
- 1318
- **1319** *1.3.5.1 Spectroscopy-based technology data filtering*
- 1320

1321 Due to highly variable and frequently stagnant wind conditions observed during testing, we

developed a quality control (QC) criterion based on wind speed. The goal of this QC step was toensure gas from earlier releases will not contaminate measurements conducted by aircraft teams.

1324 Specifically, for a given aircraft overpass, we look at the gas released in the preceding ten

minutes and determine if the gas is from the current release or the previous release (releases
defined using the criteria above). Next, we identify the trajectory of the gas using 3-D wind data
collected onsite. Finally, we determine how much of the gas from any previous releases is within
a certain distance threshold of release source. If the amount of gas from previous releases and
within the distance threshold is greater than a certain percent of the gas from the current release,
the measurement is considered contaminated.

1331 1332

2 This criterion requires determine two specific parameters:

1333 1334

> 1335 1336

> 1337

1. Distance threshold: the distance in meters that gas from previous releases must travel in order to be considered *not* interfering with the current release.

2. Mass threshold: the maximum amount of gas from a previous release allowed within the distance threshold.

1338 1339 We made all quality control decisions, including setting the values of both mass and distance 1340 thresholds, prior to viewing any blinded data reported by aircraft operators. We determined technology specific distance thresholds for each aircraft operator based on our knowledge of 1341 their technology (see Table S15). We use a mass threshold value of 10%, meaning that 10% of 1342 1343 the mass within the distance threshold may be from previous gas releases. Because we only use 1344 wind transport in modeling the gas plume trajectory, and do not consider dispersion or diffusion, 1345 this approach is inherently conservative. Thus, we use a high mass threshold value of 10% as 1346 default in our analysis. However, after receiving operator results, we evaluated the error profile each operator using both 10% and 1% mass threshold values. We compared overpasses that 1347 failed with the 1% criteria but passed with the 10% criteria with those that passed both criteria, 1348 1349 summarized in Table S16. For all operators, we found no statistically significant difference between these two groups using a t-test, with a p-value of 0.05 (a threshold determined prior to 1350 1351 viewing any blinded data reports).

1353 Table S15 Distance threshold for each aircraft operator and information used in determining the	? value
--	---------

Operator	Distance Threshold	Source		
	<u>[m]</u>			
Carbon Mapper	300	Based on maximum fetch distance of 150 m		
		reported in Duren et al., 2019 supplemental		
		information, doubled to add buffer		
GHGSat	300	Based on plume images provided to Stanford		
		Team by GHGSAT during 2021 Controlled		
		Release testing, to be published in Rutherford et		
		al, 2023		
Kairos	500	Based on plume images from Sherwin, Chen et al.,		
		2021		
MethaneAIR	500	Based on plume images provided to Stanford		
		Team by MethaneAIR during 2021 Controlled		
		Release testing, included in Chulakadabba et al,		
		2023.		

1355 Table S16: Summary of quality control comparison imaging technologies, including number of measurements to pass both the

1356 10% and 1% mass thresholds. The threshold comparison column includes the calculated p-value when comparing operator

quantification estimates for two groups of measurements: those that passed the 1% mass threshold, and those that passed the
10% mass threshold but failed the 1% mass threshold. Because the threshold comparison value is greater than 0.05 for all
operators, we use the 10% mass threshold.

Pass 10% Mass Pass 1% Mass Threshold Operator Threshold (no. of Threshold (no. of **Comparison** measurements) measurements) (p-value) Carbon Mapper 67 71 0.48 **GHGSat-AV** 104 121 0.96 Kairos Aerospace 171 191 0.08 **MethaneAIR** 18 20 0.85

1360

1361

1362Figure S14 shows example overpasses that would pass or fail Stanford QC criteria. In both

images, we depict the trajectory of gas participles emitted in the ten minutes leading up to the

1364 overpass. Points in green represent gas particles emitted from the current release, whereas gas in

red represents gas from previous release(s). Within the set distance threshold, if the total mass of red particles is greater than 10% of the total mass of green particles, the overpass is considered

contaminated and filtered. In Figure S14, the image on the left passes Stanford quality controlfiltering while the image on the right fails.

1369



1371 1372

Figure S14 Sample releases that pass (left) and fail (right) Stanford's quality control criteria. Colored points represent gas released in the last 10 minutes. Particles colored green are from the current release, and gas colored red are not.

We did not use this wind exclusion criteria for Scientific Aviation, due to the different technological approach to measurement. Rather, we excluded any plumes where the standard deviation of the flow rate was greater than 10% of the mean flow rate. Only one release was removed in this manner.

1378 *1.3.5.2* In situ technologies data filtering

1379

Due to the difference in measurement approach, for Scientific Aviation we excluded points based
on variability in flow rate. We considered the entire measurement period, as reported by the
operator, and excluded any measurements where the standard deviation over this period was
greater than 10% of the mean flow rate for the same period.

1384

1385 Using this method, Stanford filtered one measurement from the Scientific Aviation dataset. The 1386 high variability in flow rate was the result of changing gas trailers mid-release. While conducting 1387 the release, the Stanford ground team observed a steady drop in flow rate, as indicated by the laptop dashboard. Upon communicating with the Rawhide personnel operating gas equipment, 1388 1389 we learned this was caused by falling pressure in the CNG supply trailer. The Rawhide personnel 1390 switched to the other trailer, which was full and at a much higher pressure. As a result of the 1391 increased pressure on the system while maintaining a fixed position of the gate vale, pressure 1392 jumped when the source trailer was switched. We attempted to slowly return the flow rate to the

- previous level, resulting in the slope observed in the second half of the release in Figure S15.
- 1394



 1395

 1396

 Figure S15: Release for Scientific Aviation filtered by Stanford due to high variability in flow rate (standard devation was greater

 1397

 than 10% of the mean flow rate)

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- 1399
- 1400 1.3.6 Stage 3 data selection
- 1401

In Stage 3, our goal was to provide operators with ground-truth data for roughly half of theiroverpasses, allowing them to revise and resubmit estimates for their remaining blinded

1404 measurements. We provided operators with a list of overpass timestamps, as documented by the

- 1405 Stanford field team, with the associated 60-second time-averaged methane flow rate (in 1406 $kg(CH_4)/hr$). Due to delays in acquiring detailed information on interpreting the irregularities in
- 1406 kg(CH4)/m). Due to delays in acquiring detailed information on interpreting the megularities in 1407 the gas compositional vales discussed above, we used raw Eurofins data for Stage 3 methane
- 1407 the gas compositional vales discussed above, we used faw Euroritis data for Stage 5 methane 1408 mole fraction. Upstream measurement station data were only acquired after completion of Stage
- 1409

3.

- 1410
- 1411 Stage 3 data were selected to provide roughly half of all overpasses that meet Stanford quality
- 1412 control criteria, while also ensuring the subset is representative of the flow rates for all1413 overpasses. Additionally, we did not want to provide operators with any additional information
- 1414 about overpasses that remained blinded. Thus, when an overpass from one release was selected,
- 1415 we also provided data on all other overpasses that occurred during the same release. With this
- approach, we avoid a situation in which we provide an operator with overpasses at the start and
 end of a release, but not those in between a scenario that could provide operators with
- 1418 information on the true release rate for the middle overpasses that remained blinded.
- 1419
- 1420 In order to select which releases to unblind for operators, we first generated a cumulative
- 1421 probability function of the flow rates for all releases conducted for each operator. We then
- 1422 uniformly selected a number of points on this distribution function equal to half of the total
- 1423 number of releases (if we conducted N releases for a given operator, we selected N/2 points on
- the cumulative probability function). For each probability selected, we then determined the
- 1425 closest maximum gas flow rate with a corresponding cumulative probability lower than the
- selected probability. This results in a selection of roughly 50% of the releases with a very similarto that of the total distribution.
- 1428
- 1429 Finally, for each release, we then identified all corresponding operator overpasses. Because the
- number of overpasses varies per release, operators were provided with time-averaged flow ratesfor close to but not exactly half of their all overpasses. Figure S16 depicts a sample release
- 1431 distribution for all spectroscopy-based aircrafts (including MethaneAIR, who did not participate
- 1433 in Stage 3 analysis).
- 1434



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Figure S16 Cumulative probability distribution each operator (including MethaneAIR, who did not participate in Stage 3 of unblinding). We selected a number of points on the distribution function equal to half the total number of releases conducted for the operator. We then determined the corresponding release rate, and provide operators with releases representative of their total distribution. Panel A shows the distribution of all releases. Panel B shows the distribution of releases selected for unblinding in Stage 3.

1442 2 Supplementary Results

1443

1444 2.1 Aircraft data reporting

1445

1450

All aircraft were requested to use typical data collection and analysis procedures to best represent
real world operations, including for any quality control filtering. Several operators raised
concerns regarding stagnant wind conditions onsite during testing. Table S17 summarizes the
timeline for reporting results for all three stages.

Operator	Testing	Stage 1	<u>Start</u>	Stage 2	<u>Start</u>	Stage 3
	Complete	Submitted	Stage 2	Submitted	Stage 3	Submitted
Carbon	10/31/22	01/03/23	01/11/23	2023-02-13	02/15/23	02/28/23
Mapper						
GHGSat	11/07/22	11/21/22	12/22/22	12/23/22	02/15/23	02/17/23
Kairos	10/28/22	11/17/22	12/19/22	12/20/22	02/15/23	02/23/23
Methane	10/29/22	03/22/23	NA	NA	NA	NA
Air						
Scientific	11/11/22	02/21/23	NA	NA	NA	NA
Aviation						

1451 Table S17: Operator data reporting timeline

1453

1454 2.1.1 Carbon Mapper

1455

1456 Carbon Mapper apply filtering criteria to determine if a measurement was high enough quality for quantification. Prior to submitting any results, Carbon Mapper requested the ability to change 1457 1458 their filtering with each stage of data submission, based on new data provided during unblinding. 1459 For full transparency, Carbon Mapper included quantification estimates for all measurements in 1460 each stage, including those that failed their filtering criteria. These are available in the original operator data reports. In Stage 2, unblinded wind data did not change the quality control filtering 1461 of any measurements. However, in Stage 3, two measurements filtered from both Stages 1 and 2 1462 1463 passed the filtering criteria and were included in quantification estimates.

1464

1465 All Stanford analysis only includes measurements that pass the Carbon Mapper filtering. As a 1466 result, the results classification histogram and probability of detection plots in the main text detection capabilities for measurements that Carbon Mapper has confidence in quantifying. We 1467

1468 use this approach for consistency when comparing with other operators, who did not distinguish

if their filtering applied different to detection vs. quantification. 1469

1470

2.1.2 1471 Kairos

1472

1473 To determine variability in their measurement instruments, Kairos flew with two measurement 1474 units during testing, one on each wing. They submitted two separate data reports, one for each 1475 measurement unit. In the main text, we consider the detection and quantification performance of the entire deployed system, rather than reporting results for each unit individually. If either unit 1476 1477 detected an emission, we consider that a detection. We report quantification estimates using the average of two reported units. If one unit reported values and the other did not, either due to a 1478 1479 non-detect or QC filtering, we use the value for the reporting unit. Individual analysis for each 1480 measurement unit is included in Supplementary Information.

1481

1482 2.1.3 **MethaneAIR**

1483

1484 MethaneAIR uses two methods for analysis, described previously in Chulakadabba et al. (2023): divergence integral (DI) and modified integrated mass enhancement (mIME). In their main 1485 1486 unblinded results submission, they report the average value of the two quantification methods. Results for each independent method are included in Supplemental Results. 1487

1488

1489 Scientific Aviation 2.1.4

1490

1491 Four of these measurements were all conducted on November 8th, when wind conditions were 1492 not conducive towards measurement. The Scientific Aviation flight team informed Stanford on 1493 Nov 8th after testing was completed that the measurements were affected by wind conditions that 1494 blew the plume towards extremely tall power lines which prevented them from using the ideal flight path. Two additional points were due to too few laps conducted in the field (on November 1495 10th and 11th), and an additional measurement was discarded on November 11th because not 1496

- 1497 enough of the plume near the ground surface was captured.
- 1498

- 1499
- Additional analysis of operator-reported results 1500 2.2
- 1501 1502
 - Quantification Accuracy: error profile and best-fit residuals 2.2.1
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Carbon Ma Partially u {n=34} Carbon Mappe Fully blinded (n=71) (% t Quantification Error (%) tification Errol 10 oue -10 Perc -20 -20 -30 400 600 800 1000 1200 1400 Methane Release Rate (kg/hr) 400 600 800 1000 1200 1400 Methane Release Rate (kg/hr) 400 600 800 1000 1200 1400 Methane Release Rate (kg/hr) GHGSat Partially unb (n=63) (%) (%) (% Error Quantification Erro Percent Quantification Errol 10 10 Ouantific -10 Perc 400 600 800 1000 1200 Methane Release Rate (kg/hr) 1400 400 600 800 1000 1200 Methane Release Rate (kg/hr) 400 600 800 1000 1200 Methane Release Rate (kg/hr) 1400 1400 Kairo Unb (n= Kairos Fully bl (n=124 (n=49) (% (%) Percent Quantification Error (%) Quantification Error Error 100 100 ent Quantification 310 -10 Perce -20 -20 400 600 800 1000 1200 Methane Release Rate (kg/hr) 1400 400 600 800 1000 1200 Methane Release Rate (kg/hr) 400 600 800 1000 1200 Methane Release Rate (kg/hr) 1400 Scientific Aviation Fully blinded resu (n=8) Methane Fully bli (n=18) (%) Quantification Error (%) Quantification Error 100 -10 -20 -300 400 600 800 1000 1200 1400 Methane Release Rate (kg/hr) 20 400 600 800 1000 1200 Methane Release Rate (kg/hr)







Figure S18: Percent quantification error for all points included in parity plots in the main text



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Figure S20 Percent quantification error for measurements with metered methane release rates greater than 50 kg / hr. Note yaxis is adjusted compared to previous percent quantification error for improved clarity. Value of n represents all data points including those beyond the range of the x-axis in this plot.







1520
1521 Figure S22 Percent error of residuals for linear best fit on all aircraft quantification estimates. Calculated as the difference
1522 between operator quantification estimate and linear best fit value, as a fraction of the linear best fit value.

- 1523 2.2.2 Kairos individual pod analysis
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Kairos Aerospace flew with two measurement pods, one attached to each wing and reported results for each pod individually. In the main manuscript, we averaged both values. Where one pod reported a non-zero release and the other did not (either reporting a zero, or filtering through quality control), we use the single reported value. Here, we use the individual reported pod

1528 quality control), we use the single reported value. Here, we use the individual reported pod

values to generate the same figures reported in the main manuscript for quantification accuracy

and lower detection limit.









2.2.3 Kairos wind normalized probability of detection

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1538 Kairos Aerospace typically reports quantification estimates using wind-normalized flow rate, as 1539 in Sherwin, Chen et al., 2021. Here, we produce probability of detection plots using units of kg 1540 methane per hour per meter per second to allow for comparison with previous testing. In the 1541 Kairos operator results report, they include a column with the Dark Sky wind speed used in 1542 determining the quantification estimate. We divide their reported flow rate by the reported wind

same bins reported in Sherwin, Chen et al., 2021. In the flight configuration tested previously,

- Kairos Aerospace did not detect the one release smaller than 5 kgh / mps, and only detected 1 of
 14 releases between 5 10 kgh / mps, and 8 of 12 releases between 10 and 15 kgh / mps. In the
- 1547 current study, Kairos detected all releases above 1 kgh / mps.
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1550Methane Release Rate (kgn/mps)Methane Release Rate (kgn/mps)1550
1551Figure S25 Kairos probability of detection using wind normalized methane flow rate, for comparison with Sherwin, Chen et al.,
2021. Kairos reported wind speed from Dark Sky used in each quantification estimate in their Stage 1 report, and we use this as
the input wind value for normalization. Figure on the left has x-axis that match Sherwin, Chen et al., 2021 for ready comparison
between the two tests. Figure on the right shows all releases below 5 kgh/mps.

1554 2.2.4 MethaneAIR Results for DI and mIME Methods

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As described in the main text, MethaneAIR reported results that are the average of two different methodological approaches: divergence integral (DI) and modified methane mass enhancement (mIME). Here we provide parity plots for the individual methods. For each method, MethaneAIR reported 24 measurements. However, the mIME method detected two non-zero releases that were not detected using the DI method. Hence, the total number of points in Figure S26 differ by this amount.



Figure S26: MethaneAIR quantification results for mIME and DI methods

1564	
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1614 4 Appendix

- 1615 4.1 Plume release definition
- 1616

1617 Here we present the plume definitions as determined using the algorithm described in SI Section

1618 S1.3.3. Plume definitions were used to determine which measurements to unblind in Stage 3.1619



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	Plume definition algorithm results for 10/31																_																										
ite (kgh) 1500 1750 2000		Plume 1 (~ 219 kg/h)	Plume 2 (~ 320 kg/h)	Plume 3 (~ 115 kg/h)	Plume 4 (~ 63 kg/h) Plume 5 (~ 115 kg/h)	Plume 6 (= 697 kg/h)	Plume 7 (= 23 kg/h) Plume 8 (= 287 kg/h)	Plume 9 (= 330 kg/h)	Plume 10 (= 273 kg/h)	Plume 11 (~ 1024 kg/h)	Plume 13 (~ 175 kg/h)	Plume 14 (~ 1168 kg/h) Plume 15 (~ 19 kg/h)	Plume 16 (~ 652 kg/h)	Plume 17 (= 24 kg/h)	Plume 18 (= 483 kg/h)	Plume 19 (~ 0 kg/h)	Plume 20 (= 649 kg/h)	Plume 21 (= 168 kg/h)	Plume 22 (= 588 kg/h)	Plume 23 (~ 2 kg/h) Plume 24 (~ 7 kg/h)	Plume 25 (~ 0 kg/h)	Plume 26 (~ 347 kg/h) Plume 27 (~ 1 kg/h)	Plume 28 (= 1174 kg/h)	Plume 29 (= 95 kg/h)	Plume 30 (= 0 kg/h)	Plume 31 (~ 3 kg/h) Plume 32 (~ 330 kg/h)	Plume 33 (~ 8 kg/h)	Plume 34 (~ 886 kg/h)	Plume 35 (= 10 kg/h)	Plume 36 (= 401 kg/h)	Plume 37 (~ 80 kg/h)	Plume 38 (~ 0 kg/h)	Plume 39 (= 244 kg/n) Plume 40 (= 1119 kg/h)	Plime 41 (= 2 kr/h)	(v)v4 VV - J CV emild	Plume 43 (~ 11 kg/h)	Dlime AA (~ 0 ko/h)	Plume 45 (~ 138 kg/h)	Plume 46 (= 489 kg/h)	Plume 47 (~ 45 kg/h)	Plume 48 (~ 0 kg/h)	Plume 49 (~ 543 kg/h)	
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Time (UTC)

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Metered Release Rate (kgh) 1000 1500 2000 2500

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17:00

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Plume 1 (~ 352 kg/h)

Plume 4 (~ 511 kg/h) Plume 5 (~ 24 kg/h)

17.17 17.25 17.30

Plume 6 (~ 424 kg/h)

Plume 3 (~ 700 kg/h)

Plume 2 (~ 0 kg/h)

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Plume 7 (= 381 kg/h) Plume 8 (= 23 kg/h)

17.43 17.52 17.55 Plume 9 (= 35 kg/h)

1635 1636 Plume definition algorithm results for 10/29

Plume 10 (= 356 kg/h) Plume 11 (= 145 kg/h) Plume 12 (= 859 kg/h) Plume 13 (= 603 kg/h)

18:22

Plume 15 (~ 450 kg/h) Plume 16 (~ 335 kg/h) Plume 17 (~ 1372 kg/h)

19.19 19.25 19.31 19 40 19 46 19 50 20:01 20:09

Plume 18 (~ 798 kg/h) Plume 19 (~ 81 kg/h) Plume 20 (~ 852 kg/h) Plume 21 (~ 610 kg/h) Plume 22 (= 68 kg/h) Plume 23 (= 214 kg/h)

Plume 14 (~ 1134 kg/h)

Plume 24 (~ 141 kg/h) Plume 25 (~ 62 kg/h) Plume 25 (~ 62 kg/h) Plume 26 (~ 62 kg/h) Plume 27 (~ 519 kg/h) Plume 28 (~ 129 kg/h) Plume 28 (~ 129 kg/h) Plume 32 (~ 43 kg/h) Plume 33 (~ 143 kg/h) Plume 33 (~ 71 kg/h)

20.21 20.37 20.37 20.43 20.43 20.43 21.02 21.06 21.12





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1649 4.2 Daily release

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Plots of daily release rates for each day of testing for each aircraft. Vertical lines represent a
measurement, color indicates quality control filtering. For spectroscopy-based technologies,
height of vertical lines represent average release rate for the 1-minute period prior to aircraft
overpass.











Height of each bar represents the average release rate over the entire measurement period, using
start and end points reported by Scientific Aviation. The location of the bar indicates the
Scientific Aviation measurement start period.

