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Carlos Gómez-Ortiz
Department of Physical Geography and Ecosystem Science
Lund University
Sweden

Jens-Uwe Grooß
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Atmospheric Chemistry and Physics (ACP)

Dear Editor,

We appreciate the detailed feedback provided by the referee on our manuscript titled "Can $\Delta^{14}CO_2$ observations help atmospheric inversions constrain the fossil CO_2 emission budget of Europe?". While their comments have been instrumental in guiding our revisions, we would like to address two key points where we believe there may have been misunderstandings or misinterpretations of our revised manuscript:

1. The referee has repeatedly requested a mathematical description and justification for Appendix B. This appendix describes the procedure used to define the reduced grid, represented in Figure 2b. In our setup, the tracer transport is computed at a spatial resolution of 0.5°, but the emissions are resolved on a coarser grid towards the edges of the domain, where there are fewer observations to constrain the emissions. This is clearly represented in Figure 2 and described in Section 2.3.1 (lines 228 to 242 of the revised manuscript). While we welcome critical comments on this choice, it is important to note that it is a setting rather than a derivation. For practical reasons, it is defined through an iterative algorithm, but it is fundamentally an arbitrary choice, similar to the definition of the domain extent or the selection of a specific resolution for the transport model. As such, it is not feasible to provide a mathematical description for this more than the actual code referenced in the answer below.
2. The referee has criticized the lack of methodological description regarding the construction of the error covariance matrix. However, this information is clearly presented in Section 2.3.2 since the original version of the manuscript. Moreover, our inversion setup follows standard practices. While there may be aspects of this methodology that can be improved, our study's focus is not on these specific improvements. Therefore, we believe it is appropriate to refer to other studies for detailed methodologies on this topic.

Below, we provide a detailed response (in regular font) to each of the referee's comments *(in italics*), indicating how we have addressed them in the revised manuscript. We hope this clarifies any misunderstandings and demonstrates our commitment to meeting the high standards of the journal.

Referee's comments

The revised manuscript "Can $\Delta^{14}CO_2$ observations help atmospheric inversions constrain the fossil CO_2 emission budget of Europe?" by Carlos Gómez-Ortiz, Guillaume Monteil, Sourish Basu, *and Marko Scholze, provides a clear improvement of the original manuscript. This applies mainly for the part of the paper, which presents the results. The presentation of the methodology has made little improvements, however. The reviewers' guide on the clearness of the method emphasizes some essential questions about the methodological aspects. I cite from the reviewers' guide on the clearness of the method:*

- *Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?*
- *Are the scientific methods and assumptions valid and clearly outlined?*
- *Are the results sufficient to support the interpretations and conclusions?*
- *Is the description of experiments and calculations sufficiently complete and precise to allow for their reproduction by fellow scientists (traceability of results).*

The authors must explain the objectives of this paper more clearly. Simply stating that the aim is "to explore the interest of using these CO_2 and $\Delta^{14}CO_2$ observations to constrain the fossil CO_2 emissions in Europe" appears not to be a sufficiently strong motivation for performing this study and publishing it in ACP.

We believe our manuscript fits very well to the scope of ACP, in particular to the subject area 'Gases' and to the research activity 'Atmospheric Modelling and Data Analysis'. This submission is in line with previous articles submitted to and published by ACP such as: "Potential of 14C-based versus ΔCO-based $\Delta ffCO_2$ observations to estimate urban fossil fuel CO_2 ($ffCO_2$) emissions" (https://doi.org/10.5194/egusphere-2023-1239), "Atmospheric radiocarbon measurements to quantify CO_2 emissions in the UK from 2014 to 2015" (https://doi.org/10.5194/acp-19-14057-2019), and "Potential of European $^{14}CO_2$ observation network to estimate the fossil fuel CO_2 emissions via atmospheric inversions" (https://doi.org/10.5194/acp-18-4229-2018). Therefore, we consider ACP as suitable and relevant for publishing our manuscript.

To further clarify the objective and the relevance of our study, we have expanded this section of the Introduction as follows:

"In this work, we present the new capabilities of the Lund University Modular Inversion Algorithm (LUMIA) system (Monteil and Scholze, 2021) to perform simultaneous inversions of atmospheric CO_2 and $\Delta^{14}CO_2$ observations as a first attempt to develop a model capable of supporting the monitoring and verification of fossil CO_2 emissions across Europe. Such emissions monitoring and verification support capacities are essential for assessing compliance with international agreements, such as the Paris Agreement (UNFCCC, 2015), and for guiding policy decisions aimed at reducing carbon emissions as outlined by Janssen-Maenhout et al. (2020). We perform Observing System Simulation Experiments (OSSEs), recreating the current state of the ICOS network and its sampling strategy, and using different flux products (as priors and true values) to demonstrate the performance of the inversion scheme and show its capabilities. We begin by assessing the impact of oceanic fluxes on the total CO_2 and $\Delta^{14}CO_2$ concentrations. Then, we evaluate the impact of adding $\Delta^{14}CO_2$ observations on the estimation of fossil CO_2 emissions by comparing the model's ability to recover true fluxes starting from a prior flux set to zero. Finally, with a more realistic setup, i.e., prior, we evaluate the impact of the prescribed fossil CO_2 flux uncertainty and the impact of the terrestrial isotopic disequilibrium product."

The reader is entitled to a more detailed reference to the underlying LUMIA model in the model description subsection 2.1. There is no such reference here. For example, there is no outline of the Lagrangian approach embedded in the TM5 global model, let alone further reasonable specifications, suitable to acknowledge a vetted methodology.

There is no mention of a "Lagrangian approach embedded in the TM5 global model" in our manuscript. The only reference to the TM5 model appears in the discussion. We agree that the title of Section 2.1 could have been clearer and have slightly modified it. However, it is intentional that the transport model is not described in detail here, as the approach remains consistent whether the transport model is Lagrangian, Eulerian, global, or regional. The underlying transport model used in our study is FLEXPART, a widely used mesoscale Lagrangian transport model. This is described (and referenced) in Section 3.2, and a pointer to Section 3.2 is given in Section 2.1 (line 163). The specific implementation has been extensively described in Monteil & Scholze (2021), and it is standard practice to refer to the original model development paper rather than repeating the information.

To address the reviewer's concerns and improve clarity, we have modified the initial paragraph of Section 2.1 as follows:

"We are using the LUMIA (Lund University Modular Inversion Algorithm) system as described by Monteil and Scholze (2021), modifying the way background concentrations ($\boldsymbol{y}^{\mathrm{b}}$ in Equation 1) are calculated by computing a smoothed and detrended average of real observations from the ICOS network for each sampling site. Originally, the LUMIA system was developed to optimize regional Net Ecosystem Exchange (NEE) fluxes over Europe using in situ CO_2 observations from the ICOS (Integrated Carbon Observation System) Atmosphere network. In this study, we have extended LUMIA to additionally assimilate $\Delta^{14}CO_2$ observations from the same network and optimize multiple flux categories. This extension introduces a new step in the mass balance of the atmospheric transport, as follows:

$$\boldsymbol{y}_{\mathrm{CO_2}} = \boldsymbol{y}^{\mathrm{b}}_{\mathrm{CO_2}} + \sum_{\mathrm{c}} H(\boldsymbol{F}_{\mathrm{c}})$$

$$\boldsymbol{y}_{\mathrm{C\Delta^{14}C}} = \boldsymbol{y}^{\mathrm{b}}_{\mathrm{C\Delta^{14}C}} + \sum_{\mathrm{c}} H(\boldsymbol{\Delta}_{\mathrm{c}}\boldsymbol{F}_{\mathrm{c}})$$

"

This point was not addressed in my line 120 remark. Rather, only my "shifted delta" item was questioned, which simply means that the delta sign, typically located ahead of the flux F, has been displaced behind F (in the difference parentheses). In the writing of formulae, the authors do not differentiate between scalars, vectors, and operators by notation (normal vs. bold face, small vs, capital letters), which should be standard in mathematically oriented publications.

The radiocarbon signature $\boldsymbol{\Delta}$ is a vector, and its position on either side of the flux term does not affect the result of the multiplication. However, to ensure consistency in the formula, we have rewritten Equation 2b by moving the disequilibrium subtraction in front of $\boldsymbol{F}$ as follows:

$$\sum_{c} H(\boldsymbol{F}_{c}) = H(\boldsymbol{\Delta}_{\mathrm{ff}}\boldsymbol{F}_{\mathrm{ff}}) + H\big(\boldsymbol{\Delta}_{\mathrm{atm}}(\boldsymbol{F}_{\mathrm{bio}} + \boldsymbol{F}_{\mathrm{oce}})\big) + H\big((\boldsymbol{\Delta}_{\mathrm{bio}} - \boldsymbol{\Delta}_{\mathrm{atm}})\boldsymbol{F}_{\mathrm{bio2atm}}\big) + H\big((\boldsymbol{\Delta}_{\mathrm{oce}} - \boldsymbol{\Delta}_{\mathrm{atm}})\boldsymbol{F}_{\mathrm{oce2atm}}\big) + H(\boldsymbol{\Delta}_{\mathrm{nuc}}\boldsymbol{F}_{\mathrm{nuc}})$$

All the manuscript's equations were carefully revised and adjusted when needed using the appropriate notation following Table 1 from Fundamentals of data assimilation (Rayner et al., 2019).

Original L172 : In examining the space-time covariance matrix as understood by the authors, I consulted the Broquet et al. (2011) paper, as referenced in the reply. However, I could not discern any indication of the use of this spatio-temporal matrix formulation, as claimed. I would be interested in understanding the design of such a space-time matrix, which is used in data assimilation by the rarely applied Physical Space Statistical Analysis System approach (PSAS). Please provide precise information on the matrix entries Bij and the methodology used to obtain them. The repeated references to other studies, such as Basu et al., are insufficient and do not comply with the standards set by ACP and other quality journals (see above).

The construction of the matrix **B** is thoroughly detailed in Section 2.3.2, titled "Construction of the prior error covariance matrix (**B**)." We believe this section provides a clear, step-by-step explanation. Specifically, the entries of the prior error covariance matrix **B** are described by the equation found in Section 2.3.2 (L251 of the revised manuscript, originally L204).

Original L192…: The additional description, including matrix dimensions, has been noted. However, I was unable to reconcile the clustering approach detailed in Appendix B with equation (7).

We understand the referee's concern. However, it is important to clarify that Equation 7 and Appendix B address different aspects of the methodology (see also our general response to the review):

- Equation 7 describes the transition between the model grid and the optimization grid. This is a step within the inversion process.
- Appendix B explains how the optimization grid is defined, specifically how matrix $\mathbf{T}_h$ in Equation 7 is constructed. This is part of the experiment design and serves as an input to the inversion, rather than a step within it.

It is evident that Appx B is purely descriptive, devoid of any formulas or derivations that could potentially relate to (7). It appears that the authors have derived a sensitivity analysis through the use of adjoint (backward in time) integration, which has enabled them to identify clusters of emission cells.

We would like to clarify that we have not identified clusters of emissions. Instead, we have used a clustering approach to define the extent (in space) of the emission offsets optimized by the inversions. These "clusters" are based on the adjoint sensitivity of the network to the emissions (dE/dy), which is entirely independent of the emissions themselves. In other words, the grid is denser where there are more observations, as clearly illustrated in Figure 2. However, the specific approach used to define this reduced grid is not very relevant from the perspective of the inversion, therefore it is described in an appendix and not in the main text.

*In order to highlight the level of mathematical rigor that is evident in the authors' approach, it is worth noting the following literature, which features methods for achieving resembling objectives. Berliner, L. M., Lu, Z., and Snyder, C.: Statistical design for Adaptive Weather Observations, J. Atmos. Sci., 56, 2536–2552, 1998. ** Bishop, C. H. and Toth, Z.: Ensemble Transformation and Adaptive Observations, J. Atmos. Sci., 56, 1748–1765, 1998. ** Buizza, R. and Palmer, T. N.: The Singular-Vector Structure of the Atmospheric Global Circulation, J. Atmos. Sci., 52, 1434–1456, 1993. ** Goris, N. and Elbern, H.: Singular vector-based targeted observations of chemical constituents: description and first application of the EURAD-IM-SVA v1.0, Geosci. Model Dev., 8, 3929–3945, https://doi.org/10.5194/gmd-8-3929-2015, 2015.*

Thank you for highlighting these references. However, the objective in our case is more straightforward than the reviewer seems to think. Our goal is to construct a grid that has a denser resolution where observational coverage is better. This allows us to work with a smaller control vector and avoids the impracticality of solving for emission offsets at a very high resolution where there are no observations. The fine-scale structure of the emissions is maintained regardless.

Original L202: In response to the authors' initial request, I am unable to comprehend the rationale provided. A more rigorous mathematical approach would be beneficial. It is recommended that the authors provide a clear and detailed outline of the mathematical methods employed, in accordance with the criteria set out by the journal.

We do not understand the referee's comment here. The mathematical method is totally standard, and is entirely described in this section. Lines 202-208 describe how each element $\mathbf{B}_{ij}$ of the covariance matrix is constructed, in a generic way (i.e. with covariances based on the product of variances and distance-based correlation functions), while lines 209-217 describe how the elements of that product (the variances and correlations) are computed, for each category. All the equations that are used in the code are already present in the paper.

Original L282: It is unclear whether the authors have fully grasped the distinction between OSSEs and identical twin experiments. To illustrate, if meteorological simulations (wind transport) are conducted using the same model configuration for observational modelling and inversion procedures, the latter is provided. In such a case, the assimilation system may be considered to "err on the optimistic side", as outlined in the textbook by Daley (1991): Atmospheric Data Analysis, due to tacitly assuming the transport simulation as perfect. The authors are encouraged to subsume their approach in terms of this aspect.

Original L282: I don't know whether the authors have fully understood the difference between OSSEs and identical twin experiments. For example, if the meteo simulations (wind transport) are performed by the same model setup for observation modelling and the inversion procedure, then the latter is given, in which case the assimilation system "errs on the optimistic side" (see textbook Daley, 1991, Atmospheric data analysis).

We acknowledge that, in strict meteorological terminology, our approach aligns more closely with identical twin experiments rather than OSSEs. As the referee correctly point out, a 'real' OSSE would typically involve using a different transport model for generating pseudo-observations to avoid the assumption of perfect transport simulation. However, it is important to note that within the atmospheric inversion community, the term "OSSE" is commonly used to describe experiments like ours, even when the same model configuration is employed for both observational modeling and inversion procedures. This usage is prevalent in the literature and aligns with the established terminology in our field.

To illustrate this, we refer to studies where identical twin experiments are referred to as OSSEs, such as in Philip et al. (2019), Byrne et al. (2019), Liu et al. (2014), Chen et al. (2023), and Wang et al. (2014). These references demonstrate the conventional use of the term "OSSE" in the context of atmospheric inversions. Therefore, while we understand and acknowledge the meteorological distinction, we rather call it “perfect transport OSSEs" in our manuscript to remain consistent with the common terminology in atmospheric inversion studies.

Original L480: It would appear that the authors' response indicates a fundamental point of confusion. Firstly, it is unclear whether the Chi-squared value is set to approach ½ or 1. In practice, it is more likely to be set to ½, as exemplified by Talagrand. The successful approximation to the target value is contingent upon a consistent design of R and B, which may not be readily apparent even through educated guesses of these matrices. It is of the utmost importance to "tune" the covariances and validate the minimisation process in order to draw valid conclusions from a consistently designed OSSE. For further details, please refer to Desroziers et al. (2005). Furthermore, the remarks made in relation to original line 282 are equally applicable here.
I am unable to understand the explanation of the authors to my initial request. Substatially more mathematical rigour will help. The authors are strongly recommended to clearly outline the mathematical methods, following the journals’ criteria mentioned above.

Original L480: Apparently, the authors’ answer indicates a fundamental point of confusion: Firstly, is the Chi2 value set to ideally approaching at ½ or at 1 (mostly the former, e.g. Talagrand)? The successful approximation to the target value requires (only as necessary, not sufficient condition) a consistent design of R and B, not necessarily given even by educated guesses of these matrices. “Tuning” the covariances and validation of the minimization is essential to extract conclusions from a consistently designed OSSEs. See Desroziers et al. 2005 for details. In addition, the remarks to original line 282 apply here as well.

We calculate the reduced chi-square statistic as a diagnostic to ensure that we have improved upon the initial state, which is clearly supported by the change from the prior to posterior values (1.77 to 1.06), and as a way to guarantee that we are not under or overfitting the model’s optimization. We calculated the χ_ν^2 as:

$$\chi_\nu^2 = \frac{1}{\nu}\sum_{i=1}^{\mathrm{N}}\left(\frac{\boldsymbol{y}_i^{\mathrm{so}} - \boldsymbol{y}_i^{\mathrm{b,a}}}{\boldsymbol{\epsilon}_i}\right)^2$$

Where $\boldsymbol{y}_i^{\mathrm{so}}$ is the synthetic observation i, $\boldsymbol{y}_i^{\mathrm{b,a}}$ is either the prior (b) or the posterior (a) concentration i, $\boldsymbol{\epsilon}_i$ is the error of the synthetic observation i, N is the number of observations, and ν are the degrees of freedom calculated as $\nu = \mathrm{N} - \mathrm{p}$, being p the number of fitted parameters in the model. Since p is difficult to calculate due to the different time and space clusters, we keep the number of observations as the degrees of freedom $(\nu = \mathrm{N})$. We added this explanation to the manuscript to make it clearer.

We acknowledge the importance of a consistent design of the covariance matrices **R** and **B**, as we discussed in our methodology, however, we would like to emphasize that our current study represents a first demonstrator of the inversion scheme rather than an operational system. The level of fine-tuning expected for an operational system, as highlighted in the referee's comments, is indeed a goal for future work. At this stage, our primary aim is to demonstrate the feasibility and potential of our approach, which naturally involves some initial assumptions and approximations such as the perfect transport and boundary conditions and it does not make sense to fine tune on the current approach.

Appendix B:
The authors seem reluctant to respond positively to suggestions for a more comprehensive and rigorous presentation of the mathematical foundations. In particular, Appendix B is inadequate in its purely verbal presentation. I find an implementation hardly reproducible.

As stated above, there is no mathematical foundation for the clustering process. It is just a practical way to construct the "reduced grid" represented in Figure 2b. The code is provided for reproducibility (along with the entire code used in this project), the role of Appendix B is to explain to an interested reader how that code works. However, it is not a central part of the methodology (therefore it is in an Appendix). The clustering algorithm can be found in the code available as supplementary material (https://doi.org/10.5281/zenodo.8426217) on lumia/Tools/optimization_tools.py or in the GitHub repository (https://github.com/lumia-dev/lumia/blob/lumia_multitracer/lumia/Tools/optimization_tools.py).

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