Toward a versatile spaceborne architecture for immediate monitoring of the global methane pledge

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Abstract.

The global methane pledge paves a fresh, critical way toward Carbon Neutrality. However, it remains largely invisible and highly controversial due to the fact that planet-scale and plant-level methane retrievals have rarely been coordinated. This has never been more essential within a narrow window to reach the Paris target. Here we present a two-tiered spaceborne architecture to address this issue. Using this framework, we patrol the world, like the United States, China, the Middle East, and North Africa, and simultaneously uncover methane-abundant regions and plumes. These include new super-emitters, potential 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-emitters. Our results show that, in principle, the above framework can be extended to be multi-tiered by adding upcoming stereoscopic measurements and suitable artificial intelligence, thus versatile for immediate and future monitoring of the global methane pledge.

1. Introduction

Global methane pledges finalized at the COP26 (the 26th United Nations Climate Change Conference of the Parties) have been never more ambitious (Schellnhuber et al., 2016; Schurer et al., 2018; United Nations, 2021). More than 100 countries have promised 30% methane emission reductions by 2030. Also, energy giants (e.g., Shell and BP) have committed to clear targets of methane mitigation. Such pledges have never been more essential within a narrow window (< ten years) to reach the 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 al., 2016; Saunois et al., 2016, 2020). Worse still, it is thought to rise since 2007 (Mikaloff and Hinrich, 2019), surge since 2014 (Nisbet et al., 2019), and set another record in 2021 (National Oceanic and Atmospheric Administration, 2022). Fortunately, methane is short-lived (~ ten years) (Shoemaker et al., 2013), and, particularly, that from human activities can be reduced in half using existing technologies by 2030 (Ocko 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 (Allen et al., 2015; Alvarez, Ramón A. and Zavala-Araiza, Daniel and Lyon, David R. and Allen, David T. and Barkley, Zachary R. and Brandt, Adam R. and Davis, Kenneth J. and Herndon, Scott C. and Jacob, Daniel J. and Karion, Anna and Kort, Eric A. and Lamb, Brian K. and Lauva, 2018; 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 (Alvarez, Ramón A. and Zavala-Araiza, Daniel and Lyon, David R. and Allen, David T. and Barkley, Zachary R. and Brandt, Adam R. and Davis, Kenneth J. and Herndon, Scott C. and Jacob, Daniel J. and Karion, Anna and Kort, Eric A. and Lamb, Brian K. and Lauva, 2018).

To this end, widespread super-emitters present a unique opportunity worldwide (Duren et al., 2019; Lauvaux et al., 2022; Pandey et al., 2019; Zavala-Araiza et al., 2015, 2017). Super-emitters can generally be defined to be emission sources that comprise highly concentrated methane plumes and dominate localized methane budgets (~ 5×5 km^2). In contrast to region-scale hotspots (or area sources), they can be attributed to individual facilities (e.g., factories, chimneys, and pipelines), typically with side lengths varying from several meters to tens of meters depending on monitoring instruments. Super-emitters are typically responsible for the underestimates of methane emissions (Alvarez et al., 2018; Duren et al., 2019; Itziar et al., 2021; Lauvaux et al., 2022; Thompson et al., 2016). Moreover, there is increasing evidence that methane emissions follow a heavy-tailed distribution (Duren et al., 2019; Frankenberg et al., 2016; Lauvaux et al., 2022), for which relatively small number of sources (so-called super-emitters) can account for a disproportionately large share of total emissions. In contrast to area sources (e.g., cities), super-emitters are typically coal mines, wells, gathering stations, storage tanks, pipelines, and flares, with even less than dozens of metres in diameter but high-concentrated methane plumes (Allen et al., 2013; Miller et al., 2019;

75 Subramanian et al., 2015; Varon et al., 2019). We thus anticipate that significant emission mitigation could be achieved by

76 deploying well-designed systems to identify methane super-emitters. For instance, in support of the Paris agreement, the 17th

77 World Meteorological Congress (2015) requested an Integrated Global Greenhouse Gas Information System (IG3IS) that

78 aimed to develop a measurement framework for methane emission reductions (Phil DeCola and WMO Secretariat, 2017).

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

80 methane super-emitters. Despite this, they are spatially confined (e.g., regionally) and temporally infrequent (e.g., a few weeks),

81 incapable of exploring global methane super-emitters (Alvarez, Ramón A. and Zavala-Araiza, Daniel and Lyon, David R. and

82 Allen, David T. and Barkley, Zachary R. and Brandt, Adam R. and Davis, Kenneth J. and Herndon, Scott C. and Jacob, Daniel

83 J. and Karion, Anna and Kort, Eric A. and Lamb, Brian K. and Lauva, 2018; Conley et al., 2016; Duren et al., 2019; Marchese

84 et al., 2015; Nisbet et al., 2020; Smith et al., 2017; Thompson et al., 2016; Thorpe et al., 2016). Today, substantial advances

85 have been made towards detecting and quantifying methane super-emitters from space (Cusworth et al., 2019; Hu et al., 2018;

86 Irakulis-Loitxate et al., 2022; Jacob et al., 2016; Pandey et al., 2019; Thompson et al., 2016) (Table 1). Such advances, however,

87 have rarely been expanded to measure the global methane pledge because large-scale swath and high-resolution sampling have

88 not been coordinated. First, global methane monitoring has become possible. A flagship satellite mission is the TROPOspheric

89 Monitoring Instrument (TROPOMI) onboard the Copernicus Sentinel-5 Precursor satellite (Lauvaux et al., 2022; Veefkind et

90 al., 2012). It can offer daily global insights for methane column concentrations, with a large swath width of ~ 2600 km, a

91 moderate resolution of 7.0×5.5 km^2 (since August 2019), and high signal-to-noise ratios. However, its relatively coarse spatial

92 sampling still limits its application to detect methane super-emitters (Lauvaux et al., 2022). Second, next-generation satellite

93 missions, pioneered by the GHGSat constellation (three satellites at the moment), emerge for mapping methane super-emitters

94 (Cusworth et al., 2019), with a narrow swath (e.g., ~ 12 km) but a ground-breaking high-resolution spatial sampling (e.g., 25

95 ~ 50 m) (Jervis et al., 2021; Varon et al., 2020). Complementary to the GHGSat constellation, satellite-based hyperspectral

96 imager spectrometers, such as PRISMA, GF-5, ZY1, Sentinel-2, and Worldview-3, have shown great potentials (Guanter et

97 al., 2021; Itziar et al., 2021; Sánchez-García et al., 2021; Varon et al., 2021). They can resolve methane enhancements and

98 attribute them to specific infrastructures via similar narrow swath and high-resolution sampling (e.g., 30 m). Note that regions

99 those satellites usually gazed at are originally well-known home to methane super-emitters. Narrow swath coverage thus

100 remains a crucial limitation for global surveys of methane super-emitters. Collectively, existing studies still struggle to

101 surveillance global methane super-emitters due to the fact that individual satellite missions, either TROPOIM or PRISMA,

102 cannot coordinate large-scale swath and high-resolution sampling.

103 To address this issue, we present a two-tiered, space-based framework that coordinates TROPOIM and PRISMA for both

104 planet-scale and plant-level methane retrievals. The key is that ready-made satellite missions alone have the potential to initiate

105 immediate monitoring of the global methane pledge. Using this framework, we patrol the world, with an experimental focus

106 on China, the United States, Iraq, Kuwait, and Algeria, and reveal both region-scale hotspots and plant-level super-emitters.

107 We can even gaze at a single source to map multiple plumes and inspect possible methane leakages. These results can challenge

108 national reports that possibly miss unexpected super-emitters or mislead emission magnitude. On the eve of the Paris target,

109   at least while a global methane monitoring network is not in place, this multi-tiered satellite constellation presented in this

110   study has important implications for measuring global methane pledges.

2. Materials and Methods

2.1 Multi-tiered satellite constellation

113   The multi-tiered satellite constellation is designed to reconcile global-scale and high-resolution methane monitoring. First,

114   TROPOMI offers a unique potential for global methane monitoring, depending on its large-scale (i.e., 2600 km) swath, daily

115   revisit time, regional footprint (i.e., $5.5 \times 7\,km^2$ since August 2019), and sounding precision and accuracy (i.e., < 1 %)

116   (Veefkind et al., 2012). Approximately, TROPOMI observes a full swath per second, resulting in ~ 216 spectra per second.

117   This instrument comprises two spectrometer modules, the first involving near-infrared (NIR) spectral channels, and the second

118   dedicated to the shortwave-infrared (SWIR) spectral channel. The NIR and SWIR channels are equipped with spectral

119   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

120   are incorporated in different instrument modules, the NIR spectra will be co-registered with the SWIR spectra before

121   performing methane retrievals. The methane total column-averaged dry-air mole fraction (XCH_4) is retrieved from near-

122   infrared (NIR) (757 ~ 774 nm) and shortwave-infrared (SWIR) (2305 ~ 2385 nm) spectral measurements for sunlight

123   backscattered by Earth's surface and atmosphere (Hu et al., 2018). In this study, only high-quality measurements, retrieved

124   under cloud-free and low aerosol load conditions, are used. These measurements are filtered, in addition, for solar zenith angle

125   (< 70°), low viewing zenith angle (< 60°), and smooth topography (the surface elevation of < 80 m within 5 km radius) as

126   described in Hu et al. (28) (Hu et al., 2018).

127   Hyperspectral satellite missions serve as the second tier, responsible for mapping localized methane super-emitters depending

128   on their unprecedented resolution (i.e., 3m ~ 50m). Therein PRISMA, as an open-access representative, is specifically suitable

129   for this work. It can image the solar radiation reflected by the Earth's surface and atmosphere via hundreds of spectral channels

130   between the visible and SWIR spectrum (~ 400 ~ 2500 nm). Measurements in the SWIR spectrum from 2000 to 2500 nm

131   sample absorption features from water vapor, carbon dioxide, and methane. Therein the 2100 nm and 2450 nm windows are

132   especially sensitive to methane. Furthermore, the signal-to-noise ratio is reported to be about 100 in the SWIR for a relatively

133   dark vegetation pixel and increases up to above 200 for bright soil surfaces in oil and gas extraction sites. More importantly,

134   it covers areas of $30 \times 30\,km^2$ with a 30 m spatial sampling.

135   We collect dozens of daily measurements from the multi-tiered satellite constellation. These measurements experimentally

136   map regional methane hotspots and localize methane super-emitters across the United States, China, the Middle East (Iraq and

137   Kuwait), and North Africa (Algeria). The acquisitions are mostly taken between April 2020 and January 2022.

2.2 Multi-tiered methane retrievals

In the first tier of our framework, we employ the operational methane products via TROPOMI onboard the Sentinel 5 satellite. The target product is the column-averaged dry-air volume mixing ratio of methane (XCH_4), which will be retrieved simultaneously with scattering properties of the atmosphere. The operational retrieval algorithm is based on RemoTeC (Butz et al., 2009; Hasekamp and Butz, 2008), which is originally developed for CO_2 and methane retrievals from GOSAT observations (Butz et al., 2011). It attempts to fit spectra observed by the TROPOMI-based NIR and SWIR channels. Its sensitivities to atmospheric scattering properties, atmospheric input data, and instrument calibration errors have been extensively evaluated (Sha et al., 2021; Verhoelst et al., 2021). As a result, the operational products are proved to be critically stable, with a convergence rate of 99% and high significance as compared with both satellite-based (e.g., GOSAT) and ground-based (e.g., TCCON) measurements. The required accuracy and precision of < 1 % for the XCH_4 product are met for clear-sky measurements over land surfaces and after appropriate filtering of difficult scenes. Moreover, the forward model error is less than 1 % for about 95 % of the valid retrievals. Model errors in the input profile of water do 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 are the most sensitive to an error in the instrument spectral response function of the shortwave infrared channel.

In the second tier of our framework, we apply 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 the hyperspectral spectrometers. A key representative is the family of differential optical absorption spectroscopy (DOAS) methods (Cusworth et al., 2019, 2020, 2021b, 2021a). The latter seeks a methane absorption spectrum across a hyperspectral image using statistical methods. It is commonly based on the matched-filter and the singular vector decomposition concepts. 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 al., 2021; Thompson et al., 2016; Thorpe et al., 2016). In this study, the data-driven retrieval based on the matched-filter concept is used. The main reason is that it could implicitly account for potential radiometric and spectral errors in satellite-based imaging spectroscopy. For instance, vertical striping is prevalent in hyperspectral measurements due to detector inhomogeneity, thus substantially degrading methane retrievals. The matched-filter algorithm focuses on the individual columns rather than the whole scene to resolve methane enhancements. This means that the methane enhancement per column is calculated separately (i.e., methane enhancements were calculated on a per-column basis). More explanations can be found in Guanter et al. (2021). Besides, the physically-based method has to consider background concentrations that are difficult to determine around the super-emitters. In contrast, the data-driven method is independent of background levels and can directly

170   seek methane enhancements. Finally, the data-driven method generally has a substantially superior computational efficiency

171   compared to the physically-based method.

172   The matched-filter retrieval used here is similar to the one used by Thompson et al. (2016) for the Hyperion imaging

173   spectrometer onboard the EO-1 satellite. The calculation processes of methane enhancements ($\mathbf{\Delta XCH_4}$, **ppb**) are as follows.

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

175   The $\vec{\boldsymbol{x}}$ denotes the spectrum under analysis. The $\vec{\mu}$ and Σ represent the mean background radiance and corresponding

176   covariance, respectively, calculated with their common formulas after subtracting the current signal estimates from the data.

177   Specifically, the $\vec{\mu}$ is calculated from the data with the removal of the most recent enhancement estimates, while the Σ is then

178   calculated with updated $\vec{\mu}$ and the most recent enhancement estimates. More technical details are reported in previous studies

179   (Foote et al., 2020). Note that, owing to the non-uniform response of individual detectors in PRISMA, they are calculated

180   based on per-column spectrums in order to consider different responses of across-track sensors to radiance. The $\vec{\boldsymbol{t}}$ is the target

181   spectrum that reflects the background radiance enhanced by the methane plume. It is generated by the elementwise

182   multiplication of $\vec{\mu}$ and $\vec{\boldsymbol{k}}$, This implicit parameter $\vec{\boldsymbol{k}}$ represents a unit methane absorption spectrum derived from a look-up

183   table simulated by the MODTRAN radiative transfer model. Similarly, the spectral convolution is also performed on a per-

184   column basis.

185   In principle, it would be more difficult to detect methane enhancements in pixels over low-albedo surfaces. Although methane

186   absorption is independent of albedo, the resulting signal in absolute radiance is weakened with surface albedo decreasing. A

187   major measure to compensate for the albedo effect is to scale the target spectrum $\vec{\boldsymbol{t}}$ by the pixel-specific albedo factor due to

188   the fact that the Beer–Lambert absorption law depends on the initial radiance in the absence of the absorber. Here the pixel-

189   specific scalar $\boldsymbol{f}$ is calculated based on the spectral average $\vec{\mu}$ and the analysis spectrum $\vec{\boldsymbol{x}}$ as follows:

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

191   This solution makes $\mathbf{\Delta XCH_4}$ normalized by the albedo term, similar to the per-pixel normalization in previous hyperspectral

192   analysis (Kraut et al., 2005). The premise to launch the matched-filter algorithm is the accurate knowledge of the response of

193   the instrument spectra to the methane absorption nature. To this end, the objective is to gain the best fit between the simulated

194   and reference spectra. An initial step is thus conducted to update the spectral calibration for the channels within the 2100 -

195   2400 nm window, in which the channel wavelength centre and width are updated for each across-track position in each scene.

196   Other details are illustrated in previous attempts (Foote et al., 2020; Guanter et al., 2021; Irakulis-Loitxate et al., 2022).

197   **2.3 Multi-tiered attribution of methane hotspots and plumes**

198   In the first tier of our framework, we apply visual inspection to identify methane hotspots using the TROPOMI-based methane

199   retrievals. The transformation from visual inspection to automatic recognition would significantly advance long-term, global

200   methane monitoring. However, no satisfactory set of criteria was found that could be suitable for this study. This was mainly

because, in localized regions, methane budgets respond to the changes in not only super-emitters but also complex external factors (e.g., meteorology, topography, and background concentrations). Similar compromises are also adopted in previous studies. Therefore, automatic recognition enabled by artificial intelligence would play an essential role in the versatile spaceborne architecture for long-term, global methane monitoring (Ouerghi et al., 2021; Paoletti et al., 2018; Yang et al., 2018; Yu et al., 2017; Zhang et al., 2018).

Regarding the identified methane hotspots, we utilize a Boolean mask to select plume-influenced pixels downwind of the source. The background distribution (mean ± standard deviation) is defined by an upwind sample of the measured columns, in which the hourly wind field data come from the ERA5 reanalysis dataset produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) (Hoffmann et al., 2019). We then sample the surrounding (5×5) pixels centred on each pixel and compare the corresponding distributions to the background distribution based on a Student's t-test. Pixels with a distribution substantially higher than the background at a confidence level of 95% are assigned to the plume. More details in the Boolean plume mask can be found in previous studies (Pandey et al., 2019; Varon et al., 2018).

Regarding the identified regional hotspots, we also apply visual inspection to search for plumes within their surrounding 30 km scales (i.e., corresponding to the swath width of PRISMA) in the second tier of our framework (Itziar et al., 2021; Lauvaux et al., 2022; Martin et al., 2018; Varon et al., 2020). To date, it is still challenging to distinguish methane plumes in hyperspectral images using full physically-based algorithms. The main cause is potential methane retrieval artifacts from hyperspectral satellites that are spatially correlated to surface features. Specifically, we manually search for methane enhancement pixels with gas-plume-like shapes, i.e., high methane enhancements progressively decrease downwind. The resulting pixels are subsequently compared to the spectral radiance data at the 2300 nm absorption feature sensitive to low surface albedos. In this way, the fake positives due to specific surface features are prevented. On this basis, the candidate pixels are 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 align with simultaneous wind direction and origin from explicit infrastructures. Here the high-resolution satellite imageries are taken from the Google Map. The hourly wind field data also come from the ERA5 reanalysis dataset. Finally, we manually draw polygons to mask such resulting plumes out. As preparation for plume emission quantification, we remove the background using the threshold of the median values of the scenes.

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

2.4 Multi-tiered quantification of methane emissions

In our framework, we calculate 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 define the background levels as the 10% of the average methane concentrations in the TROPOMI-based and PRISMA-based scenes (Figs. 1b ~ 1g) (Frankenberg et al., 2016; Varon et al., 2018). On this basis, we eliminate the interferences from the background concentrations and calculate IMEs as the methane masses of the masked hotspots and plumes.

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

$$Q = \frac{U_{eff} \cdot IME}{L}. \text{ (Eq. 3)}$$

Specifically, the IME and L can be inferred from the observations of the hotspots or plumes. During this process, we carefully apply a Boolean plume mask that separates the pixels (i) with notable signals ($\Delta\Omega_i$) from background pixels and thus defines the total areas ($\Sigma_{i=1}^{N} A_i$) of the hotspots or plumes. The L is defined as the square root of the total plume areas. Hence, the IME is calculated as follows:

$$IME = \Sigma_{i=1}^{N} \Delta\Omega_i A_i. \text{ (Eq. 4)}$$

In the first tier of our framework, the effective wind speed (U_{eff}) is 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 are used.

In the second tier of our framework, we apply an ensemble of large eddy simulations (LES) to establish an empirical, linear relationship between U_{eff} and the measured 10-m wind speed U_{10} as follows (Fig. S8)

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

The configurations of these simulations, such as spatial resolution and precision, are comparable to our PRISMA data. Other details in this methodology were described in Varon et al. (2018) (Varon et al., 2018).

We estimate the uncertainties of Q by propagating the random errors in U_{10} and IME. This processes are conducted in previous studies (Cusworth et al., 2019, 2021b; Itziar et al., 2021). As shown in previous findings, the major error source come from the U_{10} term. Its random distributions typically correspond to the 50% random error. On this basis, this error is integrated quadratically with the standard error of the IME, the result of which can be treated as the final random error of Q. The intrinsic errors of the IME model are quantified in the following uncertain analysis.

2.5 Uncertainty Analysis

The objective of this work is to promote a multi-tiered satellite constellation that can monitoring global methane pledges. To better understand the performance of our framework, we conduct comprehensive uncertain analysis. Note that the protocol of the uncertain analysis on our framework origins from previous studies (Itziar et al., 2021; Varon et al., 2020). Specifically, we require to account for the uncertainties in the TROPOMI-based and PRISMA-based methane retrievals and subsequent

emission estimates. Therein the operational TROPOMI-based methane retrieval products have been evaluated strictly and proved to be reliable globally (except in low- and high-albedo and snow-covered areas) (Lorente et al., 2021; Sha et al., 2021). In this work, we thus focus on three main sources of uncertainties, specifically including (1) uncertainties in the PRISMA-based methane retrievals; (2) uncertainties in the TROPOMI-based methane emission estimates; and (3) uncertainties in PRISMA-based methane emission estimates. During the analysis for the latter two uncertain sources, we would further investigate the potential wind impacts on the methane emission estimates. Note that it remains challenging to directly quantify the uncertainties in the wind fields across our cases due to the lack of measurements. We would thus assess the 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.

The detection limit of this framework depends mainly on the TROPOMI-based and PRISMA-based methane retrievals, which have been well discussed in previous studies (Guanter et al., 2021; Hu et al., 2018). As the robust relationship between the "minimum source" and the related methane enhancement interpreted by Jacob et al. (2016) and Guanter et al. (2021), the detection threshold for the TROPOMI instrument is 4000 kg/h with a wind speed of 5 km/h. Following the same relationship in the PRISMA instrument, we estimate that a retrieval precision of 114 ppb (6.1% with the assumed background concentration of 1850 ppb), such as in the case of the Hassi Messaoud site (Fig. S10e1), would lead to a detection limit of 800 kg/h for the same wind speed (analogous to the reported range of 500 ~ 900 kg/h) (Guanter et al., 2021; Irakulis-Loitxate et al., 2022). Similar instruments and detection limits are generally comparable to emissions from anthropogenic sectors, like O&G and coal mines in this study or landfills, agriculture, and waste management in previous studies (Lauvaux et al., 2022; Maasakkers et al., 2023; Sadavarte et al., 2021). However, no conclusive evidence shows by far that short-term (e.g., daily) satellite-based measurements with such detection limits can capture methane hotspots driven by natural sources (e.g., wetlands). In contrast, long-term (e.g., year-round) satellite-based measurements with much higher detection limits have shown the potential (Pandey et al., 2021).

3 Results and discussions

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 clarifies a methane-abundant region and further focuses on explicit super-emitters. Among them, five methane-abundant regions are captured in Wattenberg (the United States), Yangquan (China), Rumaila (Iraq), Burgan (Kuwait), and Hassi Messaoud (Algeria) (Fig. 1a and Table S1). These account for 4805 ~ 46138 kg/h methane emissions based on our daily first-tiered (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 rank among the top 1% regarding emission intensities per unit area (km^2) (Fig. S1) (Crippa et

al., 2020). The Rumaila field, for example, is known as the largest oil field in Iraq (in terms of both reserves and yields). In this work, it is found with a significant methane emission intensity exceeding 45000 kg/h (Fig. 1b). In addition to such well-known oil fields (Figs. 1c ~ 1f), methane hotspots emerge in developing coal mines, like Yangquan, with comparable emission levels (> 30000 kg/h) (Fig. 1g).

We attribute these methane enhancements to specific methane plumes via the second-tiered (i.e., PRISMA-based) monitoring (Figs. 1b1 ~ 1g2). There are substantial variations in the methane plumes' amounts, types, and magnitude, even in a single methane-abundant region. For instance, in the Burgan field, the second-tiered monitoring detects up to eight methane plumes in a handful of grids in the first-tiered monitoring (Figs. 1c1 ~ 1c4 and 1d1 ~ 1d4). Such intensive distributions are also found in previous region-oriented surveys in the Permian basin and California (Duren et al., 2019; Itziar et al., 2021). Together with high-definition images (Fig. S2), we find that such plumes origin from various sources, such as flares, factories, and wells. A breakthrough is the capture of two distinctive plumes in an individual methane source with extremely high emissions (> 10000 kg/h), unprecedented in previous satellite-based exploration and only observable in aircraft surveys (Fig. 1b1). Such precise distinctions benefit from the high resolution of the second-tiered monitoring, despite being limited by the relatively higher detection threshold (~ 300 kg/h) (Guanter et al., 2021). Besides, factories and wells can also emit such evident plumes (Fig. 1c1 and Figs. 1e1 and 1e2). By comparison, other plumes are typically more diffuse but with comparable 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) (Figure 1). Specifically, for a given set (including both a methane-abundant region and associated super-emitters), the overpass timing of TROPOMI can be nearly concordant with that of PRISMA. The temporal gaps could be frequently controlled within 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, 2021, November 15th and 17th, 2021), our multi-tiered satellite constellation goes through the Hassi Messaoud field and the Yangquan coal mine and provides 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 uncover methane enhancements in the Wattenberg field (Fig. 1f) but also track them back to explicit methane super-emitters (Figs. 1f1 and 1f2). As expected, if we extend the monitoring window of our framework to years, more methane super-emitters are subsequently captured (Fig. S3). Moreover, our framework via multi-tiered satellite constellation paves an in-time way for routine monitoring of global methane hotspots and associated super-emitters.

3.2 Multi-tiered verification of global methane super-emitters

Four unexpected cases occur in Burgan (Iraq), Hassi Messaoud (Algeria), and Yangquan (China), potentially explainable if we take mutual verification of the first- and second-tiered monitoring into consideration. First, an anomalous methane plume is detected in the Burgan field (Fig. 1c4) of high emission magnitude (> 1500 kg/h), notably exceeding typical O&G facilities, from an elusive source (i.e., no clear source could be attributed) (Fig. S2). The long-term measurements of our multi-tiered

satellite constellation intermittently, rather than accidentally, observe this abnormal plume (Figs. S4). Furthermore, uncertain analysis (see Materials and Methods) helps to confirm this real plume. In particular, the methane plumes are clearly uncorrelated with the surface brightness from space (Fig. S4). Consequently, the most likely hypothesis for this super-emitter is methane leakage from gigantic O&G pipelines as shown in the Google Map (Fig. S2).

Second, we observe suspect trails of methane plumes above the storage tanks in the Burgan field (Fig. 1d4). Conceivably, the technical noise driven by albedo effects bore the brunt, although it is believed to be corrected reliably (See Materials and Methods). To this end, we apply a multi-spectral retrieval algorithm to eliminate this effect to a large extent. The detailed illustrations are shown in Supplementary Information (Fig. S5). Consequently, we provide 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 tanks. This is only seen in previous aircraft-based surveys (Frankenberg et al., 2016). Therefore, our multi-tiered outcomes 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 and innumerable O&G fields.

Third, our framework detect 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. S6), 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 appears 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).

Figure 2 illustrates that, in our multi-tiered satellite constellation, the extent to which the explicit plumes in the second tier explain the regional budget detected by the first tier. The overpass moments are explicitly shown Fig. 1, most of which are inconsistent between for the first- and second-tier monitoring. Overall, the plumes in the former are mostly responsible for large shares (> 8.2%) of regional budgets in the latter. In the Rumaila, Burgan, and Wattenberg fields, the detected methane plumes play a more critical role, with contributions up to 53.8 ~ 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 comparison, the relatively low but still significant contributions in the Hassi Messaoud field (8.2%) and Yangquan coal mine (35.7%) are partly due to the technical limitation of our framework in detecting methane plumes on top of high background 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), is possibly applicable worldwide. To further explore such a hypothesis, we extend the temporal sample window of our multi-tiered framework. Using year-round snapshots in the second tier of our framework, we inspect the identified super-emitters (Figs. 1b ~ 1g) repeatedly and find more methane plumes as expected (Fig. S3). This reinforces the above hypothesis for the widespread occurrence of methane super-emitters. Note that there are differences in the order of magnitude between the TROPOMI-based and PRISMA-based results. The main cause is that the TROPOMI-based and PRIMSA-based results represent the methane emissions from different special scales.

The former results represent region-scale methane budgets, while the latter ones resolve the emission magnitude from the individual methane super-emitter therein (Fig. 1). Although the latter results can explain a large fraction of the former ones (Fig. 2), the gaps remain mainly due to inconsistent overpass moments between the two-tiered results or sources still missed by the PRIMSA-based results. In other words, closing the temporal gaps between the two tiers or improving the detection ability of the second tier would help to reconcile the first- and second-tiered results.

A regional survey in a California field is considered as the best reference, owing to its utilization of systematic airborne measurements to detect and quantify methane super-emitters (Duren et al., 2019). The California survey aims to provide the first view of methane super-emitters across the state. This survey is conducted with the Next Generation Airborne Visible/Infrared Imaging Spectrometer (AVIRIS-NG), with 5 nm SWIR spectral sampling, 1.8 km view field, 3 m horizontal resolution, and 3 km cruise altitude, and contains five campaigns over several months from 2016 to 2018. Moreover, this instrument is unique due to its high signal-to-noise ratio and is capable of characterizing methane super-emitters with emissions as small as 2 ~ 10 kg/h for typical surface winds of 5 m/s. They survey reports 1181 methane plumes, more than 500 times larger than previous aerial studies (Englander et al., 2018), with a median emission intensity of 170 kg/h. These results are thus used to directly evaluate the outcomes in the second tier (Fig. 3). Even though some regions of interest in this study are far less famous than the California field, their emission intensities are much higher. Specifically, these plumes detected by the second-tiered monitoring have emission intensities (1142 ~ 11698 kg/h) that exceed the median value in the California field. Satellite-based surveys are conducted repeatedly for the Permian basin (one of the top O&G bases worldwide) from 2019 to 2020 (Fig. 3). The Permian survey takes advantage of imaging spectroscopy technologies to provide the first spaceborne region-scale and high-resolution survey of methane super-emitters in the Permian basin. This survey is acquired by 30 hyperspectral images from three satellite missions, including Gaofen-5, ZY1, and PRISMA, and focuses on an area of roughly 200×150 km^2 in the Delaware sub-basin of the Permian basin within several days (mostly on four different dates: 15 May 2019, 1 November 2019, 29 December 2019, and 8 February 2020). More technical details on these two surveys can be found in previous studies (Duren et al., 2019; Itziar et al., 2021). Compared to the surveys in the California field, those in the Permian basin achieves a much higher number of strong methane super-emitters, the median emission rates (1850 kg/h) much closer to ours (2888 kg/h). Collectively, although such comparisons are not quantitative comparisons due to measurement divergencies between these datasets (e.g., spatial resolution and detection limit), they offer further context for the emission magnitude of the identified methane super-emitters and indicate the outstanding strength of our results that could be analogous to abundant outcomes from field campaigns. More importantly, this highlights the urgent need for global monitoring of 'nameless' O&G facilities that possibly emit methane as much as the California field and Permian basin.

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 is 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 is mostly consistent with the present results (-49.9 ~ 91.8%), with a fine average bias (63.2%). The Hassi Messaoud field in Algeria is a unique exception, where the O&G sector is in rapid development, with a relatively larger bias (489.2%). By comparison, this inventory significantly undervalues 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. S7). Moreover, the EDGARv6.0 is designed for the year 2018, missing the newly established O&G plants with high methane emissions. Second, in principle, conventional inventories directly miss 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 is downwind measurements, yet insufficiently reliable as shown in previous findings (Alvarez, Ramón A. and Zavala-Araiza, Daniel and Lyon, David R. and Allen, David T. and Barkley, Zachary R. and Brandt, Adam R. and Davis, Kenneth J. and Herndon, Scott C. and Jacob, Daniel J. and Karion, Anna and Kort, Eric A. and Lamb, Brian K. and Lauva, 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 (Alvarez, Ramón A. and Zavala-Araiza, Daniel and Lyon, David R. and Allen, David T. and Barkley, Zachary R. and Brandt, Adam R. and Davis, Kenneth J. and Herndon, Scott C. and Jacob, Daniel J. and Karion, Anna and Kort, Eric A. and Lamb, Brian K. and Lauva, 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 aligns 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.

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.

428 Our data prove that depending on ready-made satellite missions alone can initiate immediate, proactive monitoring of global

429 methane pledges, in contrast to existing surveys that have to focus on a priori methane-abundant regions. As such, as the

430 window for achieving the Paris target is rapidly closing, we will not need to sit back and wait for upcoming space missions,

431 like MethaneSAT and SBG in the United States, EnMAP in Germany, a new version of GF-5 in China, and, later, the European

432 Space Agency's CHIME from 2025 to 2030 (Cusworth et al., 2019). In addition, while scientific communities persistently

433 debates the drivers of the recent methane surge (Nisbet et al., 2014, 2019; Turner et al., 2019), the consequences of our

434 outcomes are clear, not only holding clues but also facilitating mitigation.

435 It should be noted that the multi-tiered framework is sustainable (Fig. 5). First, it can harmonize multiple satellites. The

436 potential representatives include upcoming official missions (e.g., the GF-5) (Itziar et al., 2021), current private constellations

437 (e.g., the GHGSat series) (Jervis et al., 2021; Varon et al., 2020), and explorable multispectral products (e.g., the Worldview-

438 3 and Sentinel-2) (Sánchez-García et al., 2021). Second, the framework is not confined to satellites and can be expanded by

439 integrating in situ (e.g., Global Atmosphere Watch Programme) (World Meteorological Organization, 2022), aircraft, and

440 unmanned aerial vehicles (UAVs) (Cusworth et al., 2020; Gålfalk et al., 2021; Tuzson et al., 2020). Note that such a multi-

441 tiered framework based on multifarious satellites, aircrafts, and UAVs keeps pursuing wider coverages and faster revisits. We

442 would thus derive the next objective in this manner, i.e., how to achieve effective, efficient, and economic monitoring of global

443 methane pledges, in which how to make better coverage-resolution balance between instruments is crucial. This will be the

444 topic of the next separate study. Third, nighttime methane monitoring is important because abnormal leakages or pulses might

445 also occur during nighttime (Plant et al., 2022; Poindexter et al., 2016). In these events, the LIDAR-equipped ones (involving

446 satellites, e.g., MERLIN) can allow to retrieve methane fluxes at all-latitudes, all-seasons, and all-weather (involving nighttime)

447 as they are not relying on sunlight. Third, better characterizing methane vertical profile would help to optimize our analysis,

448 like minimizing the uncertainties in tropospheric air mass factors and subsequent methane enhancements. Finally, on the basis

449 of our framework, rapid advances in artificial intelligence (AI) techniques are projected to completely replace manpower to

450 seek faint signals of methane enhancements in Earth's surface, and to significantly optimize data-driven algorithms of methane

451 emission estimates (Reichstein et al., 2019; Yuan et al., 2020). In principle, subsequent mitigation of such super-emitters via

452 routine maintenances, leak detections, or emergent repairs can provide effective, efficient, and economic solutions toward the

453 Paris target (Mayfield et al., 2017).

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

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

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

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

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

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

460 negotiations between countries.

461

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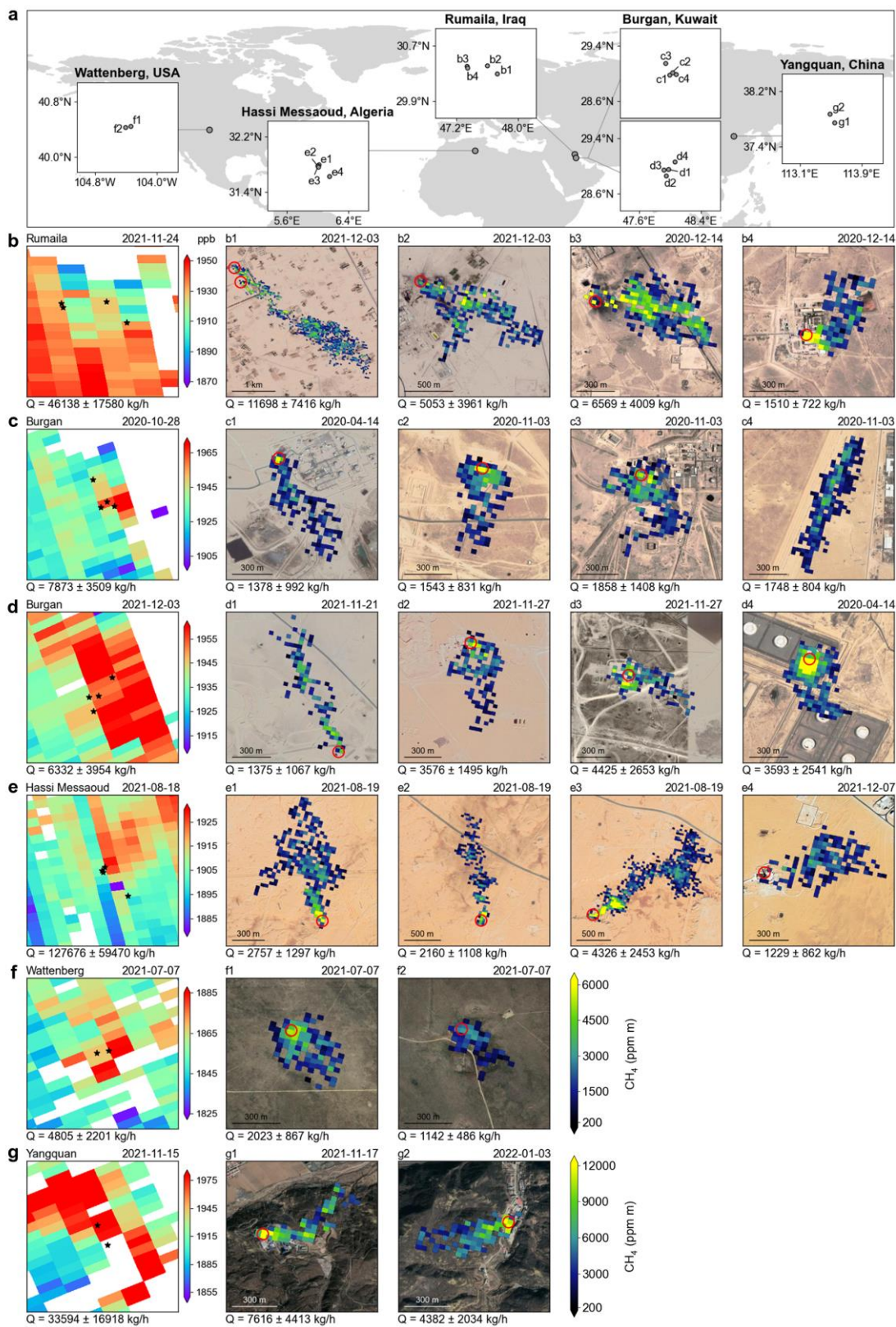
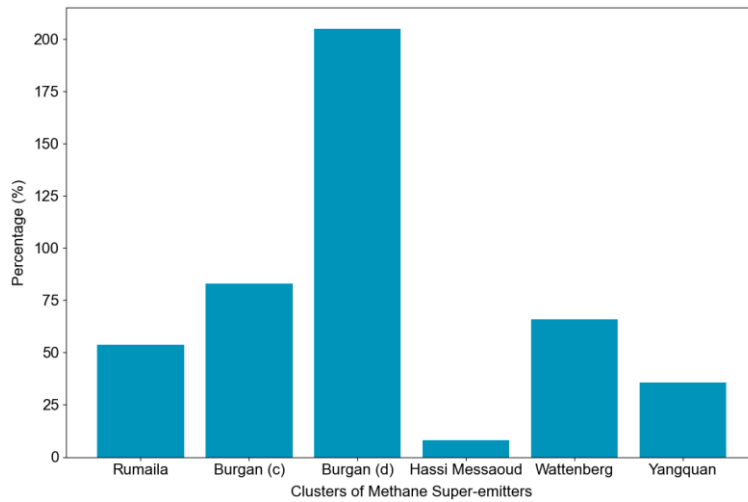
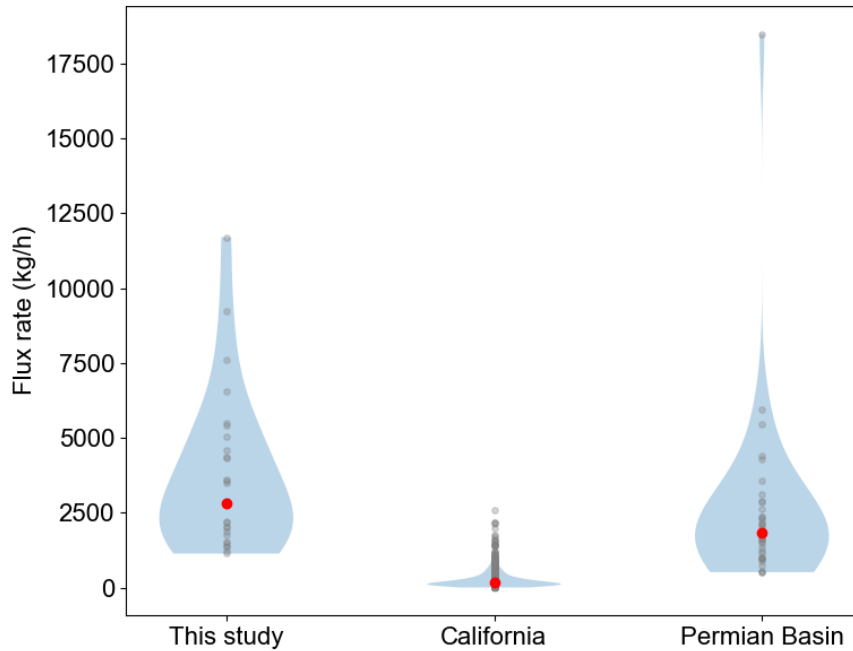


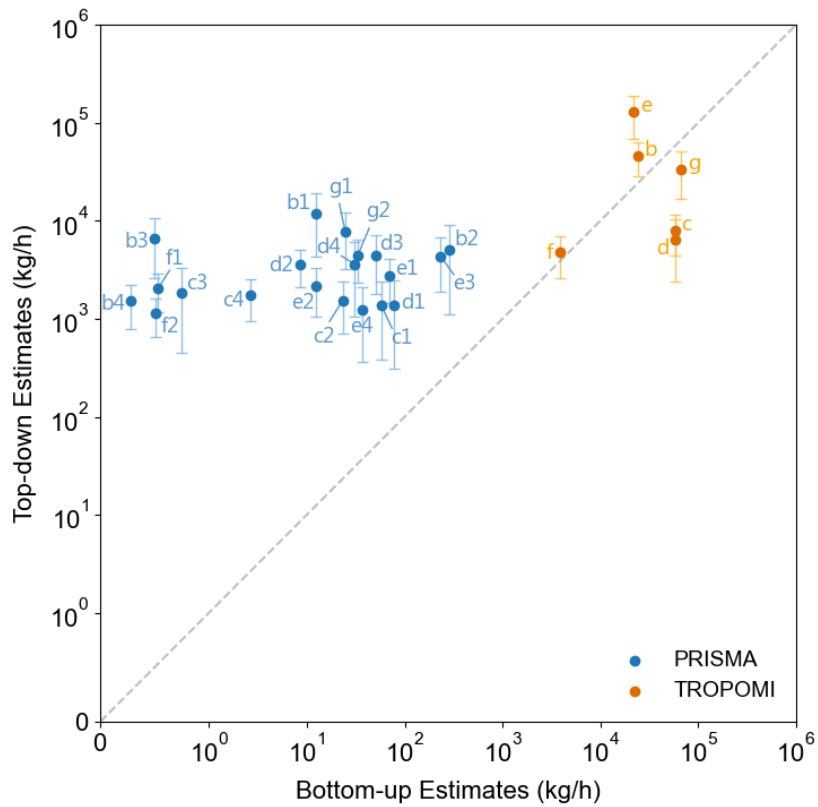
Fig. 1. Methane hotspots and associated super-emitters across the United States, China, Iraq, Kuwait, and Algeria via the multi-tiered daily satellite constellation. (**a**) Methane-abundant regions and associated super-emitters are captured by the TROPOMI and PRISMA, respectively. Their locations are marked by black rectangles and dots. Their names are obtained from the Google Map, usually being the names of the nearest O&G fields and coal mines. (**b ~ 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-emitter (five-pointed stars), the overpass moments of the multi-tiered satellite constellation and the consequent emission 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. Plume sources in the PRISMA results are marked by red circles.

472

473 **Fig. 2. High contributions of methane super-emitters to corresponding regional methane budgets.**

474

475 **Fig. 3. Comparison of emission estimates of methane plumes between surveys.** The surveys for the California field and

476 Permian basin are selected as the references. They report 1181 and 39 methane plumes, while our second-tiered survey attempts

477 29 plumes. Violin plots show statistical distributions of methane plume emission rates for these surveys. For each survey, the

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

479
480 **Fig. 4. Multi-tiered emission estimates versus bottom-up emission inventories.** We first interpolate the bottom-up emission
481 inventories into the resolution consistent with our multi-tiered results. On this basis, the bottom-up emission rates in the grids
482 that the detected hotspots and plumes cover are summed up to compare with the results. The detected hotspots (yellow dots)
483 and plumes (blue dots) correspond to those as shown in Fig. 1. The grey dashed line represents the ratio of the bottom-up
484 emissions to the top-down ones of 1:1.

485

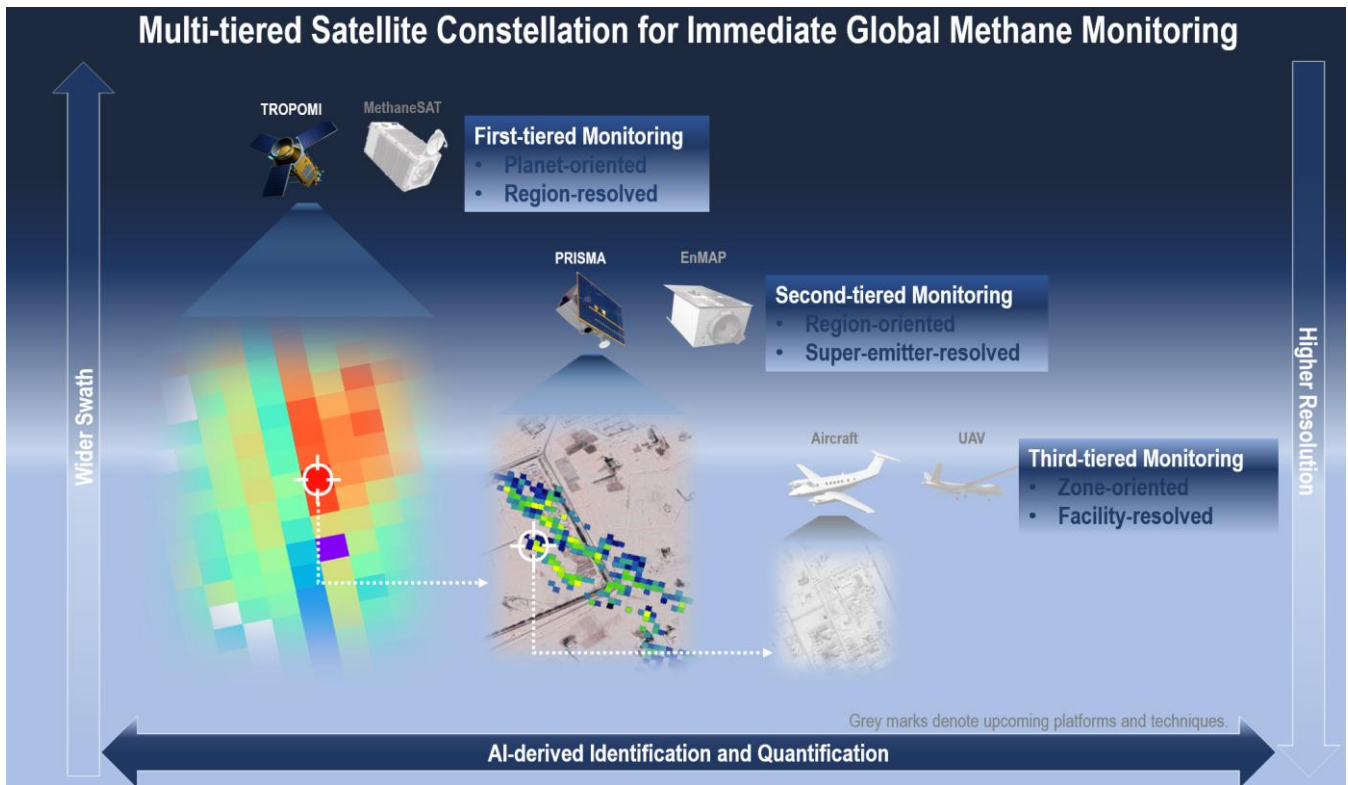



Fig. 5. Multi-tiered satellite framework for immediate global methane monitoring. This framework harmonizes global-scale and high-resolution methane retrievals, with a dual focus on mapping region-scale and plant-level drivers. In this work, the framework reconciles the spacious swath of TROPOMI (i.e., ~ 2600 km) with the high resolution of PRISMA (i.e., 30 × 30 m^2), in contrast to conventional satellite-based surveys that were of either insufficient samplings or narrow views. Looking 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 Paris target, at least while a super methane satellite with spacious swath, high resolution, and agile analysis is not in place, our multi-tiered satellite constellation has important implications for measuring global methane pledges. The appearances of the 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 TROPOMI and PRISMA refer to the results in Figs. 1e and 1b1. The grey marks indicate upcoming platforms (i.e., MethaneSAT and EnMAP) and techniques (e.g., AI techniques that can optimize the identification and quantification of methane super-emitters).

500 **Table 1. Spaceborne measurements for global methane monitoring.**

Satellite	Coverage/ Swath	Pixel Size (km^2)	SWIR (nm)	Spectral Resolution (nm)	Overpass (Local Time)	Period	Reference
SCIAMACHY	960 km	30×60	1630–1670	1.4	10:00	2002–2012	(Frankenberg et al., 2006)
GOSAT	790 km	10×10	1630–1700	0.06	13:00	2009–present	(Kuze et al., 2016)
GOSAT-2	1000 km	10×10	1630–1700, 2330–2380	0.06	13:00	2018–present	(Suto et al., 2021)
TROPOMI	2600 km	5.5×7, 7×7	2305–2385	0.25	13:30	2017–present	(Butz et al., 2012)
Sentinel-3	1420 km	0.5×0.5	1580–1640, 2230–2280	0.025	10:00	2016–present	(Pandey et al., 2022)
GHGSat	12×12 km²	0.05×0.05	1600–1700	0.3–0.7	9:30	2016–present	(Varon et al., 2018)
PRISMA	30×30 km²	0.03×0.03	1600–1700, 2200–2500	10	10:30	2019–present	(Guanter et al., 2021)
GF-5	60×60 km²	0.03×0.03	2100–2400	10	13:30	2018–present	(Irakulis-Loitxate et al., 2022)
ZY1	60×60 km²	0.03×0.03	2100–2400	10	10:50		(Irakulis-Loitxate et al., 2022)
Landsat-8	185×185 km²	0.03×0.03	2300	200	10:50	2013–present	(Ehret et al., 2022)
Sentinel-2	290 km	0.02×0.02	1610, 2190	200	10:30	2015–present	(Varon et al., 2021)
Worldview-3	66.5×112 km²	0.0037×0.0037	2295–2365	50	10:30	2014–present	(Sánchez-García et al., 2021)
EnMAP	30×30 km²	0.03×0.03	1600–1700, 2200–2450	10	11:00	2020–present	(Cusworth et al., 2019)
EMIT	80 km	0.06×0.06	1600–1700, 2200–2510	7.4		2022–present	(EMIT, 2023)

Data availability.

The operational TROPOMI product is available at https://scihub.copernicus.eu/, https://www.temis.nl/emissions/data.php. 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 EDGARv6.0 dataset comes from https://edgar.jrc.ec.europa.eu/gallery?release=v60ghg&substance=CH4§or=TOTALS.

Code availability.

The codes are available upon request to corresponding author.

Supplementary information.

Supplementary information accompanies this paper.

Author contributions.

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. H., and Y. P. performed the data analysis. S. Y., A.B., D. R., and J. H. S. contributed to the manuscript.

Competing interests.

The authors declare no competing interests.

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