Reply to comments on "Toward a versatile spaceborne architecture for immediate monitoring of the global methane pledge" by Yuchen Wang et al.

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5 **Reply to Reviewer #1:**

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This paper proposes an interesting method to address the important issue of quantifying current methane emissions. The authors justifiably argue that no current satellite instrument provides both the coverage and the spatial resolution to accurately measure global methane concentrations; to address this lack they propose a two-step method that uses data from two very different instruments: the wide swath, coarse spatial resolution TROPOMI and narrow swath but very high spatial resolution PRISMA. The TROPOMI data are used to locate high methane emission regions and the methane hotspots within these regions, then the co-located PRISMA data are examined for the presence of plumes. Emissions over the hotspots and plumes are estimated by combining wind speed information with an integrated mass enhancement model.

The approach is demonstrated for short periods over five small regions and the results are compared with surveys over two other regions. The median and range of the plume emissions are qualitatively consistent with those obtained using data from another (non-specified) satellite instrument over the Permian basin, and much higher than those from an aircraft campaign over California. The hotspot and plume emissions are also compared with emissions from the EDGAR_v6.0 inventory; the hotspot emissions were somewhat consistent with the inventory, while the plume emissions were much higher.

Summarizing the above, this is an interesting method with very interesting results. The authors evidently put a great deal of effort and enthusiasm into this work. However, the paper presents several problems, principally lack of detail on how some of the results were obtained. I have listed the main technical issues below, which need to be addressed before the paper can be published. An overarching issue is English language usage. Verb tenses are frequently used incorrectly (e.g, past or conditional future for present), and nouns and adjectives are interchanged. Before resubmitting the authors should have either a native English speaker or someone with excellent English revise the paper. I will be happy to provide more specific wording changes once this been done, if they are still necessary.

26 Response: We truly appreciate these positive responses and thorough summarizations. We are also very grateful for the 27 valuable comments and suggestions and have addressed all of them in our revised manuscript. Particularly, we have 28 supplemented more technical details to clarify the procedure of our framework. In addition, our co-authors (involving native 29 English speakers) have carefully gone through the entire manuscript to improve the English level.

The followings are our point-to-point responses to the reviewer's comments. The responses are shown in brown font,while the added/rewritten parts are presented in blue font. All revised figures and tables are also included in the manuscripts.

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33 1. The method for identifying high emission areas and plumes appears to be visual identification. The authors do mention 34 a Boolean mask for identifying the former, but no details are provided and the reader is left wondering what this means. This 35 needs to be clarified. Such an intensive method is feasible for a small analysis, such as presented in figures 1-3, but obviously not for long term, global emission estimates. Here the authors suggest a machine learning approach for further applications of 36 37 their method, which is a reasonable suggestion. However, this issue makes the year long results presented in S3 and S4 38 questionable. Were the TROPOMI maps obtained by applying the Sun oversampling method for an entire year over the original 39 methane concentrations? If so which wind fields were used to obtain the emissions, both for the regional and plume estimates? 40 How were the PRISMA data averaged over the year? Given the variability in wind direction, I don't think it makes sense to 41 look for plumes in averaged data. These plots need to either explained in much greater detail, or omitted entirely from the 42 paper. If they are to be included, then the authors need to be clear which results (short term or annual) are used in all other 43 plots.

44 **Response:** Thank you for these valuable comments and suggestions. First, we have supplemented more technical details 45 to clarify the role of the Boolean mask method. As you pointed out, in the first tier of our framework, we apply visual inspection to identify methane hotspots using the TROPOMI-based methane retrievals. The transformation from visual inspection to 46 47 automatic recognition would significantly advance long-term, global methane monitoring. However, no satisfactory set of 48 criteria is found that could be suitable for this study. This was mainly because, in localized regions, methane budgets respond 49 to the changes in not only super-emitters but also complex external factors (e.g., meteorology, topography, and background 50 concentrations). Similar compromises are also adopted in previous studies. Therefore, automatic recognition enabled by 51 artificial intelligence would play an essential role in the versatile spaceborne architecture for long-term, global methane 52 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 \pm standard deviation) is defined by an upwind sample of the measured columns, in which the hourly wind field data came 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).

Second, we agree that it might make no sense to look for plumes in averaged data due to the variable wind direction and have thus omitted the oversampled methane maps in the first tier of our framework (Fig. S3). In turn, 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. S4). This reinforces the above hypothesis for the widespread occurrence of methane super-emitters.

Added/rewritten part in Sect. 2.3: In the first tier of our framework, we apply visual inspection to identify methane
 hotspots using the TROPOMI-based methane retrievals. The transformation from visual inspection to automatic recognition

would significantly advance long-term, global methane monitoring. However, no satisfactory set of criteria is 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., 2012; Paoletti et al., 2018; Yang et al., 2018; Yu et al., 2017; Zhang et al., 2018).

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80 Added/rewritten part in Sect. 3.2: To further explore such a hypothesis, we extend the temporal sample window of our 81 multi-tiered framework. Using year-round snapshots in the second tier of our framework, we inspect the identified super-82 emitters (Figs. 1b ~ 1g) repeatedly and find more methane plumes as expected (Fig. S3). This reinforces the above hypothesis 83 for the widespread occurrence of methane super-emitters.

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85 2. The plume maps would be more interesting if the plume source were clearly marked.

Response: Thanks. We have marked all the plume sources in Fig. 1 and Fig. S3.

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3. How was the background vector used in equation 1 derived?

Response: Thanks. We have supplemented brief descriptions for this issue. The $\vec{\mu}$ and Σ represent the mean background radiance and corresponding covariance, respectively, calculated with their common formulas after subtracting the current signal estimates from the data. Specifically, the $\vec{\mu}$ is calculated from the data with the removal of the most recent enhancement estimates, while the Σ is then calculated with updated $\vec{\mu}$ and the most recent enhancement estimates. More technical details are reported in previous studies (Foote et al., 2020). Note that, owing to the non-uniform response of individual detectors in PRISMA, they are calculated based on per-column spectrums in order to consider different responses of across-track detectors to radiance.

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100 al., 2020). Note that, owing to the non-uniform response of individual detectors in PRISMA, they are calculated based on per-

101 column spectrums in order to consider different responses of across-track sensors to radiance.

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4. What does this sentence mean: methane enhancements detected in spectrometers generally exhibit sparsity, especiallyover low albedo surfaces.

105 Response: Sorry for the confusion we caused. We have revised this sentence to clarify this issue. In principle, it would 106 be more difficult to detect methane enhancements in pixels over low-albedo surfaces. Although methane absorption is 107 independent of albedo, the resulting signal in absolute radiance is weakened with surface albedo decreasing.

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112 5. Please define the co-location criteria between the TROPOMI and PRISMA datasets.

113 Response: Thanks. We have supplemented the definition the co-location criteria between the TROPOMI and PRISMA 114 datasets. Regarding the identified regional hotspots, we also apply visual inspection to search for plumes within their 115 surrounding 30 km scales (i.e., corresponding to the swath width of PRISMA) in the second tier of our framework.

Added/rewritten part in Sect. 2.3: 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 (Irakulis-Loitxate et al., 2021; Lauvaux et al., 2022; Martin et al., 2018; Varon et al., 2020).

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6. The section on comparing the TROPOMI/PRISMA results with the California and Permian surveys needs to provide more detail on those surveys (instrument, time of year, temporal and spatial extent). It also needs to emphasize that these comparisons are basically tests of reasonableness, not true quantitative comparisons.

Response: Thanks. We have supplemented more technical details on these surveys. 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 $2 \sim 10$ kg/h for typical surface winds of 5 m/s.

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 km² in the Delaware sub-basin of the Permian basin within several days (mostly on four different dates: 15 May 2019, 1 November 2019, 133 29 December 2019, and 8 February 2020). More technical details on these two surveys can be found in previous studies (Duren

134 et al., 2019; Irakulis-Loitxate et al., 2021).

Moreover, we agree that such comparisons are basically reasonableness test rather than stringently quantitative validations due to measurement divergencies between these datasets (e.g., spatial resolution and detection limit). 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.

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160 7. The phrase "on a per column basis" is frequently used: what does this mean?

161 Response: Sorry for the confusion we caused. We have supplemented some sentences to explain this phrase at its first 162 appearance. The matched-filter algorithm focuses on the individual columns rather than the whole scene to resolve methane 163 enhancements. This means that the methane enhancement per column is calculated separately (i.e., methane enhancements 164 were calculated on a per-column basis). More explanations can be found in Guanter et al. (2021).

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 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).
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8. The detailed uncertainty analysis is confusing, disorganized and hard to follow. Please put some more thought in howto present this information.

171 **Response:** Thank you very much for this constructive suggestion. We have reorganized and revised the detailed 172 uncertainty analysis in Supplementary Information to clarify this issue, which has been explicitly divided into three sub-issues: (1) uncertainties in the PRISMA-based methane retrievals; (2) uncertainties in the TROPOMI-based methane emission 173 174 estimates; and (3) uncertainties in PRISMA-based methane emission estimates. Note that operational TROPOMI-based methane retrieval products have been evaluated strictly and proved to be reliable globally (except in low- and high-albedo and 175 176 snow-covered areas) (Lorente et al., 2021; Sha et al., 2021) and the related uncertainty analysis is thus omitted here. As a result, 177 we could confirm the reliable performance of our framework. Comprehensive uncertainty analysis is illustrated in 178 Supplementary Information.

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180 **Reference**

- Duren, R. M., Thorpe, A. K., Foster, K. T., Rafiq, T., Hopkins, F. M., Yadav, V., Bue, B. D., Thompson, D. R., Conley, S.
 and Colombi, N. K.: California's methane super-emitters, Nature, 575(7781), 180–184, 2019.
- 183 Foote, M. D., Dennison, P. E., Thorpe, A. K., Thompson, D. R., Jongaramrungruang, S., Frankenberg, C. and Joshi, S. C.:
- 184 Fast and Accurate Retrieval of Methane Concentration From Imaging Spectrometer Data Using Sparsity Prior, IEEE Trans.
- 185 Geosci. Remote Sens., 58(9), 6480–6492, doi:10.1109/TGRS.2020.2976888, 2020.
- 186 Hoffmann, L., Günther, G., Li, D., Stein, O., Wu, X., Griessbach, S., Heng, Y., Konopka, P., Müller, R., Vogel, B. and
- Wright, J. S.: From ERA-Interim to ERA5: the considerable impact of ECMWF's next-generation reanalysis on Lagrangian
 transport simulations, Atmos. Chem. Phys., 19(5), 3097–3124, doi:10.5194/acp-19-3097-2019, 2019.
- 189 Irakulis-Loitxate, I., Guanter, L., Liu, Y.-N., Varon, D. J., Maasakkers, J. D., Zhang, Y., Chulakadabba, A., Wofsy, S. C.,
- 190 Thorpe, A. K., Duren, R. M. and Others: Satellite-based survey of extreme methane emissions in the Permian basin, Sci.
- 191 Adv., 7(27), eabf4507, doi:10.1126/sciadv.abf4507, 2021.
- 192 Lauvaux, T., Giron, C., Mazzolini, M., d'Aspremont, A., Duren, R., Cusworth, D., Shindell, D. and Ciais, P.: Global
- assessment of oil and gas methane ultra-emitters, Science (80-.)., 375(6580), 557–561, 2022.
- 194 Lorente, A., Borsdorff, T., Butz, A., Hasekamp, O., aan de Brugh, J., Schneider, A., Wu, L., Hase, F., Kivi, R., Wunch, D.,
- 195 Pollard, D. F., Shiomi, K., Deutscher, N. M., Velazco, V. A., Roehl, C. M., Wennberg, P. O., Warneke, T. and Landgraf, J.:
- 196 Methane retrieved from TROPOMI: improvement of the data product and validation of the first 2 years of measurements,
- 197 Atmos. Meas. Tech., 14(1), 665–684, doi:10.5194/amt-14-665-2021, 2021.

- 198 Martin, Van, Damme, Lieven, Clarisse, Simon, Whitburn, Juliette, Hadji-Lazaro and Daniel: Industrial and agricultural
- ammonia point sources exposed, Nature, 2018.
- 200 Ouerghi, E., Ehret, T., de Franchis, C., Facciolo, G., Lauvaux, T., Meinhardt, E. and Morel, J.-M.: Detection of methane
- 201 plumes in hyperspectral images from SENTINEL-5P by coupling anomaly detection and pattern recognition, ISPRS Ann.
- 202 Photogramm. Remote Sens. Spat. Inf. Sci., V-3–2021, 81–87, doi:10.5194/isprs-annals-V-3-2021-81-2021, 2021.
- 203 Pandey, S., Gautam, R., Houweling, S., Van Der Gon, H. D., Sadavarte, P., Borsdorff, T., Hasekamp, O., Landgraf, J., Tol,
- 204 P. and Van Kempen, T.: Satellite observations reveal extreme methane leakage from a natural gas well blowout, Proc. Natl.
- 205 Acad. Sci., 116(52), 26376–26381, 2019.
- 206 Paoletti, M. E., Haut, J. M., Plaza, J. and Plaza, A.: A new deep convolutional neural network for fast hyperspectral image
- 207 classification, ISPRS J. Photogramm. Remote Sens., 145, 120–147, doi:https://doi.org/10.1016/j.isprsjprs.2017.11.021, 2018.
- 208 Sha, M. K., Langerock, B., Blavier, J.-F. L., Blumenstock, T., Borsdorff, T., Buschmann, M., Dehn, A., De Mazière, M.,
- 209 Deutscher, N. M., Feist, D. G., García, O. E., Griffith, D. W. T., Grutter, M., Hannigan, J. W., Hase, F., Heikkinen, P.,
- 210 Hermans, C., Iraci, L. T., Jeseck, P., Jones, N., Kivi, R., Kumps, N., Landgraf, J., Lorente, A., Mahieu, E., Makarova, M. V,
- 211 Mellqvist, J., Metzger, J.-M., Morino, I., Nagahama, T., Notholt, J., Ohyama, H., Ortega, I., Palm, M., Petri, C., Pollard, D.
- 212 F., Rettinger, M., Robinson, J., Roche, S., Roehl, C. M., Röhling, A. N., Rousogenous, C., Schneider, M., Shiomi, K., Smale,
- 213 D., Stremme, W., Strong, K., Sussmann, R., Té, Y., Uchino, O., Velazco, V. A., Vigouroux, C., Vrekoussis, M., Wang, P.,
- 214 Warneke, T., Wizenberg, T., Wunch, D., Yamanouchi, S., Yang, Y. and Zhou, M.: Validation of methane and carbon
- 215 monoxide from Sentinel-5 Precursor using TCCON and NDACC-IRWG stations, Atmos. Meas. Tech., 14(9), 6249–6304,
- 216 doi:10.5194/amt-14-6249-2021, 2021.
- 217 Varon, D. J., Jacob, D. J., McKeever, J., Jervis, D., Durak, B. O. A., Xia, Y. and Huang, Y.: Quantifying methane point
- sources from fine-scale satellite observations of atmospheric methane plumes, Atmos. Meas. Tech., 11(10), 5673–5686,
- 219 doi:10.5194/amt-11-5673-2018, 2018.
- 220 Varon, D. J., Jacob, D. J., Jervis, D. and McKeever, J.: Quantifying Time-Averaged Methane Emissions from Individual
- 221 Coal Mine Vents with GHGSat-D Satellite Observations, Environ. Sci. Technol., 54(16), 10246–10253,
- 222 doi:10.1021/acs.est.0c01213, 2020.
- 223 Yang, X., Ye, Y., Li, X., Lau, R. Y. K., Zhang, X. and Huang, X.: Hyperspectral Image Classification With Deep Learning
- 224 Models, IEEE Trans. Geosci. Remote Sens., 56(9), 5408–5423, doi:10.1109/TGRS.2018.2815613, 2018.
- Yu, S., Jia, S. and Xu, C.: Convolutional neural networks for hyperspectral image classification, Neurocomputing, 219, 88–
 98, doi:https://doi.org/10.1016/j.neucom.2016.09.010, 2017.
- 227 Zhang, M., Li, W. and Du, Q.: Diverse Region-Based CNN for Hyperspectral Image Classification, IEEE Trans. Image
- 228 Process., 27(6), 2623–2634, doi:10.1109/TIP.2018.2809606, 2018.
- 229