Efficient use of a Lagrangian Particle Dispersion Model for atmospheric inversions using satellite observations of column mixing ratios

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I thank authors for revising the manuscript by taking into account of my comments. But, I noticed that the authors' responses to my comments differ in format from those addressed to Reviewers 1 and 2. While their replies are structured as point-by-point responses with reviewer comments quoted in blue, mine are listed by number without the original comments included. This formatting made it difficult to track and cross-reference of my original concerns. For future revisions, I suggest maintaining a consistent structure that includes the reviewer's comments, as this facilitates the process for both reviewers and the editor.

Even though the manuscript improved significantly from the previous version, not all of the concerns I raised during the initial review have been fully addressed. While the revised methods section is overall clearer, several aspects of the FLEXPART implementation remain vague. I appreciate that the authors corrected and rewrote Equations (6) to (11), as the original formulations contained significant errors. However, it is still unclear whether these errors were confined to the manuscript or if they were also present in the model implementation. I wonder whether the discrepancies arose from a miscommunication between the code developer and the person who wrote the article. If that is the case, it raises the concern that the same misinterpretation may have affected the code implementation itself. I hope this is not the case. Nonetheless, since no new simulations have been provided in this revision, it remains uncertain whether the code implementation is consistent with the corrected equations.

A key unresolved concern in the methods section is with the description of the affine transformation applied to particle coordinates. In my initial review, I requested a clear mathematical description of this transformation. In the revised manuscript, the authors describe the process only qualitatively and refer readers to the Fortran source code in a public repository. This is not an adequate substitute for proper documentation in the manuscript or supplementary material. A clear and explicit mathematical description of the transformation is essential for transparency and reproducibility, particularly because this step directly affects the spatial accuracy of the satellite pixel representation.

Regarding the results and discussion, the authors state that they performed sensitivity tests but chose not to include them, as the paper's emphasis is on methodology. However, if the primary contribution of the manuscript is methodological, it becomes even more important to describe all technical steps, including the transformation, particle release setup, and super-observations procedure, rigorously. Providing this level of detail is critical to ensure that the approach can be reproduced and built upon by other researchers.

To be clear, my goal is not to obstruct publication. I recognize that the manuscript presents valuable developments and has strong potential. However, given that the stated focus is methodological, I believe the technical content should be presented with greater clarity and rigor to meet the standards of transparency and reproducibility expected in the field.

Further comments are below based on the response of the authors:

Comment 1:

I recommend adding a brief statement to the main text, perhaps in the Introduction, as there is no dedicated discussion section, to clarify how the developments presented in this study relate to FLEXPART v11. Specifically, while FLEXPART v11 introduces the option for custom particle initialization, it does not currently support simulation of total column averages from satellite retrievals. In contrast, the work presented here, based on FLEXPART v10.4, provides an operational method for calculating total column source-receptor relationships (SRRs) from satellite observations.

It would also be valuable to note that the developments described in this paper are planned for future integration into FLEXPART v11. Including this clarification will help readers and potential users select the appropriate FLEXPART version for their applications and will underscore the significance of the methodological contribution presented here.

Comment 3:

In my initial review, I raised concerns regarding the formulation of Equation (6), which originally appeared in the manuscript as:

$$H_{ijk}^{col} \sum_{n=1}^{N} a_n w_n \frac{t}{m_n \rho_{ijk}} \sum_{p=1}^{J_{i,jk,n}} m_p$$

At that time, I pointed out that the variable t should represent the residence time of particles within the surface layer as defined in Seibert and Frank (2004, Eq. 8) and Wu et al. (2018, Eq. 4), rather than a fixed sampling duration. I now see that the authors have updated Equation (6) as follows:

$$H_{in} = \frac{1}{P_n} \sum_{p=1} \frac{l_{pin} \Delta t_{pin}}{\rho_i}$$

While I appreciate the authors' revision of Equation (6) and subsequent equations and their explanation of the transmission function l_pin, I suggest improving the clarity of the description. The current phrasing : l_pin "represents the fraction of the mass remaining in the particle" could be misinterpreted by readers, particularly those less familiar with Lagrangian particle dispersion models operating in backward mode. In this context, particles are tracers and do not carry real mass. Rather, the transmission function serves as a scaling factor to account for atmospheric processes (e.g., chemical losses), modifying each particle's residence time contribution to the SRR.

Comment 5:

In my initial review, I suggested the authors to provide the explicit form of the affine transformation applied to particle coordinates (i.e., the mathematical equations used). However, the revised manuscript still describes the method only in qualitative terms, and the authors refer readers to the subroutine releaseparticles_satellite.f90 in the public Fortran code base to understand the implementation.

While I appreciate that the code is openly available, I would like to reiterate that it is neither feasible nor expected for a voluntary reviewer to examine and interpret a large Fortran code base to assess a methodological detail that could, and should, be documented clearly in the manuscript or its supplementary material. Given that the affine transformation plays a critical role in accurately representing satellite pixel geometry, I strongly recommend that the authors include the mathematical formulation of this transformation for transparency and reproducibility.

Comment 6:

It would significantly improve the manuscript if the authors provided the explicit equations used to perform the super-observation (super-orbiting) processing of TROPOMI data. Currently, this procedure is only described qualitatively, which limits the reproducibility of the method and may confuse readers unfamiliar with the approach.

Comment 8:

Indeed the reduced-chi-square criterion can be ambiguous and alone is not a sufficient criterion for assessing the appropriateness of the uncertainties. However, as stated by Chevallier et al 2007, in an idealized system (OSSE experiment), the cost function J(x) converges toward N (number of observations). Hopefully, the author made this test with TROPOMI before attempting to run real case inversion.

The reduced chi-square value of 4.86 for the ground-based observations suggests that the observation or model errors may be underestimated, or that there is unaccounted model–data mismatch. It would be helpful if the authors clarified how the observation uncertainty was defined for the in situ network. particularly whether representativeness and model transport errors were included, and at what magnitude.

I made this comment because, the authors mention that *'the satellite observations are more uncertain compared to the ground-based observations (the median uncertainty for XCH4 in our study was 16 ppb compared to 8 ppb for the ground-based observations) and the model-observation errors are weighted by the inverse square of the uncertainty'*. It could be possible that the uncertainties of ground based observations in combination with the model errors are larger that 8 ppb, hence the reduced chi-square value is super high (4.86).

Comment 9:

The authors note that they performed several sensitivity tests but chose not to include them in the manuscript, as the focus of the paper is "primarily on the methodology of using satellite observations in an inversion framework based on a Lagrangian transport model." However, if the manuscript is intended to be primarily methodological, then the methodological components should be described in greater detail.

Given the concise length and the technical nature of the work, specifically, the adaptation of FLEXPART v10.4 for use with satellite-based retrievals, the manuscript should contain the formal presentation of the algorithmic steps, including the mathematical formulation of transformations and other implementation choices, which are currently either omitted or described only qualitatively.