

A versatile spaceborne architecture for immediate monitoring of the 2 global methane pledge

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32 Abstract.

33 The global methane pledge paves a fresh, critical way toward Carbon Neutrality. However, it remains largely invisible and 34 highly controversial due to the fact that planet-scale and plant-level methane retrievals have rarely been coordinated. This has 35 never been more essential within a narrow window to reach the Paris target. Here we present a versatile spaceborne architecture to address this issue. Using this framework, we patrol the world, like the United States, China, the Middle East, and North 36 37 Africa, and simultaneously uncover methane-abundant regions and plumes. These include new super-emitters, potential 38 leakages, and unprecedented multiple plumes in a single source. More importantly, this framework is shown to challenge official emission reports that possibly mislead estimates from global, regional, to site scales, particularly by missing super-39 40 emitters. We reveal that this framework can enable ready-made satellites to initiate monitoring of the global methane pledge

41 immediately and is also versatile for upcoming stereoscopic measurements and artificial intelligence techniques.





42 1. Introduction

Global methane pledges finalized at the COP26 (the 26th United Nations Climate Change Conference of the Parties) have 43 been never more ambitious (Schellnhuber et al., 2016; Schurer et al., 2018; United Nations, 2021). More than 100 countries 44 have promised 30% methane emission reductions by 2030. Also, energy giants (e.g., Shell and BP) have committed to clear 45 46 targets of methane mitigation. Such pledges have never been more essential within a narrow window (< ten years) to reach the 47 Paris target. The scientific context is that atmospheric methane is a powerful greenhouse gas second only to carbon dioxide (CO_2) , trapping ~ 80 times more heat than the same amount of CO_2 (per molecule) over a 20-year time horizon (Etminan et 48 49 al., 2016; Saunois et al., 2016, 2020). Worse still, it is thought to rise since 2007 (Mikaloff and Hinrich, 2019), surge since 50 2014 (Nisbet et al., 2019), and set another record in 2021 (National Oceanic and Atmospheric Administration, 2022). 51 Fortunately, methane is short-lived (~ ten years) (K. et al., 2013) and can be reduced in half using existing technologies (Ocko 52 et al., 2021).

However, a classic dilemma emerges, dimming the hopes of scientists and policymakes (Masood and Tollefson, 2021). That is, on the eve of the Paris target, those targets and emissions remain largely invisible worldwide and thus hinder effective mitigation. The main issue is the Paris framework relies on countries or corporate giants to report emissions (A. et al., 2018, 2015; Ganesan et al., 2019). Moreover, the reports are based on indirect statistics, such as O&G inventories, rather than direct measurements(Deng et al., 2022). This leads to a broad consensus that prominent discrepancies exist between the reports. For example, field campaigns nearly double official claims of methane emissions in the United States by correcting leak detection(A. et al., 2018).

To this end, widespread super-emitters present a unique opportunity worldwide (Duren et al., 2019; Pandey et al., 2019; T. et 60 61 al., 2022; Zavala-Araiza et al., 2015, 2017). They are typically responsible for the underestimates of methane emissions (Alvarez et al., 2018; Duren et al., 2019; Itziar et al., 2021; T. et al., 2022; Thompson et al., 2016). Moreover, there is increasing 62 evidence that methane emissions follow a heavy-tailed distribution (Duren et al., 2019; Frankenberg et al., 2016; T. et al., 63 64 2022), for which relatively small number of sources (so-called super-emitters) can account for a disproportionately large share 65 of total emissions. In contrast to area sources (e.g., cities), super-emitters are typically coal mines, wells, gathering stations, 66 storage tanks, pipelines, and flares, with even less than dozens of metres in diameter but high-concentrated methane plumes 67 (Allen et al., 2013; Miller et al., 2019; Subramanian et al., 2015; Varon et al., 2019). We thus anticipate that significant emission mitigation could be achieved by deploying well-designed systems to identify methane super-emitters. For instance, 68 69 in support of the Paris agreement, the 17th World Meteorological Congress (2015) requested an Integrated Global Greenhouse 70 Gas Information System (IG3IS) that aimed to develop a measurement framework for methane emission reductions (Phil 71 DeCola and WMO Secretariat, 2017).

72 To date, a large body of field measurements (e.g., in situ and aircraft surveys) between 2012 and 2020 has been designed for

- 73 methane super-emitters. Despite this, they are spatially confined (e.g., regionally) and temporally infrequent (e.g., a few weeks),
- ⁷⁴ incapable of exploring global methane super-emitters (A. et al., 2018; Conley et al., 2016; Duren et al., 2019; Marchese et al.,





2015; Nisbet et al., 2020; Smith et al., 2017; Thompson et al., 2016; Thorpe et al., 2016). Today, substantial advances have 75 76 been made towards detecting and quantifying methane super-emitters from space (Cusworth et al., 2019; Hu et al., 2018; Itziar 77 et al., 2022; Jacob et al., 2016; Pandey et al., 2019; Thompson et al., 2016). Such advances, however, have rarely been expanded 78 to measure the global methane pledge because large-scale swath and high-resolution sampling have not been coordinated. First, 79 global methane monitoring has become possible. A flagship satellite mission is the TROPOspheric Monitoring Instrument 80 (TROPOMI) onboard the Copernicus Sentinel-5 Precursor satellite (T. et al., 2022; Veefkind et al., 2012). It can offer daily 81 global insights for methane column concentrations, with a large swath width of ~ 2600 km, a moderate resolution of 7.0×5.5 82 km² (since August 2019), and high signal-to-noise ratios. However, its relatively coarse spatial sampling still limits its 83 application to detect methane super-emitters (T. et al., 2022). Second, next-generation satellite missions, pioneered by the 84 GHGSat constellation (three satellites at the moment), emerge for mapping methane super-emitters (Cusworth et al., 2019), 85 with a narrow swath (e.g., ~ 12 km) but a ground-breaking high-resolution spatial sampling (e.g., $25 \sim 50$ m)(Jervis et al., 2021; 86 Varon et al., 2020). Complementary to the GHGSat constellation, satellite-based hyperspectral imager spectrometers, such as 87 PRISMA, Gaofen-5, ZY1, Sentinel-2, and Worldview-3, have shown great potentials (Guanter et al., 2021; Itziar et al., 2021; 88 Sánchez-García et al., 2021; Varon et al., 2021). They can resolve methane enhancements and attribute them to specific 89 infrastructures via similar narrow swath and high-resolution sampling (e.g., 30 m). Note that regions those satellites usually 90 gazed at are originally well-known home to methane super-emitters. Narrow swath coverage thus remains a crucial limitation 91 for global surveys of methane super-emitters. Collectively, existing satellite missions still lack both global vision and keen 92 insight and thus cannot sustain the global methane pledge. Here we present a multi-tiered, space-based framework for global-scale and high-resolution methane retrievals. The key is that 93

ready-made satellite missions alone have the potential to initiate immediate monitoring of the global methane pledge. Using this framework, we patrol the world, with an experimental focus on China, the United States, Iraq, Kuwait, and Algeria, and reveal both region-scale hotspots and plant-level super-emitters. We can even gaze at a single source to map multiple plumes and inspect possible methane leakages. These results can challenge national reports that possibly miss unexpected superemitters or mislead emission magnitude. On the eve of the Paris target, at least while a global methane monitoring network is not in place, this multi-tiered satellite constellation presented in this study has important implications for measuring global methane pledges. Further information on methane retrievals and emission estimates, as well as uncertainty analysis, are shown

101 in Materials and Methods.

102 2. Materials and Methods

103 2.1 Multi-tiered satellite constellation

The multi-tiered satellite constellation was designed to reconcile global-scale and high-resolution methane monitoring. First, TROPOMI offered a unique potential for global methane monitoring, depending on its large-scale (i.e., 2600 km) swath, daily revisit time, regional footprint (i.e., $5.5 \times 7 \text{ km}^2$ since August 2019), and sounding precision and accuracy (i.e., < 1%)





(Veefkind et al., 2012). Approximately, TROPOMI observed a full swath per second, which resulted in \sim 216 spectra per 107 108 second. This instrument comprised two spectrometer modules, the first involving near-infrared (NIR) spectral channels, and the second dedicated to the shortwave-infrared (SWIR) spectral channel. The NIR and SWIR channels were equipped with 109 110 spectral resolutions of 0.38 and 0.25 nm and spectral sampling ratios of 2.8 and 2.5, respectively. Since the NIR and SWIR detectors are incorporated in different instrument modules, the NIR spectra will be co-registered with the SWIR spectra before 111 112 performing methane retrievals. The methane total column-averaged dry-air mole fraction (XCH₄) is retrieved from near-113 infrared (NIR) (757 ~ 774 nm) and shortwave-infrared (SWIR) (2305 ~ 2385 nm) spectral measurements for sunlight backscattered by Earth's surface and atmosphere (Hu et al., 2018). In this study, only high-quality measurements, retrieved 114 115 under cloud-free and low aerosol load conditions, were used. These measurements were filtered, in addition, for solar zenith angle ($<70^{\circ}$), low viewing zenith angle ($<60^{\circ}$), and smooth topography (the surface elevation of <80 m within 5 km radius) 116 117 as described in Hu et al. (28) (Hu et al., 2018).

118 Hyperspectral satellite missions severed as the second tier, responsible for mapping localized methane super-emitters 119 depending on their unprecedented resolution (i.e., 3m ~ 50m). Therein PRISMA, as an open-access representative, was 120 specifically suitable for this work. It can image the solar radiation reflected by the Earth's surface and atmosphere via hundreds of spectral channels between the visible and SWIR spectrum (~ 400 ~ 2500 nm). Measurements in the SWIR spectrum from 121 122 2000 to 2500 nm sampled absorption features from water vapor, carbon dioxide, and methane. Therein the 2100 nm and 2450 123 nm windows were especially sensitive to methane. Furthermore, the signal-to-noise ratio was reported to be about 100 in the 124 SWIR for a relatively dark vegetation pixel and increased to above 200 for bright soil surfaces in oil and gas extraction sites. More importantly, it covered areas of 30×30 km² with a 30 m spatial sampling. 125

We collected dozens of daily measurements from the multi-tiered satellite constellation. These measurements experimentally mapped regional methane hotspots and localized methane super-emitters across the United States, China, the Middle East (Iraq and Kuwait), and North Africa (Algeria). The acquisitions were mostly taken between April 2020 and January 2022.

129 2.2 Multi-tiered methane retrievals

130 In the first tier of our framework, we employed the operational methane products via TROPOMI onboard the Sentinel 5 satellite. The target product was the column-averaged dry-air volume mixing ratio of methane (XCH₄), which will be retrieved 131 132 simultaneously with scattering properties of the atmosphere. The operational retrieval algorithm is based on RemoTeC (Butz et al., 2009; Hasekamp and Butz, 2008), which was originally developed for CO_2 and methane retrievals from GOSAT 133 observations (Butz et al., 2011). It attempted to fit spectra observed by the TROPOMI-based NIR and SWIR channels. Its 134 135 sensitivities to atmospheric scattering properties, atmospheric input data, and instrument calibration errors had been extensively evaluated (Sha et al., 2021; Verhoelst et al., 2021). As a result, the operational products were proved to be critically 136 137 stable, with a convergence rate of 99%, and high significance by comparisons with both satellite-based (e.g., GOSAT) and ground-based (e.g., TCCON) measurements. The required accuracy and precision of < 1 % for the XCH₄ product were met 138

139 for clear-sky measurements over land surfaces and after appropriate filtering of difficult scenes. Moreover, the forward model





error was less than 1 % for about 95 % of the valid retrievals. Model errors in the input profile of water did not influence the retrieval outcome noticeably. The methane product is expected to meet the requirements if errors in input profiles of pressure and temperature remain below 0.3% and 2 K, respectively. Of all instrument calibration errors, the retrieval results were the most sensitive to an error in the instrument spectral response function of the shortwave infrared channel.

- To achieve long-term (i.e., one year) methane retrievals, we oversampled the TROPOMI data at 5×5 km² resolution following Sun et al. (2018) (Sun et al., 2018) where the full spatial footprint of the observation was taken into account by attributing the observed value to grid cells weighted by the spatial overlap of the observation with those grid cells.
- 147 In the second tier of our framework, we applied the matched-filter algorithm to calculate per-pixel methane enhancements with
- respect to background levels based on the SWIR sample spectrum (i.e., the 2100 2450 nm window) onboard the PRISMA (Foote et al., 2020; Guanter et al., 2021; Itziar et al., 2021). In theory, the retrieval method can depend on physically-based or data-driven algorithms. The former aims to explicitly resolve the radiative transfer between the surface, the atmosphere, and
- 151 the hyperspectral spectrometers. A key representative is the family of differential optical absorption spectroscopy (DOAS)
- 153 image using statistical methods. It is commonly based on the matched-filter and the singular vector decomposition concepts.

methods (Cusworth et al., 2019, 2020, 2021b, 2021a). The latter seeks a methane absorption spectrum across a hyperspectral

- 154 These methods are both widely applied and evaluated, especially onboard satellite (e.g., PRISMA, GF-5, and ZY-1) and
- airborne (e.g., AVIRIS and AVIRIS-NG) platforms (Cusworth et al., 2020; Foote et al., 2020; Guanter et al., 2021; Itziar et
- 156 al., 2021; Thompson et al., 2016; Thorpe et al., 2016).
- In this study, the data-driven retrieval based on the matched-filter concept was used. The main reason was that it could implicitly account for potential radiometric and spectral errors in satellite-based imaging spectroscopy. For instance, vertical striping was prevalent in hyperspectral measurements due to detector inhomogeneity, thus substantially degrading methane retrievals. The matched-filter algorithm focused on the per-pixel columns and thus tackled this issue in principle. Besides, the physically-based method had to consider background concentrations that were difficult to determine around the super-emitters. In contrast, the data-driven method was independent of background levels and can directly seek methane enhancements. Finally, the data-driven method generally had a substantially superior computational efficiency compared to the physically-based
- 164 method.

152

- 165 The matched-filter retrieval used here was similar to the one used by Thompson et al. (2016) (Thompson et al., 2016) for the 166 Hyperion imaging spectrometer onboard the EO-1 satellite. The calculation processes of methane enhancements (Δ **XCH**₄,
- 167 **ppb**) were as follows.

168
$$\Delta XCH_4(\vec{x}) = \frac{(\vec{x}-\vec{\mu})^T \Sigma^{-1} \vec{t}}{\vec{t}^T \Sigma^{-1} \vec{t}} \text{ (Eq. 1)}.$$

169 The \vec{x} denoted the spectrum under analysis. The $\vec{\mu}$ and Σ represented the mean background radiance and corresponding 170 covariance, respectively. They were calculated based on per-column spectrums in order to consider different responses of 171 across-track detectors to radiance. The \vec{t} was the target spectrum that reflected the background radiance enhanced by the 172 methane plume. It was generated by the elementwise multiplication of $\vec{\mu}$ and \vec{k} . This implicit parameter \vec{k} represented a unit





- methane absorption spectrum derived from a look-up table simulated by the MODTRAN radiative transfer model. Similarly,
 the spectral convolution was also performed on a per-column basis.
- 175 In theory, methane enhancements detected in spectrometers generally exhibit sparsity, especially over low albedo surfaces.
- 176 We thus accounted for such non-specificity effects to improve the basic version of the matched-filter model. A major measure
- 177 to compensate for the albedo effect was to scale the target spectrum \vec{t} by the pixel-specific albedo factor due to the fact that
- 178 the Beer–Lambert absorption law depended on the initial radiance in the absence of the absorber. Here the pixel-specific scalar
- 179 f was calculated based on the spectral average $\vec{\mu}$ and the analysis spectrum \vec{x} as follows:

180
$$f = \frac{\vec{x}^T \mu}{\mu^T \mu}$$
 (Eq. 2)

181 This solution made ΔXCH_4 normalized by the albedo term, which was similar to the per-pixel normalization in previous 182 hyperspectral analysis (Kraut et al., 2005).

183 The premise to launch the matched-filter algorithm was the accurate knowledge of the response of the instrument spectra to

184 the methane absorption nature. To this end, the objective was to gain the best fit between the simulated and reference spectra.

185 An initial step was thus conducted to update the spectral calibration for the channels within the 2100 - 2400 nm window, in

186 which the channel wavelength centre and width were updated for each across-track position in each scene. Other details are

187 illustrated in previous attempts (Foote et al., 2020; Guanter et al., 2021; Itziar et al., 2022).

188 2.3 Multi-tiered attribution of methane hotspots and plumes

In the first tier of our framework, we filtered the TROPOMI-based methane retrievals to identify region-scale hotspots. In each 189 190 scene, we focused on the grids with anomalous values that were noticeably higher than the average. In this study, we used a 191 Boolean mask to define the pixels that were affected by the methane emissions (Pandev et al., 2019). Although automatic 192 threshold and algorithm might result in more consistent and flexible identifications, no satisfactory set of criteria was found 193 that could be applied for this study. This was mainly because, in localized regions, the methane budgets responded to the 194 changes in not only the super-emitters but also the background. Assisted by artificial intelligence techniques in the future 195 (Ouerghi et al., 2021; Paoletti et al., 2018; Yang et al., 2018; Yu et al., 2017; Zhang et al., 2018), our framework can derive a 196 global, operational, and open-access methane monitoring network. As expected, multiple hotspots of interest result, and here

- 197 we focused on those in the United States, China, Iraq, Kuwait, and Algeria.
- In the second tier of our framework, we applied visual inspection to detect plumes using the PRISMA-based methane retrievals (Itziar et al., 2021; Martin et al., 2018; T. et al., 2022; Varon et al., 2020). To date, it was still challenging to distinguish methane plumes in hyperspectral images using full physically-based algorithms. The main cause was potential methane retrieval artifacts from hyperspectral satellites that were spatially correlated to surface features. Specifically, we manually searched for methane enhancement pixels with gas-plume-like shapes, i.e., high methane enhancements progressively decreased downwind. The resulting pixels were subsequently compared to the spectral radiance data at the 2300 nm absorption
- 204 feature sensitive to low surface albedos. In this way, the fake positives due to specific surface features were prevented. On this





basis, the candidate pixels were overlaid over simultaneous (i.e., hourly) wind fields and high-resolution imageries in individual scenes. They would be considered to be true plumes if they roughly aligned with simultaneous wind direction and originated from explicit infrastructures. Here the high-resolution satellite imageries were taken from the Google Map. The hourly wind field data came from the ERA5 reanalysis dataset produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) (Hersbach et al., 2020; Hoffmann et al., 2019). Finally, we manually drew polygons to mask such resulting plumes out. As preparation for plume emission quantification, we removed the background using the threshold of the median values of the scenes.

212 These satellite imageries allowed us to categorize methane plumes within narrow spatial scales between 50 to 500 m², such as 213 O&G extraction platforms, storage tanks, and compressor stations. They even enabled the attribution of plumes to specific 214 emission ports in individual sources due to their very high resolution. Furthermore, we could name them based on points of 215 interest in the Google Map. On this basis, such sources could be visually retrospected via long-term, high-resolution (i.e., 10 216 m) satellite images from the Sentinel-2 mission (Ehret et al., 2021; Varon et al., 2021). Their key details, like ages and statuses 217 (e.g., active or inactive), were thus collected reliably. Note that, regarding such information, national reports were typically 218 credible but inaccessible, particularly in global missions. In addition, it should be highlighted that, on top of considerably high 219 budgets, like megacities, there must be super-emitters undetectable in our way. Other causes are discussed in uncertainty 220 analysis in Supplement Information.

221 2.4 Multi-tiered quantification of methane emissions

In our framework, we calculated the total excess mass of methane in kilograms in the detected hotspots (in the first tier) and plumes (in the second tier) using the so-called integrated mass enhancement (IME) model(Frankenberg et al., 2016; Varon et al., 2018). To make conservative estimates, we defined the background levels as the 10% of the average methane concentrations in the TROPOMI-based and PRISMA-based scenes (Figs. 1b \sim 1g)(Frankenberg et al., 2016; Varon et al., 2018). On this basis, we eliminated the interferences from the background concentrations and calculated IMEs as the methane masses of the masked hotspots and plumes.

Overall, this method linked the emission rate (Q) with the measured IME via the residence time of methane (IME/Q). This residence time relied on an effective wind speed (U_{eff}) and a characteristic plume size (L) as follows:

230
$$\boldsymbol{Q} = \frac{\boldsymbol{U}_{\text{eff}} \cdot \text{IME}}{L}.$$
 (Eq. 3)

- 231 Specifically, the *IME* and *L* can be inferred from the observations of the hotspots or plumes. During this process, we carefully
- applied a Boolean plume mask that separated the pixels (*i*) with notable signals ($\Delta \Omega_i$) from background pixels and thus defined
- 233 the total areas $(\Sigma_{i=1}^{N}A_{i})$ of the hotspots or plumes. The *L* was defined as the square root of the total plume areas. Hence, the
- 234 *IME* was calculated as follows:
- 235 IME = $\sum_{i=1}^{N} \Delta \Omega_i A_i$. (Eq. 4)





- In the first tier of our framework, the effective wind speed (U_{eff}) was defined as the 10-m wind speed U_{10} obtained from the ERA5 reanalysis dataset. According to the detected hotspot, the value at the nearest hour and location were used.
- 238 In the second tier of our framework, we applied an ensemble of large eddy simulations (LES) to establish an empirical, linear
- 239 relationship between U_{eff} and the measured 10-m wind speed U_{10} as follows (Fig. S9)

240 $U_{eff} = 0.8602In(U_{10}) + 1.1513.$ (Eq. 5)

- The configurations of these simulations, such as spatial resolution and precision, were comparable to our PRISMA data. Other details in this methodology were described in Varon et al. (2018) (Varon et al., 2018).
- We estimated the uncertainties of Q by propagating the random errors in U_{10} and IME. This processes were conducted in previous studies(Cusworth et al., 2019, 2021b; Itziar et al., 2021). As shown in previous findings, the major error source came from the U_{10} term. Its random distributions typically corresponded to the 50% random error. On this basis, this error was integrated quadratically with the standard error of the IME, the result of which can be treated as the final random error of Q.
- 247 The intrinsic errors of the IME model were quantified in the following uncertain analysis.

248 2.5 Uncertainty Analysis

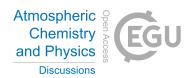
249 The objective of this work was to promote a multi-tiered satellite constellation that can monitoring global methane pledges. 250 To better understand the performance of our framework, we conducted comprehensive uncertain analysis. Note that the protocol of the uncertain analysis on our framework originated from previous studies (Itziar et al., 2021; Varon et al., 2020). 251 252 Specifically, we required to account for the uncertainties in the TROPOMI-based and PRISMA-based methane retrievals and 253 subsequent emission estimates. Therein the operational TROPOMI-based methane retrieval products had been evaluated strictly and proved to be reliable globally (except in low- and high-albedo and snow-covered areas) (Lorente et al., 2021; Sha 254 255 et al., 2021). In this work, we thus focused on three main sources of uncertainties, specifically including (1) uncertainties in 256 the PRISMA-based methane retrievals; (2) uncertainties in the TROPOMI-based methane emission estimates; and (3) 257 uncertainties in PRISMA-based methane emission estimates. During the analysis for the latter two uncertain sources, we would 258 further investigate the potential wind impacts on the methane emission estimates. Note that it remained challenging to directly 259 quantify the uncertainties in the wind fields across our cases due to the lack of measurements. We would thus assess the 260 variations in the methane emission estimates driven by distinct wind data. From such analysis, we could confirm the reliable performance of our framework. Details can be found in Supplementary Information. 261

262 3 Results and discussions

263 **3.1 Multi-tiered imaging of global methane hotspots and super-emitters**

Figure 1 presents representative sets of methane hotspots and associated super-emitters across the United States, China, the Middle East (Iraq and Kuwait), and North Africa (Algeria) via our multi-tiered satellite constellation. Each group first clarified



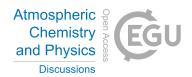


266 a methane-abundant region and further focused in on explicit super-emitters. Among them, five methane-abundant regions 267 were captured in Wattenberg (the United States), Yangquan (China), Rumaila (Iraq), Burgan (Kuwait), and Hassi Messaoud (Algeria) (Fig. 1a and Table S1). These accounted for 4805 ~ 46138 kg/h methane emissions based on our daily first-tiered 268 269 (i.e., TROPOMI-based) monitoring. From the perspective of a state-of-the-art global methane emission inventory (i.e., EDGARv6.0), such high values ranked among the top 1% regarding emission intensities per unit area (km²) (Fig. S1)(Crippa 270 et al., 2020). The Rumaila field, for example, was known as the largest oil field in Iraq (in terms of both reserves and yields). 271 272 In this work, it was found with a significant methane emission intensity exceeding 45000 kg/h (Fig. 1b). In addition to such 273 well-known oil fields (Figs. 1c ~ 1f), methane hotspots emerged in developing coal mines, like Yangquan, with comparable 274 emission levels (> 30000 kg/h) (Fig. 1g).

275 We attributed these methane enhancements to specific methane plumes via the second-tiered (i.e., PRISMA-based) monitoring 276 (Figs. $1b1 \sim 1g2$). There are substantial variations in the methane plumes' amounts, types, and magnitude, even in a single 277 methane-abundant region. For instance, in the Burgan field, the second-tiered monitoring detected up to eight methane plumes 278 in a handful of grids in the first-tiered monitoring (Figs. 1c1 ~ 1c4 and 1d1 ~ 1d4). Such intensive distributions were also 279 found in previous region-oriented surveys in the Permian basin and California (Duren et al., 2019; Itziar et al., 2021). Together 280 with high-definition images (Fig. S2), we found that such plumes originated from various sources, such as flares, factories, 281 and wells. A breakthrough was the capture of two distinctive plumes in an individual methane source with extremely high 282 emissions (> 10000 kg/h), unprecedented in previous satellite-based exploration and only observable in aircraft surveys (Fig. 283 1b1). Such precise distinction benefited from the high resolution of the second-tiered monitoring, despite being limited by the 284 relatively higher detection threshold (~ 300 kg/h) (Guanter et al., 2021). Besides, factories and wells can also emit such evident 285 plumes (Fig. 1c1 and Figs. 1e1 and 1e2). By comparison, other plumes were typically more diffuse but with comparable 286 emission magnitude (~ 1000 ~ 7000 kg/h).

Note that the above results represent only snapshots at the overpass moments of the satellites (i.e., TROPOMI and PRISMA) 287 288 (Figure 1). Specifically, for a given set (including both a methane-abundant region and associated super-emitters), the overpass 289 timing of TROPOMI can be nearly concordant with that of PRISMA. The temporal gaps could be frequently controlled within 290 ten days (e.g., Figs. 1b and 1d), even two days (Figs. 1e, 1f, and 1g). For instance, within only two days (August 18th and 19th, 291 2021, November 15th and 17th, 2021), our multi-tiered satellite constellation went through the Hassi Messaoud field and the 292 Yangquan coal mine and provided in-depth views of methane budgets, including methane-abundant regions and their drivers (Figs. 1e and 1g). Even, in just one day (July 7th, 2021), our multi-tiered satellite constellation not only uncovered methane 293 294 enhancements in the Wattenberg field (Fig. 1f) but also tracked them back to explicit methane super-emitters (Figs. 1f1 and 295 1f2). As expected, if we extended the monitoring window of our framework to years, more methane super-emitters were 296 subsequently captured (Figs. S3 and S4). Moreover, our framework via multi-tiered satellite constellation paves an in-time 297 way for routine monitoring of global methane hotspots and associated super-emitters.





298 3.2 Multi-tiered verification of global methane super-emitters

299 Four unexpected cases occurred in Burgan (Iraq), Hassi Messaoud (Algeria), and Yangquan (China), potentially explainable if we took mutual verification of the first- and second-tiered monitoring into consideration. First, an anomalous methane plume 300 301 was detected in the Burgan field (Fig. 1c4) of high emission magnitude (> 1500 kg/h), notably exceeding typical O&G facilities, 302 from an elusive source (i.e., no clear source could be attributed) (Fig. S2). The long-term measurements of our multi-tiered 303 satellite constellation intermittently, rather than accidentally, observed this abnormal plume (Figs. S4). Furthermore, uncertain analysis (see Materials and Methods) helped confirm this real plume. In particular, the methane plumes were clearly 304 uncorrelated with the surface brightness from space (Fig. S5). Consequently, the most likely hypothesis for this super-emitter 305 306 was methane leakage from gigantic O&G pipelines as shown in the Google Map (Fig. S2).

307 Second, we observed suspect trails of methane plumes above the storage tanks in the Burgan field (Fig. 1d4). Conceivably, the 308 technical noise driven by albedo effects bore the brunt, although it was believed to be corrected reliably (See Materials and 309 Methods). To this end, we applied a multi-spectral retrieval algorithm to eliminate this effect to a large extent. The detailed 310 illustrations are shown in Materials and Methods (Fig. S6). Consequently, we provided evidence that un-negligible methane emissions (> 3500 kg/h) may very well be the unique explanation, likely related to fugitive methane leaks from the storage 311 tanks. This was only seen in previous aircraft-based surveys(Frankenberg et al., 2016). Therefore, our multi-tiered outcomes 312 313 indicate even more widespread methane leaks than expected. Note that the multi-spectral retrieval algorithm cannot completely remove the albedo effects on our framework. As such, our framework could lead to efficient on-site re-inspection on worldwide 314 315 and innumerable O&G fields.

316 Third, our framework detected a new methane super-emitter in the Hassi Messaoud field on December 7, 2021 (Fig. 1e4). By

revisiting historical satellite images in the second-tiered monitoring (Fig. S7), we could confirm that this super-emitter arose between October 18th and November 12, 2021. These results indicate that monitoring of global methane super-emitters can attain monthly resolution via current satellite constellation alone. Conceivably, more satellite observations would further close the time window. Fourth, a distinct methane plume appeared in a coal mine in a mountainous area (Yangquan, China), exceeding all of the detected O&G super-emitters regarding the emission rate (> 7000 kg/h) (Fig. 1g1).

322 Figure 2a illustrates that, in our multi-tiered satellite constellation, the extent to which the explicit plumes in the second tier 323 explained the regional budget detected by the first tier. Overall, the plumes in the former were mostly responsible for large 324 shares (> 8.2%) of regional budgets in the latter. In the Rumaila, Burgan, and Wattenberg fields, the detected methane plumes 325 played a more critical role, with contributions up to $53.8 \sim 65.9\%$. Note that such contribution estimates might occasionally exceed 100% mainly owing to the inconsistent overpass moments between the first- and second-tier monitoring. By 326 comparison, the relatively low but still significant contributions in the Hassi Messaoud field (8.2%) and Yangquan coal mine 327 328 (35.7%) were partly due to the technical limitation of our framework in detecting methane plumes on top of high background 329 levels. Collectively, the heavy-tail law of methane plume distributions, early reported for regional O&G fields (like the Permian

basin and California) (Duren et al., 2019; Itziar et al., 2021), possibly applied worldwide.





To further explore such a hypothesis, we extended the temporal sample window of our multi-tiered framework. As expected, the first tier pictured less vivid methane enhancements mainly due to wind averages (Martin et al., 2018; McLinden et al., 2016; Sun et al., 2018) (Fig. S3), while the second tier could capture more methane plumes (Fig. S4). This would lead to more volatile contributions of the plumes to regional budgets, which, however, remained at high levels. This reinforces the above hypothesis for the widespread occurrence of methane super-emitters.

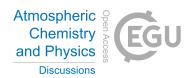
A regional survey in a California field was considered as the best reference, owing to its utilization of systematic airborne 336 337 measurements to detect and quantify methane super-emitters (Duren et al., 2019). They survey reported 1181 methane plumes, 338 more than 500 times larger than previous aerial studies (Englander et al., 2018), with a median emission intensity of 170 kg/h. 339 These results were thus used to directly evaluate the outcomes in the second tier (Fig. 3). Even though some regions of interest 340 in this study were far less famous than the California field, their emission intensities were much higher. Specifically, these 341 plumes detected by the second-tiered monitoring had emission intensities (1142 ~ 11698 kg/h) that exceeded the median value in the California field. Satellite-based surveys conducted repeatedly for the Permian basin (one of the top O&G bases 342 343 worldwide) from 2019 to 2020 (Fig. 3) as compared to the surveys in the California field, they achieved a much higher number 344 of strong methane super-emitters, the median emission rates (1850 kg/h) much closer to ours (2888 kg/h). Collectively, this 345 direct comparison indicates the outstanding strength of our results that, though derived from a small dataset, could be analogous 346 to abundant outcomes from field campaigns. More importantly, this highlights the urgent need for global monitoring of 347 'nameless' O&G facilities that possibly emit methane as much as the California field and Permian basin.

348 **3.3 Multi-tiered challenges of national emission inventories**

These multi-tiered results challenge traditional methane emission inventories (Fig. 4). Here the conventional emission data was obtained from a state-of-the-art bottom-up emission inventory (i.e., EDGARv6.0) for the year 2018. Consequently, for the methane hotspots, this inventory was mostly consistent with the present results (-49.9 ~ 91.8%), with a fine average bias (63.2%). The Hassi Messaoud field in Algeria was a unique exception, where the O&G sector was in rapid development, with a relatively larger bias (489.2%). By comparison, this inventory significantly undervalued the methane super-emitters (up to orders of magnitude). This indicates that traditional emission inventories might have acceptable performance for traditional methane-abundant regions while incapable of tracking methane super-emitters.

First, outdated spatial proxies might explain the large divergence between our plant-based estimates and the EDGARv6.0 (Fig. 1b1 and Fig. S8). Moreover, the EDGARv6.0 was designed for the year 2018, missing the newly established O&G plants with high methane emissions. Second, in principle, conventional inventories directly missed high emissions caused by abnormal operations (e.g., equipment failures) (Fig. 1c4 and Fig. S8) such as the O&G blowout shown in on-site surveys (Pandey et al., 2019). A compromise was downwind measurements, yet insufficiently reliable as shown in previous findings(A. et al., 2018). In addition, the relatively low bias in the Rumaila and Hassi Messaoud fields might be explained by other causes (Figs. 1b2 and 1e3) such as outdated emission factors. Empirically, a plant-level inventory, once optimized by direct measurements, can





raise total methane emissions by ~ 60%, although source categories vary substantially (A. et al., 2018). Besides, temporal variability might also explain top-down and bottom-up differences in methane emission estimates. For instance, the peak emission rate could exceed 40% higher than the average, which might occur in the middle afternoon due to specific processes, like episodic venting from manual liquid unloading (Vaughn et al., 2018). This aligned with the sampling time of the satellites, thus biasing bottom-up inventories. Collectively, it is necessary to carefully consider all factors affecting methane emissions, including emission factor updating and spatiotemporal variations, in order to develop effective strategies for mitigating methane emissions.

370 **3.4 Implications for global methane monitoring**

We present a multi-tiered, space-based framework that can harmonize planet-scale and plant-level methane retrievals (Fig. 5). Using this framework, we patrol the world, with synergistic, proactive detections on the methane-abundant regions and methane super-emitters across the United States, China, the Middle East (Iraq and Kuwait), and North Africa (Algeria). We even lock new methane super-emitters, track potential methane leakages from storage tanks, and distinguish multiple methane plumes in a single source. Such achievements are mostly unprecedented in satellite surveys and only observed in aircraft campaigns. On this basis, our results challenge national reports that possibly miss unexpected super-emitters or mislead emission magnitude, partly due to surges of oil and gas (O&G) facilities and widespread abnormal operations.

Our data prove that depending on ready-made satellite missions alone can initiate immediate, proactive monitoring of global methane pledges, in contrast to existing surveys that have to focus on a priori methane-abundant regions. As such, as the window for achieving the Paris target is rapidly closing, we will not need to sit back and wait for upcoming space missions, like MethaneSAT and SBG in the United States, EnMAP in Germany, a new version of GF-5 in China, and, later, the European Space Agency's CHIME from 2025 to 2030 (Cusworth et al., 2019). In addition, while scientific communities persistently debates the drivers of the recent methane surge (G. et al., 2014; Nisbet et al., 2019; Turner et al., 2019), the consequences of our outcomes are clear, not only holding clues but also facilitating mitigation.

It should be noted that the multi-tiered framework is sustainable (Fig. 5). On the one hand, it can harmonize multiple satellites. 385 386 The potential representatives include upcoming official missions (e.g., the GF-5) (Itziar et al., 2021), current private 387 constellations (e.g., the GHGSat series) (Jervis et al., 2021; Varon et al., 2020), and explorable multispectral products (e.g., 388 the Worldview-3 and Sentinel-2) (Sánchez-García et al., 2021). On the other hand, the framework is not confined to satellites 389 and can be expanded by integrating in situ (e.g., Global Atmosphere Watch Programme) (World Meteorological Organization, 2022), aircraft, and unmanned aerial vehicles (UAVs) (Cusworth et al., 2020; Gålfalk et al., 2021; Tuzson et al., 2020). 390 391 Particularly, on the basis of our framework, rapid advances in artificial intelligence (AI) techniques are projected to completely 392 replace manpower to seek faint signals of methane enhancements in Earth's surface, and to significantly optimize data-driven

393 algorithms of methane emission estimates (Reichstein et al., 2019; Yuan et al., 2020). In principle, subsequent mitigation of





such super-emitters via routine maintenances, leak detections, or emergent repairs can provide effective, efficient, and
 economical solutions toward the Paris target (Mayfield et al., 2017).

396 These outcomes have important ramifications for low- and middle-income countries. World powers, like the United States and

397 European Union, lead new national methane pledges. They are separately on the way to creating vast operational infrastructures

398 to monitor ambitious climate goals. Still, huge holes remain in coverage and authority, at least by the middle of this decade.

399 This situation is especially worse for low- and middle-income countries, where the tight budget dims the hopes for filling up

400 those holes by 2030, while methane emissions are likely to rise as countries develop. In this context, the present framework

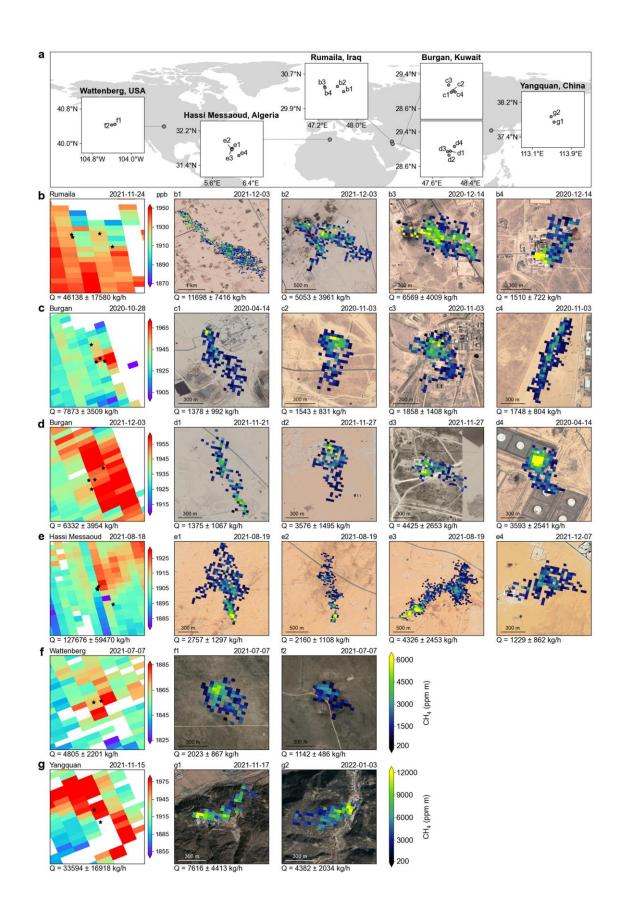
401 can at once serve as the cost-effective piece of the global methane monitoring network and thus support fair climate

402 negotiations between countries.

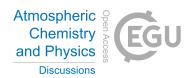
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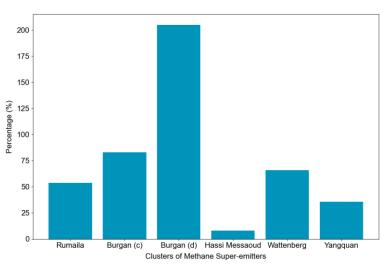




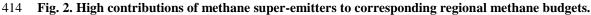
405 Fig. 1. Methane hotspots and associated super-emitters across the United States, China, Iraq, Kuwait, and Algeria via the multi-tiered daily satellite constellation. (a) Methane-abundant regions and associated super-emitters are captured by 406 407 the TROPOMI and PRISMA, respectively. Their locations are marked by black rectangles and dots. Their names are obtained 408 from the Google Map, usually being the names of the nearest O&G fields and coal mines. ($\mathbf{b} \sim \mathbf{g}$) Each group clarifies a methane-abundant region and explicit super-emitters (b1 ~ b4, c1 ~ c4, d1 ~ d4, e1 ~ e4, f1 ~ f2, and g1 ~ g2). For each super-409 emitter (five-pointed stars), the overpass moments of the multi-tiered satellite constellation and the consequent emission 410 411 estimate are presented. Its base map is obtained from the ©Google Map. The second color bar for the PRISMA is suitable for the super-emitters in China, while the first is for other countries. 412

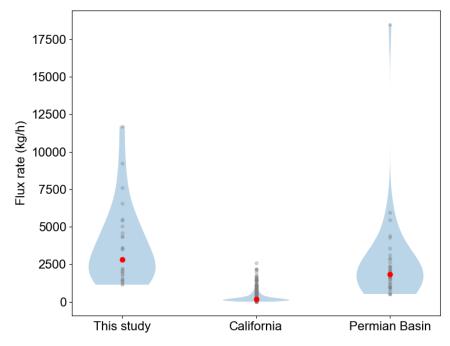






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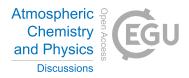




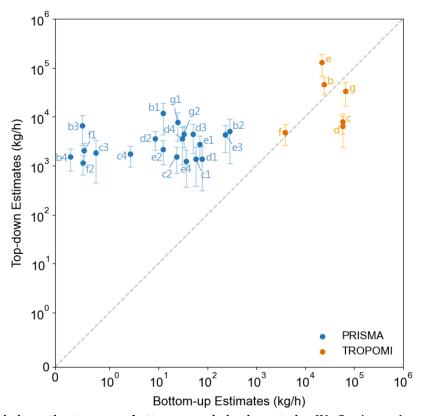
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Fig. 3. Comparison of emission estimates of methane plumes between surveys. The surveys for the California field and Permian basin are selected as the references. They report 1181 and 39 methane plumes, while our second-tiered survey attempts Plumes. Violin plots show statistical distributions of methane plume emission rates for these surveys. For each survey, the

419 grey dots refer to the emission rates of the individual plumes and the red dot represents the median value.





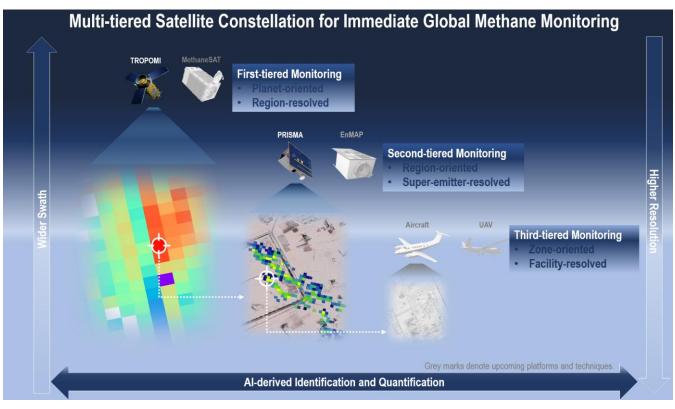


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Fig. 4. Multi-tiered emission estimates versus bottom-up emission inventories. We first interpolate the bottom-up emission inventories into the resolution consistent with our multi-tiered results. On this basis, the bottom-up emission rates in the grids that the detected hotspots and plumes cover are summed up to compare with the results. The detected hotspots (yellow dots) and plumes (blue dots) correspond to those as shown in Fig. 1. The grey dashed line represents the ratio of the bottom-up emissions to the top-down ones of 1:1.







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Fig. 5. Multi-tiered satellite framework for immediate global methane monitoring. This framework harmonizes global-427 scale and high-resolution methane retrievals, with a dual focus on mapping region-scale and plant-level drivers. In this work, 428 429 the framework reconciles the spacious swath of TROPOMI (i.e., \sim 2600 km) with the high resolution of PRISMA (i.e., 30 \times 430 30 m²), in contrast to conventional satellite-based surveys that were of either insufficient samplings or narrow views. Looking 431 forward, developments of Earth's monitoring platforms (e.g., satellites, aircrafts, and unmanned drones) and artificial intelligence will continue to strengthen the performance of methane plume retrievals and emission estimates. On eve of the 432 Paris target, at least while a super methane satellite with spacious swath, high resolution, and agile analysis is not in place, our 433 434 multi-tiered satellite constellation has important implications for measuring global methane pledges. The appearances of the 435 TROPOMI, MethaneSAT, PRISMA, and EnMAP are obtained from http://www.tropomi.eu/, https://www.methanesat.org, https://www.asi.it/en/earth-science/prisma/, and https://www.enmap.org/, respectively. The methane maps from the 436 TROPOMI and PRISMA refer to the results in Figs. 1e and 1b1. The grey marks indicate upcoming platforms (i.e., 437 MethaneSAT and EnMAP) and techniques (e.g., AI techniques that can optimize the identification and quantification of 438 439 methane super-emitters).

440





441 Data availability.

The operational TROPOMI product is available at <u>https://scihub.copernicus.eu/</u>, <u>https://www.temis.nl/emissions/data.php</u>. The PRISMA data are publicly available to registered users at https://prisma.asi.it/. The WRF-CHEM model code is available at https://ruc.noaa.gov/wrf/wrf-chem/. All Sentinel-2 satellite data are publicly available through the Copernicus Open Access Hub (https://scihub.copernicus.eu/). The HITRAN line spectra is publicly available through the HITRANonline database (https://hitran.org/). The ERA5 data come from https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5. The

447 EDGARv6.0 dataset comes from https://edgar.jrc.ec.europa.eu/gallery?release=v60ghg&substance=CH4§or=TOTALS.

448 Code availability.

449 The codes are available upon request to corresponding author.

450 Supplementary information.

451 Supplementary information accompanies this paper.

452 Author contributions.

- 453 P. L. designed this study and wrote the manuscript. P. L. and Y. W. developed the retrieval algorithm. P. L., Y. W., X. G., Y.
- 454 H., and Y. P. performed the data analysis. S. Y., A.B., D. R., and J. H. S. contributed to the manuscript.

455 Competing interests.

456 The authors declare no competing interests.

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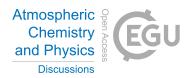
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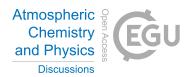
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