Author's response

Dear reviewer and associate editor,

We appreciate the positive attitudes and constructive comments provided by the reviewer and associate editor regarding our manuscript - *Separating and Quantifying Facility-Level Methane Emissions with Overlapping Plumes for Spaceborne Methane Monitoring*.

We have primarily clarified the methods description, definitions related to wind, and also addressed other concerns. Please see the point-by-point response as follows. We hope our refined manuscript will meet the high-standard requirement of AMT.

Response to Reviewer

- *"I thank the authors of "Separating and Quantifying Facility-Level Methane Emissions with Overlapping Plumes for Spaceborne Methane Monitoring" for the substantial update of this manuscript following the reviews and for their detailed answers to my comments, even when they have removed the corresponding sections in the manuscript. I also thank them for their transparency with regard to the bias in the comparison between the Gaussian plume model fitting and the IME quantification methods due to the limitation of the image size in their experiments."*

Response: Thanks for the approval. Your suggestions has greatly contributed to the improvement of our manuscript and we are delighted to mention it in the acknowledgement.

- *"Many of the updates are satisfying. However, I still push for a revision of the manuscript, because even though they partially tackled them, going in the right direction, the authors did not fully address some of the comments I have raised in the previous review. Here, I do not copy paste these comments from the 1st review, but I refer to them in the following:"*

Response: Thanks for reviewing our manuscript with patience and providing thorough and helpful feedback once again. I apologize for the misunderstandings in the last revision and hope the refinement this time can address all the comments adequately.

- *"I think that the abstract, introduction, section 2.1 and parts of the conclusion still lack a very clear and explicit overview of the split of the image processing into separation or "attribution" - detection (including the extraction of the enhancements above the background corresponding to the plumes) - quantification => the three steps that the authors promise to discuss in their answers to my comments, but which do not really appear as such in the new manuscript"*

Response: Thank you for suggesting we emphasize the description and discussion of the methods by these three stages. We believe this approach will significantly improve the clarity of our manuscript, and we intend to give it more elaboration in the content this time. We also realize that this could serve as the first clear distinction in this field, potentially benefiting future work.

- "*the 4 different combinations of methods for these 3 steps that will be tested:the method developed here, i.e., the sequence of Gaussian plume model fitting for separation, student's t-test for detection and then the IME for quantification vs. the sequence of student's t-test for the detection, the "pixel connectivity analysis" for the "attribution", and then the IME for the quantification vs. the single Gaussian plume model fitting, ignoring the problem of separation, and solving for the "attribution", detection and quantification all together (still extracting the background before this process ?) vs. multiple Gaussian plume model fitting, solving for the 3-steps all together, taking the separation problem as a problem of solving for several sources at once. => a table somewhere in section 2 may help clarify things"*

Response: We sincerely appreciate the direct suggestion. Your suggestion to add such a table helped clarify our manuscript's elaboration. We have included such a table (shown as table 1) in the manuscript. We have divided the methods into three stages, including (1) separation, which isolates overlapping plumes into individual images, each containing a single plume; (2) detection, which distinguishes the plume pixels from the background pixels; and (3) quantification, which calculates the emission rates of the point sources based on the identified pixels.

Table 1. Comparison of methods evaluated in this work.

Method	Separation	Detection	Quantification
Single-source Gaussian plume	/	Single-source Gaussian plume fitting	
Multi-source Gaussian plume	Multi-source Gaussian plume fitting		
UNSEP	/	Student's t-test & Connectivity filtering	IME method
SEP (ours)	Gaussian plume weighting separation	Student's t-test & Connectivity filtering	IME method

Table RC 1 Method combinations for quantification in the manuscript.

- *"the abstract should mainly clarify the separation between separation with Gaussian plume model and quantification with IME (line 4-5 misses something like "respectively" for this) and the alternative use of Gaussian plume model fitting for both (info missing at line 11). The other sections should bring this overall picture quite early before entering into technical details. In the introduction, lines 50-69, 73-79 and then 80-81/88/89 mix everything. The introduction of section 2.1 is focused on the new method and none of the following subsections will provide a clear overview highlighting in a distinct way the "3 steps" and the alternative methods: from 2.1.1, the text jumps into technical details."*

Response: Thanks for the suggestion. We refine the text for clear distinction in the abstract. We also improve the introduction accordingly. More importantly, we adopt the three-stage description and improve Section 2.1 with much clarity. We name our separation method as Gaussian plume weighting separation and make a clear distinction between our method and the other three method. We also clearly state the uniqueness of the detection method for IME, and the detection method is not considered a variant in this work.

- *"I think that the presentation of the "pixel connectivity analysis" could be improved: clarifying the fact that it attempts at "separating" plumes which do not overlap, but that it would merge all overlapping plumes into a single one (unless it* *includes some level of separation of overlapping plumes ?) ? I think that calling it a "pixel attribution" method is a bit misleading from that point of view."*

Response: Thanks for the advice. We rename this approach as connectivity filtering, which follows the t-test as the detection approach (shown in Table 1).

- *"- regarding the implicit optimization of sigma_y when optimizing the wind speed u in the frame of the Gaussian plume model fitting => the text is not really clear about this, sigma_y could have been fixed offline with an initial value given to u; so I think that the text should state it, and maybe the notation sigma_y(x) could be changed into sigma_y(x,u) to highlight it better ?"*

Response: Thanks for the advice. It's a good idea to highlight that σ_y is partially decided by wind speed u. However, this expression may be misleading that $\sigma_y(x,u)$ is a continuous function of u. In many definations, the $\sigma_y(x)$ functions are given in lists based on different terrain and stability (which is decided discretely by u and sunlight, i.e., decided solely by u in middle sunlight). To be rigorous, it should be written as $\sigma_y(x; u, \text{underlying condition}, \text{sunlight})$. For conciseness, we will omit the parameters in the mathematical formulas and clarify in the text that "σ_y represents the diffusion coefficient across-wind, is a function of downwind distance x and is decided by wind speed, underlying condition and sunlight (Briggs, 1973)."

- *"- observation noise: you should provide typical values using the same units (g/m2) as when plotting the plumes and compare them to the typical amplitude of the observed plumes.*

Response: It's a reasonable suggestion. However, the noise value in the concentration column (g/m^2) may vary with ground pressure and humidity. Typically, it is expressed as a percentage of the dry air column. In this domain, concentrations are usually expressed in parts per billion (ppb), so the unit is often given as ppb or a percentage. To make the content clearer, we've mentioned the typical value of noise level in g/m2 (Please see L217).

- *I think that section 3.2 should refer to the new supplementary material and summarize the conclusions from this supplementary material."*

Response: Thanks for the advice. We will mention it briefly in Section 3.2. The reason why we skip elaborating is that (1) those supplementary results are very similar to those in the content; (2) we focused on the environmental influence on the overlapping in exp2, and the observation factors don't directly decide the overlapping in our experiment. Please see L354-L355.

- *"- regarding the wind: the text is not clear; once having introduced equation 1 (l. 110-111), the authors directly speak about "the" 2D wind vector (implicitly: about the 2D effective wind) as if there was a clear definition for such a 2D wind, and later they have values for the true 2D wind (e.g. at lines 149-151, and then after equation 7, where they fit it with a log function of U10). However, the derivation of the effective 2D wind driving the 2D plumes seen from space can be a complicated topic. How do the authors get it when tackling the 3D LES simulations (it seems that they implicitly assume it to be the geostrophic wind, because of the similarity between the ranges at lines 206 and 250, which would raise questions) ? Could the optimal wind derived from the Gaussian plume model fitting differ from what the author assume to be the effective wind, because of a wrong derivation a priori of this effective wind ? In this case, may the use of the log function of U10 in the IME instead of the wind retrieved from the Gaussian plume fitting not be the best option ?"*

Response: Thanks for the advice. Our experiment involves four types of winds: (1) geostrophic wind, driving the LES simulation; (2) 10 m wind, loaded from a meteorological database to calculate effective wind speed for IME; (3) effective wind for IME; (4) effective wind for Gaussian plume. Similar to Varon et al. (2019), we derived the IME effective wind using the 10 m wind speed.

The Gaussian plume effective wind speed can be a relatively independent topic. In previous studies, such as Nassar et al. (2017), effective wind speeds are typically considered as the wind speed at the emission height. The effective wind speed for the Gaussian plume is coupled with the dispersion coefficients, defining the plume shape. Often, the dispersion coefficients are highly empirical, making the optimal wind speed differ from the actual horizontal wind speed at the emission height. In our application, our main objective is to fit the plume shape using the Gaussian plume model. Therefore, we use the optimal wind speed to mitigate the impact of inaccurate dispersion coefficients and achieve good agreement between the modeled and observed plume. Additionally, the relationship between effective wind speeds for the Gaussian plume and IME requires further discussion, as the definition of IME can also be empirical.

We also want to clarify the basis for matching 10 m and geostrophic wind speeds in exp3, where plumes are loaded according to ERA5 10 m wind speeds. As shown in Figure RC 1, our LES simulations established a boundary layer, and the relationship between geostrophic wind and 10 m wind can be expressed as $u_{10} = 0.86u_g + 0.26$ (Figure RC 2). Thus, we can match u_g using u_{10} from the meteorological database, given by $u_g = 1.16u_{10} - 0.30$. More details are elaborated in the supplement.

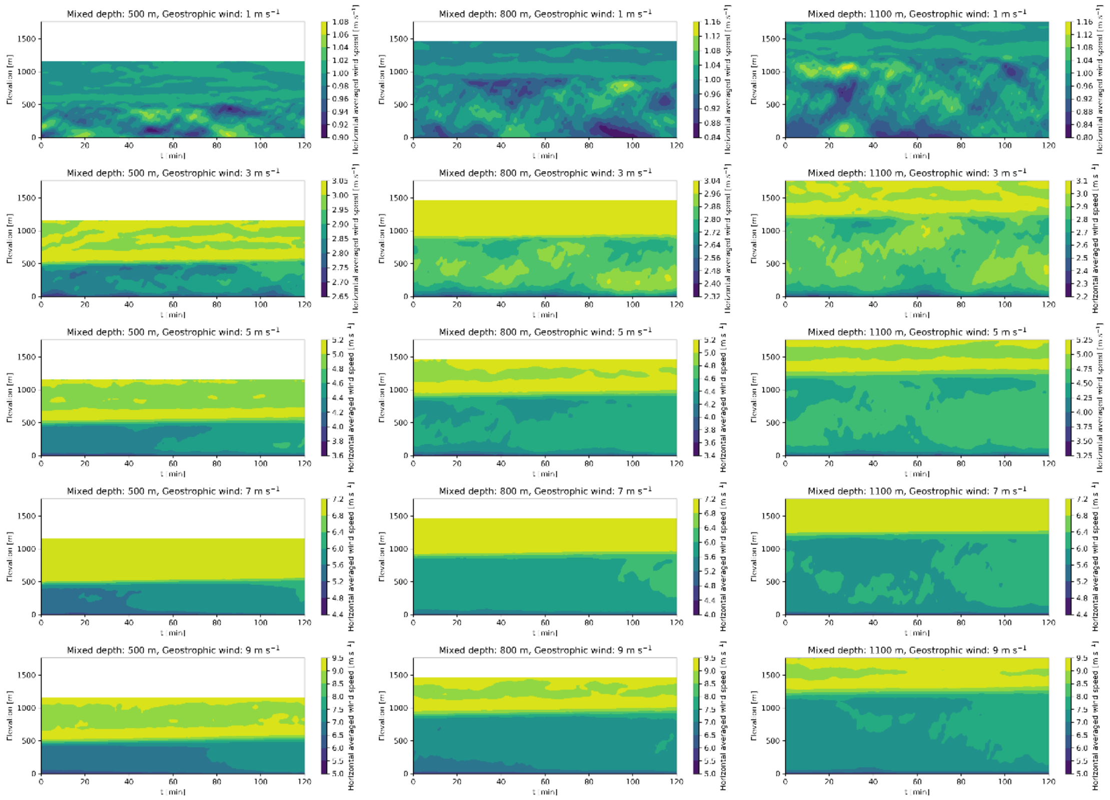

Figure RC 1 Distribution of horizontally averaged wind speed in the LES simulation.

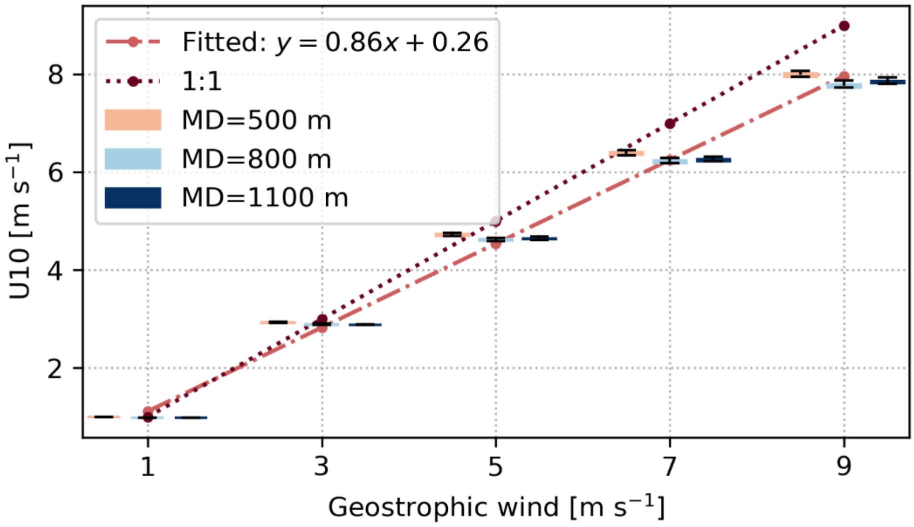

Figure RC 2 Relationship between the geostrophic wind (u_g) and 10 m wind (u_{10}) in the LES simulation.

- *"There was some misunderstanding regarding the background: in my comment, I was not speaking about the variations due to errors in the CH4 concentration retrievals. I was speaking about the impact of the CH4 sources located outside the satellite image. The frequent vicinity of sources within the images implies the frequent vicinity of other clusters of sources outside the images whose plumes all together may raise larger problems of overlapping than the sources within the image. There could also be areas sources close to the targeted sources, whose atmospheric signal overlaps the global background. If such a problem has been ignored, it should be explained in the method section. The manuscript should clarify whether the detection-quantification steps assume that the background field is uniform to derive the CH4 "enhancements" (the term is used but not really defined; it corresponds to the enhancements above the background). Lines 144-146 (and equation 5) completely ignore the background field (which adds to the general problem of the lack of overview on the "3 steps" to process the images that is discussed above)."*

Response: Thanks for the detailed clarification on the suggestion. We introduce the term "CH4 enhancements" directly from other research in this field. In this work, we consider the background uniform, and the plumes and noise solely contribute to the enhancements. The influences of the methane background are not considered as this topic is rather complicated and beyond the scope of this work. The plumes of near sources originating from outside the domain are considered well-mixed and thus can be treated as background, as supported by the results of Exp2. These sources do not contribute to the enhancements. We will clarify this point in the content.

- *"regression problem vs. parameter estimation problem in section 2.4.2: replacing the former by the latter does not solve for the issue here since the text still states in the next sentence "So, the R2 coefficient of determination is introduced to indicate the accuracy of overall estimation results", and at line 308 "regression results"*

Response: Thank you for pointing this out. We do not intend to debate the relationship between parameter estimation and regression problems, though these concepts are often mixed and not distinguished, with regression being performed through parameter estimation. To maintain coherence with the literature, we will refine the text accordingly.

- *"I still do not understand the "correction" of the SEP results in section 3.3: I understand that debiasing the results improve their accuracy, but what authorizes the authors to apply such a correction which is completely based on the knowledge of the true emissions ? What is the applicability of such a correction if considering experiments with real images ?"*

Response: Thank you for highlighting this issue. The introduction of Gaussian plume weighting can dilute the plume image and decrese the IME value. Therefore, when applying the IME method, u_{eff} needs recalibration with different plume pixel detection method. Previously, we used a "post-calibration" approach.

We are now pleased that we have solved this problem by introducing an additional calibration for u_{eff} with Gaussian plume separation practice, and the result is $u_{eff} = 0.94 + 0.64 \log(u_{10})$. In comparison, the result without separation is $u_{eff} = 0.62 + 0.55 \log(u_{10})$. We reprocessed the original results of exp3 and the new results of exp3 are shown in Figure RC 3 (Figure 6 in the manuscript). The present R2 and MAPE indicators are also comparable to previous results with "post-calibration". Necessary modifications are also made for the corresponding section.

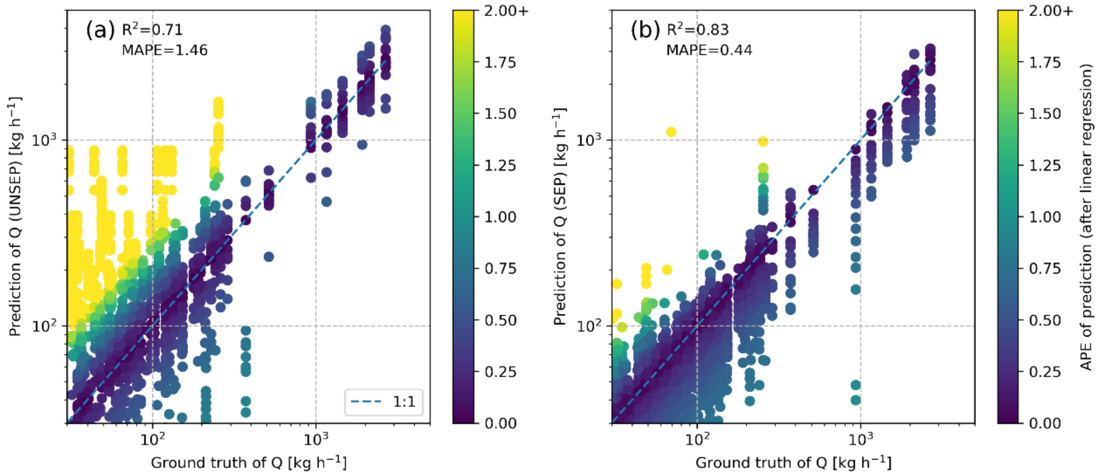

Figure RC 3 Comparison between quantification results of (a) unseparated quantification and (b) separated quantification in Exp3.

- *"- equations: C_n must be defined mathematically to be used in the right hand side of eq3; I still have the feeling that eq3 works with C instead of Cn in this right hand side, and, actually, line 130-131 is not consistent with the current eq3; l. 174: just say Cp is the modeling of plume p, but anyway, this definition has already be given around eq3 (don't redefine it several times; actually, C is redefined plenty of times throughout the manuscript); "*

Response: In our previous manuscript, C represents the concentration modeled by single Gaussian plume; C_n also represents concentration modeled by single Gaussian plume but with specified parameters for source n; C_N represents the summation of single Gaussian plumes, i.e., the multiple Gaussian plumes. It also seems that there are naming conflicts for the modeled concentration and observed concentration.

To eliminate the ambiguity, we will follow the following definations: (1) $C_{SG}(x, y; u, Q)$ or C_{SG} represents general single single Gaussian plume; (2) $C_{SG,i}(x_i', y_i'; u_i, Q_i)$ or $C_{SG,i}$ represents a specified single single Gaussian plume of source i; (3) C_{MG} represents the multiple Gaussian plume model; (4) $\Delta\Omega$ represents the observed column concentration.

- *"equation 8: should not you write i'_n j'_n ? should not eq8 look as similar as eq 3 and eq4 as possible (for the sake of clarity) since it's a similar process ? line 334: isn't it i' and j' rather than i and j ?"*

Response: Thank you for the suggestion, we will improve it accordingly.

- *“Please rewrite sentences such as: "Although there have been abundant spaceborne methane observations, these observations suffer from the demerit of the lack of priors" (l 197), "The LES run by WRF is thus a preferred option for spaceborne GHG monitoring ." (l 201-202)”*

Response: Thanks for the advice. Please see the refined text at L199-L202.