REVIEW 1

We are very thankful to the reviewer for the positive feedback and the helpful suggestions to improve our manuscript. Our responses to the reviewer's comments are marked in blue.

Review of "Can radon help to improve methane emission estimates? Results from a dual-tracer inversion"

This manuscript aims to improve the reliability of CH_4 inversions through the inclusion of radon observations. The inversions are performed within the CarboScope framework, which is based on STILT footprints (at least in its standard configuration) and uses an iterative minimization of the cost function with a conjugate-gradient algorithm. The inversions are constrained by observations from 17 in-situ stations across Europe.

The authors use radon as an additional tracer to better characterize transport errors, particularly errors associated with vertical mixing within the planetary boundary layer. This is achieved by introducing correlations between CH_4 and Rn uncertainties in the observational uncertainty covariance matrix within a dual-tracer (CH_4–Rn) inversion framework. The model-data mismatches (MDMs) are estimated from prior model simulations and so-called "leave-one-out" runs.

The study further includes extensive sensitivity analyses, investigating different radon flux models, soil-moisture assumptions, and prior uncertainty settings for radon emissions. The authors discuss both the potential and the limitations of the proposed approach in a comprehensive and transparent manner.

The results indicate that including radon information in dual-tracer inversions generally leads to a smaller increase in CH_4 emissions over Central Europe compared to corresponding single-tracer CH_4 inversions.

Overall, this is an excellent study addressing one of the most important challenges in atmospheric inversions currently: transport uncertainty. Transport errors remain a major limitation for current greenhouse-gas inversions, and the presented work represents an interesting, feasible and scientifically sound contribution towards addressing this issue. The manuscript is comprehensive, methodologically rigorous, clearly written, and provides valuable insights. The authors build upon the well-established CarboScope framework and present a carefully designed experimental setup. I strongly recommend publication after a few minor issues have been clarified.

General comments

1. Lack of independent validation

One aspect that I find missing is an independent validation of the inversion results. As presented, it is difficult to assess whether the dual-tracer inversions actually provide more accurate CH_4 flux estimates than the single-tracer inversions. I fully acknowledge that obtaining an independent validation is challenging in practice. Nevertheless, would it be possible to compare prior and posterior MDMs at stations that are not assimilated in the inversion (at least during periods when the transport model is expected to perform reasonably well)? While such an analysis would still have limitations, it could provide at least some indication of whether the dual-tracer approach improves predictive skill.

We agree with the reviewer that an independent validation of the dual-tracer inversion is challenging, as neither the true Rn nor the true CH_4 fluxes are known. Nevertheless, we evaluate our dual-tracer inversion results at two independent sites in north-western Germany, Steinkimmen (STE) and Jülich (JUE), where both CH_4 and Rn observations are available. These sites were not included in the inversion due to substantial data gaps, particularly in the Rn measurements during 2021.

The prior CH_4-Rn MDM correlation coefficients at these sites are r=0.37 (for STE) and r=0.40 (for JUE), which fall within the range observed at the sites used in the inversion but are on the lower end of that range. This suggests that, also at these sites, a part of the CH_4 MDM variability can be attributed to common transport model deficits.

In the CH_4-only inversion, the CH_4-Rn MDM correlation coefficients are reduced, resulting in r=0.21 and r=0.39 at STE and JUE, respectively (see Fig. 1). This is accompanied by a substantial 67% reduction of the mean (prior) CH_4 MDM at both sites (see Fig. 2a). In contrast, the posterior fluxes from the dual-tracer inversion exhibit higher CH_4-Rn MDM correlation coefficients (r=0.31 for STE and r=0.45 for JUE), leading to a 3-7% higher mean posterior CH_4 MDM compared to the CH_4-only inversion. Under the extreme scenario of assuming a Rn prior flux uncertainty of 5%, the dual-tracer inversion produces even higher CH_4-Rn MDM correlation coefficients (r=0.35 and r=0.51), attempting to achieve the set correlation of r=0.6, which was assumed in the MDM error covariance matrix. As a result, the prior CH_4 MDM is less reduced, and the fluxes are less adjusted.

We also evaluated the performance of the different inversion approaches during situations of low prior Rn MDM (see Fig. 2b, for a prior Rn MDM below ±0.2 Bq m^{-3}). We observe a similar pattern: the dual-tracer yields a slightly higher mean posterior CH_4 MDM than the CH_4-only inversion.

Overall, when the transport model contains systematic biases, the dual-tracer inversion is expected to yield a slightly higher mean CH_4 MDM than the CH_4-only inversion, and thus worse predictive skills. This is because the dual-tracer approach accounts for the fact that part of the CH_4 MDM may arise from transport model errors rather than biases in the prior fluxes, thereby leading to less flux adjustments.

To further improve the dual-tracer inversion, a next step would be to implement site-specific CH_4-Rn MDM correlation coefficients. This would allow for a better description of the transport model performance at individual sites. In the current study, we assumed uniform transport model errors due to vertical mixing across all sites, which does not consider the spatial variability in the model errors.

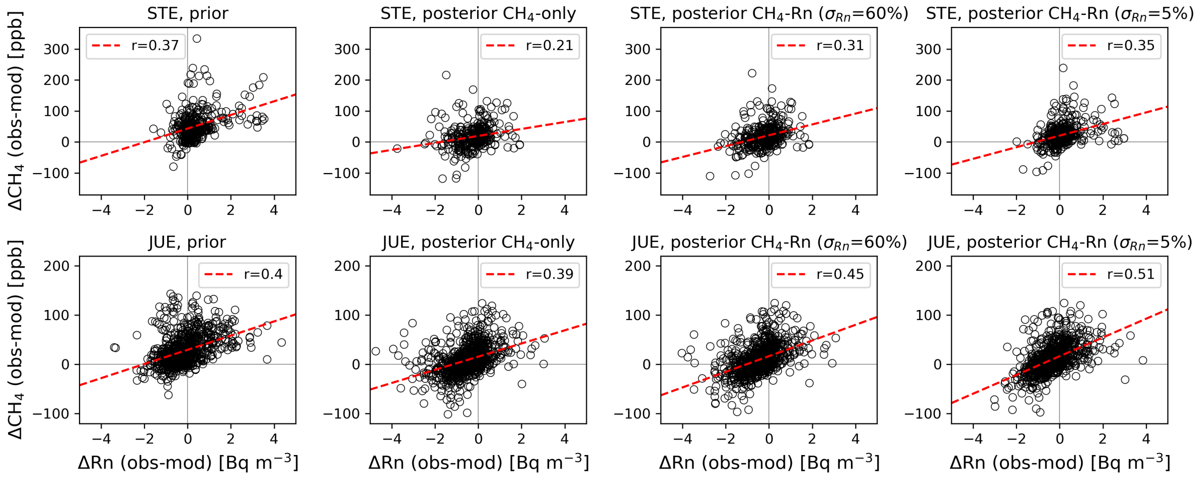

Figure 1: Correlation between CH_4 and Rn MDM at Steinkimmen (top panels) and Jülich (bottom panels) in 2021 for the prior fluxes, the posterior fluxes from the CH_4-only inversion, as well as the posterior fluxes from the dual-tracer inversion for a Rn prior flux uncertainty of 60% and 5%, respectively. The CH_4 and Rn observations for STE and JUE are taken from Fischer et al. (2024) and ICOS RI (2024), respectively.

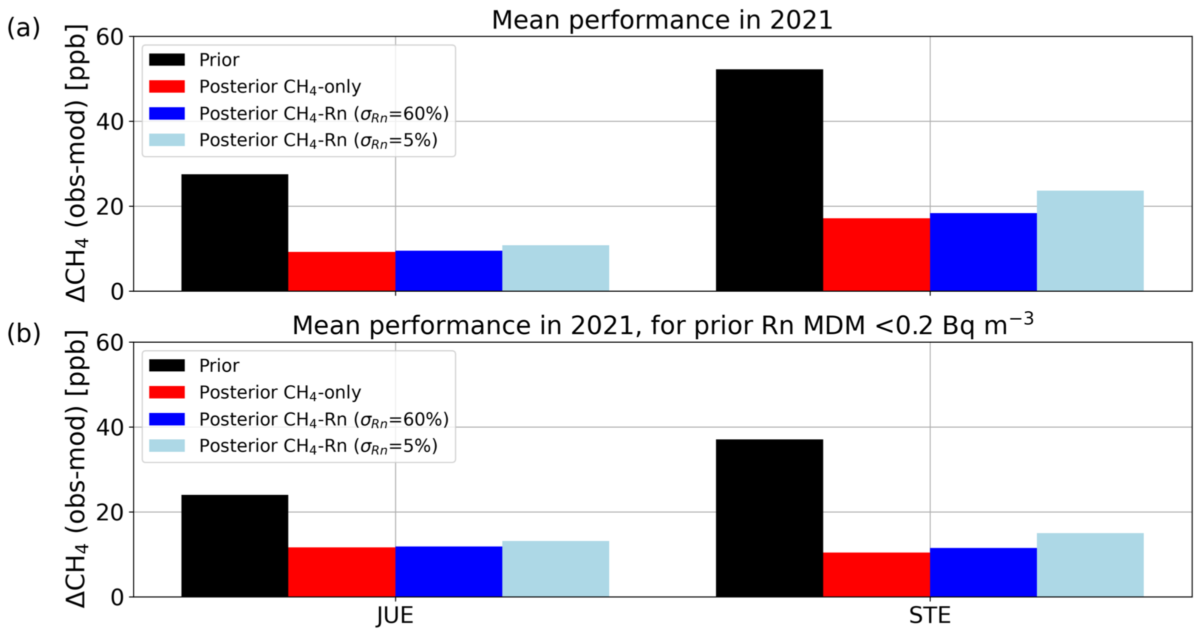

Figure 2: (a) Mean CH_4 MDM at Steinkimmen and Jülich in 2021 for the prior fluxes, the posterior fluxes from the CH_4-only inversion, as well as the posterior fluxes from the dual-tracer inversion for a Rn prior flux uncertainty of 60% and 5%, respectively. (b) Same as (a) but restricted to situations where the absolute prior Rn MDM is smaller than 0.2 Bq m^{-3}.

We have included this analysis in the appendix of the manuscript, and refer to it in the main text (l. 512-515; l. 731-762).

2. Adjustments in regions with limited observational coverage

I was surprised by the sometimes substantial flux adjustments occurring in regions with sparse observational constraints, particularly over the Iberian Peninsula and parts of the Balkans. Do the footprints of the assimilated stations provide sufficient sensitivity to these regions to support such adjustments? I see similar behaviour occasionally in other inversion systems as well, something I never fully understood. It would be helpful if the authors could discuss this point and perhaps provide the observational sensitivity of the footprints in these regions.

To illustrate the spatial sensitivity of the footprints within our model domain, we aggregated all afternoon footprints (or nighttime footprints for the two mountain sites, SSL and JFJ) from all 17 sites in 2021. The footprint sensitivity is highest in central Europe (see Fig. 3) and declines strongly with distance outside this region (note the logarithmic scale in the plot). This implies that fluxes in more distant regions have considerable flexibility to be adjusted within the range of the prior flux uncertainty, potentially allowing minor reductions in the inversion cost function. However, such adjustments are poorly constrained by the observations and therefore lack robustness and reliability. Consequently, we restricted our analysis to regions with a dense observational coverage, i.e., western and central Europe.

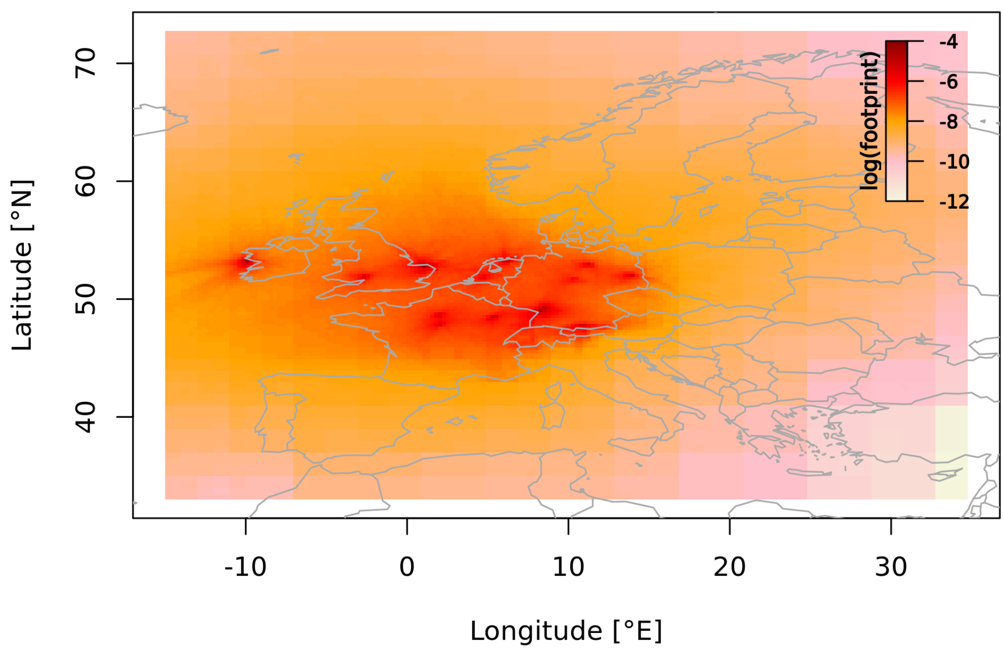

Figure 3: Average footprint of the 17 sites used in the inversion for 2021. The average footprint is shown on a logarithmic scale and is expressed in units of ppm/(μmol m^{-2}s^{-1}).

We have included this figure showing the footprint sensitivity and refer to it in the main text (Fig. S9 in the supplement; l. 417-418; l. 440).

3. Clarification of the leave-one-out experiments

It is not entirely clear to me whether the leave-one-out experiments are performed separately for each inversion setup or only once and subsequently reused across all inversion experiments. Based on the discussion around line 486 and following, I infer that they are repeated for each setup. Otherwise, it is difficult to understand how changes in the assumed prior uncertainty of radon emissions could lead to larger Rn MDMs that subsequently propagate to CH_4 through the cross-correlated uncertainties. I suggest clarifying this explicitly in the manuscript.

We performed the leave-one-out (LOO) runs only once, separately for the CH_4-only and the Rn-only inversions, to estimate the uncertainty of the CH_4 and Rn MDM at each site. For these LOO runs, we employed the standard prior uncertainty settings specified in Tab. 2 of the manuscript: 30% for each CH_4 sector, and 30% for Rn (with respect to the annual total flux of the European model domain). The resulting MDM uncertainty estimates were then used for all dual-tracer inversion runs, independent of the assumed Rn prior flux uncertainty.

By using the same uncertainty for the Rn MDM in each inversion run, the extent to which Rn information is effectively exploited in the dual-tracer inversion to improve CH_4 fluxes is directly determined by the Rn prior flux uncertainty: a lower Rn prior flux uncertainty allows less flexibility to adjust the Rn fluxes and to reduce the Rn MDM, which is propagated to CH_4 through the CH_4-Rn MDM correlation, while a higher Rn prior flux uncertainty has the opposite effect.

We have clarified this in the revised manuscript (l. 267-269) and tried to improve the discussion around line 486 (now l. 499-514 in the revised manuscript):

"[…] The CH_4-Rn MDM correlation contains this information on the transport model performance and forces the posterior CH_4 MDM to correlate with the Rn MDM. As a result, the adjustment of the CH_4 fluxes becomes effectively linked to the model's ability to simulate Rn. The Rn prior flux uncertainty plays a key role in determining the flexibility of the dual-tracer inversion to adjust the Rn fluxes, and thus, the Rn MDM. For instance, during periods when the model's representation of vertical mixing is poor, as indicated by large Rn MDMs, the inversion has limited capacity to reduce the Rn MDM, provided the Rn prior flux uncertainty is set small enough. Due to the CH_4-Rn MDM correlation, a large CH_4 MDM is also expected during such periods when the Rn MDM is large, leading the inversion to place stronger weight on the CH_4 prior and less on the CH_4 data, thereby limiting the ability to adjust the CH_4 fluxes. Conversely, when the transport model performs well, indicated by small Rn MDMs, the inversion attempts to also realize a small CH_4 MDM by adjusting the CH_4 fluxes accordingly. Therefore, the effect of the CH_4-Rn MDM correlation can be interpreted as a time-dependent weighting of the CH_4 (and Rn) data constraint relative to the prior constraint. And the magnitude of the Rn prior flux uncertainty determines whether the Rn information is mainly used to adjust the Rn fluxes (in the case of a high Rn prior flux uncertainty), or to improve the CH_4 flux estimates by diagnosing transport model performance (in the case of a lower Rn prior flux uncertainty). Overall, due to the positive biases in the annual mean (prior) CH_4 and Rn MDMs at most continental sites, the dual-tracer inversion leads to less reduction in the CH_4 MDM on an annual average compared to the CH_4-only inversion (see Fig. S4). "

4. Definition of the observational covariance matrix in the dual-tracer inversion

Another point that is not entirely clear to me concerns the treatment of the diagonal elements of **Qm**. Do the diagonal variances remain identical in the dual-tracer inversion and the corresponding single-tracer inversion? If so, introducing the off-diagonal covariance terms effectively increases the total observational uncertainty represented by **Qm**, which would generally tend to reduce the strength of the inversion adjustments. Could part of the differences observed, for example for Germany in Fig. 6, be explained by this effect rather than by the transport-error information carried by radon itself?

Yes, we assume the same diagonal elements in the Q_m matrices for both the dual-tracer and single-tracer inversions. We have clarified this in the manuscript (l. 275-276). The CH_4-Rn MDM correlation described in the off-diagonal elements of the MDM error covariance matrix provides additional information on the transport model performance. This correlation ensures that the dual-tracer inversion adjusts the prior CH_4 and Rn fluxes such that the posterior CH_4 and Rn MDMs are also correlated.

As we described in our reply above, the effect of the CH_4-Rn MDM correlation can be interpreted as a time-dependent weighting of the CH_4 (and Rn) data constraint relative to the prior constraint. The fact that the transport model systematically underestimates both CH_4 and Rn observations at many continental sites in 2021 suggests that a part of this underestimation stems from common transport model deficits. Consequently, the dual-tracer inversion yields a smaller CH_4 increase than the CH_4-only inversion in Germany. This indicates that, on an annual average, the effective CH_4 prior constraint is stronger in the dual-tracer inversion than in the CH_4-only inversion, as the dual-tracer inversion leverages Rn data to detect and account for transport model deficits. The added information from Rn can be seen in the increased uncertainty reduction of the CH_4 fluxes in the dual-tracer inversion (cf. Fig. 8 of the manuscript).

Specific comments

Figure S2

It is somewhat surprising to me that several stations exhibit a substantial number of observations with strongly negative MDMs at very low mixing heights. Based on the common tendency of models to overestimate vertical mixing, I would have expected predominantly positive MDMs under such conditions.

This behaviour is particularly noticeable at WAO and MHD. For these coastal stations, could wind-direction errors also play an important role in addition to vertical mixing errors? For example, under low-wind conditions, especially during nighttime, the model may incorrectly advect emissions from the continent towards the station, whereas the station may in reality sample relatively clean marine background air.

Furthermore, what effect has the use of the modelled mixing height rather than observed mixing heights in this analysis? How well does the modelled mixing height compare to available observations?

Yes, since both the natural CH_4 and Rn sources in the ocean are minor to negligible, respectively, a strong negative MDM at a coastal site could be attributed not only to a bias in the simulated vertical mixing but also to inaccuracies in the modelled wind direction. Specifically, if the model's back-trajectories suggest contributions of CH_4 and Rn from land areas, while the measurements indicate clean air conditions from the ocean.

To investigate this further, we plotted the CH_4 and Rn MDM at the two coastal sites, MHD and WAO, as a function of the measured wind direction (see Fig. 4). At both sites, strong negative MDM values, below -50ppb for CH_4 and below -2 Bq m^{-3} for Rn, occur across most wind sectors. Indeed, several of the most extreme MDM outliers coincide with large discrepancies between the measured and modelled wind directions. However, there are also notable outliers where the measured and simulated wind directions agree closely, suggesting that a bias in the simulated vertical mixing may be the main cause of the MDM in these cases.

A bias in the vertical mixing simulated by STILT could stem from inaccuracies in the modelled mixing heights or from an inadequate representation of convective processes. We fully agree with the reviewer that the mixing heights estimated by STILT should be evaluated in detail. Radiosonde or ceilometer measurements provide valuable data on the mixing height and can serve as a benchmark for evaluating the model performance. However, a reliable model-data comparison requires a careful preparation of these vertical profile data, including temporal and spatial alignment. Such an analysis falls beyond the scope of the present study but will be the focus of future work.

We have included in the manuscript that part of the CH_4 and Rn MDM at the coastal sites could also be explained by errors in the simulated wind direction (l. 338-341).

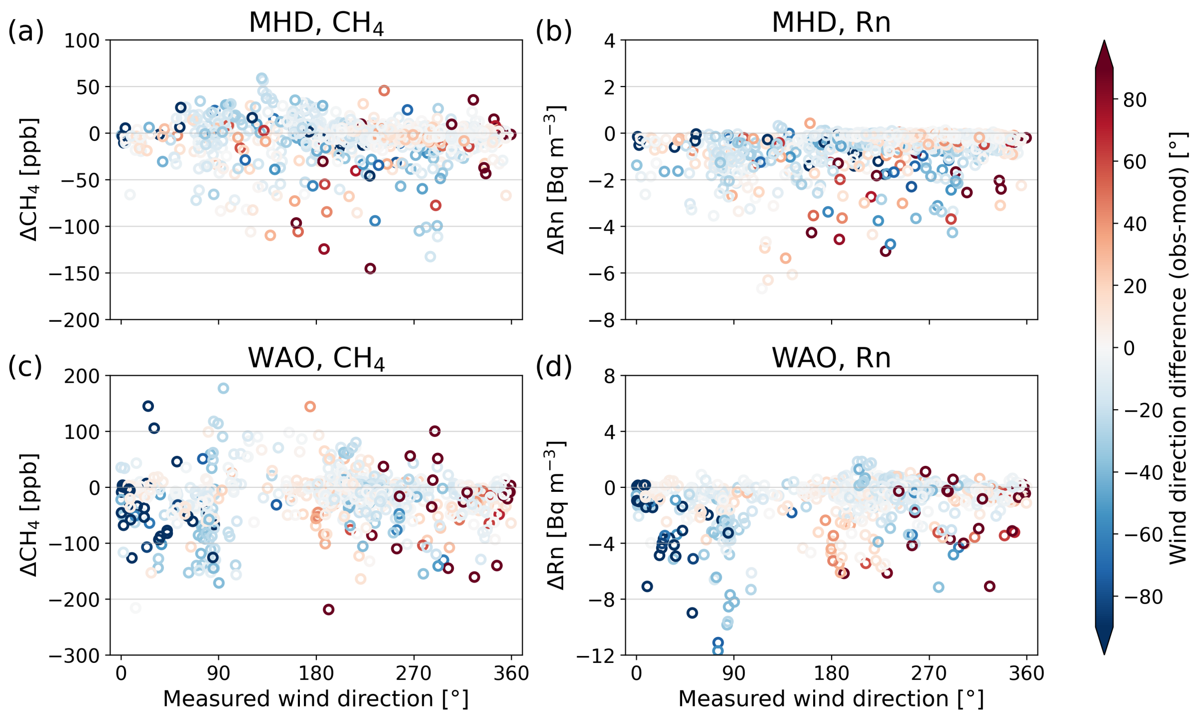

Figure 4: CH_4 (a, c) and Rn (b, d) MDMs versus measured wind direction at the two coastal sites MHD (a, b) and WAO (c, d). The marker color indicates the difference between measured and simulated wind direction. The meteorological data for MHD and WAO are taken from Lopez et al. (2026) and Forster and Manning (2026), respectively.

Line 336

In addition, during nighttime there are generally no thermal upwinds along the slopes that would transport emissions from local sources towards mountain stations.

Thank you, we have included this information in the manuscript (l. 344-345).

References:

Fischer, B., Kneuer, T., Kubistin, D., Lindauer, M., Müller-Williams, J., Plaß-Dülmer, C., Schmidt, M., and Meinhardt, F.: HRM Radon Data Germany L2 final data, ICOS ERIC – Carbon Portal [data set], https://doi.org/10.18160/Q2M8-B1HJ, 2024.

Forster, G., Manning, A.: ICOS ATC Meteo Release from Weybourne (10.0 m), 2021-07-01–2026-03-31, ICOS RI, https://hdl.handle.net/11676/8GjmIkXwFgBjKereGmZzVi91, 2026.

ICOS RI, Bergamaschi, P., Colomb, A., De Mazière, M., Emmenegger, L., Kubistin, D., Lehner, I., Lehtinen, K., Leuenberger, M., Lund Myhre, C., Marek, M.V., O'Doherty, S., Platt, S.M., Plaß-Dülmer, C., Apadula, F., Arnold, S., Blanc, P.-E., Brunner, D., Chen, H., Chmura, L., Chmura, Ł., Conil, S., Couret, C., Cristofanelli, P., Forster, G., Frumau, A., Gerbig, C., Gheusi, F., Hammer, S., Haszpra, L., Hatakka, J., Heliasz, M., Henne, S., Hensen, A., Hoheisel, A., Kneuer, T., Larmanou, E., Laurila, T., Leskinen, A., Levin, I., Lindauer, M., Lopez, M., Mammarella, I., Manca, G., Manning, A., Martin, D., Meinhardt, F., Mölder, M., Müller-Williams, J., Noe, S.M., Nęcki, J., Ottosson-Löfvenius, M., Philippon, C., Pitt, J., Ramonet, M., Rivas-Soriano, P., Scheeren, B., Schumacher, M., Sha, M.K., Spain, G., Steinbacher, M., Sørensen, L.L., Vermeulen, A., Vítková, G., Xueref-Remy, I., di Sarra, A., Conen, F., Kazan, V., Roulet, Y.-A., Biermann, T., Delmotte, M., Heltai, D., Hermansen, O., Komínková, K., Laurent, O., Levula, J., Lunder, C., Marklund, P., Morguí, J.-A., Pichon, J.-M., Schmidt, M., Sferlazzo, D., Smith, P., Stanley, K., Trisolino, P., Zazzeri, G., ICOS Carbon Portal, ICOS Atmosphere Thematic Centre, ICOS Flask And Calibration Laboratory, ICOS Central Radiocarbon Laboratory: European Obspack compilation of atmospheric methane data from ICOS and non-ICOS European stations for the period 1984-2024; obspack_ch4_466_GVeu_v10.0_20240729 [data set], https://doi.org/10.18160/PAWA-B2Y4, 2024.

Lopez, M., Martin, D., Ramonet, M.: ICOS ATC Meteo Release from Mace Head (24.0 m), 2018-03-01–2023-03-15, ICOS RI, https://hdl.handle.net/11676/JsE16hjsbqU18CIVIE23qL_C, 2026.

REVIEW 2

We thank the anonymous reviewer for reviewing our manuscript and for the constructive questions, which helped us clarify the information content of Rn in the dual-tracer inversion. Our responses to the reviewer's comments are marked in blue.

Maier et al perform a dual tracer CH_4 and ^{222}Rn inversion, utilizing transport-induced correlations in the modeled time series of CH_4 and ^{222}Rn to improve CH_4 flux estimates. The possibility of using ^{222}Rn to trace the boundary layer dynamics in transport models and thereby quantify transport errors in a GHG inversion has long been considered a possibility, and the current effort follows earlier work from several coauthors on developing the components needed (Gachkivskyi et al., 2025; Karstens et al., 2015; Levin et al., 2021; Maier et al., 2025). I am heartened to see the progress and this eventual implementation of a dual tracer CH_4+^{222}Rn inversion framework to address transport biases, a long-standing limitation of GHG inversions using boundary layer data.

The work itself is very thorough, and I have little to recommend by the way of improvements. The methods are clearly described, including the implementation of new features such as the off-diagonal Q_m matrix, and the authors have done an extensive set of sensitivity tests to quantify the different sources of error. This is an excellent study that should be published. The only clarification I would like is about their metric of judging whether a dual-tracer inversion is better than a single tracer, CH_4-only inversion. There are two issues, as follows.

Information flow due to off-diagonal elements in Q_m

The authors motivate their work by citing a seminal CO+CO_2 dual tracer inversion by Palmer et al (2006), in which both transport-induced correlations in Q_m and source-induced correlations in Q_p were implemented. As far as I understand that paper, Palmer et al (2006) did not see much improvements to CO fluxes in the dual tracer inversion (compared to the single tracer inversion). For CO_2 fluxes, the primary metric for judging that the dual tracer inversion was better was that it provided for a separation of NEE and biomass burning, as judged by their posterior correlation. They suggested that this was primarily due to CO:CO_2 transport correlations, since source correlations were too weak. This makes sense intuitively, since measurements of biomass burning CO plumes and enhancements could now provide information on how large the biomass burning CO_2 signal *should be* at those measurement locations. This would then be traced back to biomass burning CO_2 emissions, effectively separating it from NEE.

However, in their own work, the authors use the uncertainty reduction of CH_4 emissions as a metric of the added value of ^{222}Rn measurements (lines 547—549, and again line 586). I do not understand why adding CH_4:^{222}Rn correlations in

Q_m would result in a stronger uncertainty reduction for CH_4 posterior emissions. In my mind, adding positive off-diagonal elements without changing the diagonals of Q_m should reduce the information content in the measurements. Take the extreme example of a correlation of +1 between two y-Hx. This means that two measurements could be collapsed into one without any loss of information, or reduction in the uncertainty of that average measurement. Then why does adding positive but <1 correlations in $Q_m$result in a stronger uncertainty reduction in methane emissions? I'm sure it does in the authors' setup (as they report), but I'm confused as to why. Some explanation of the flow of information here would be helpful.

We thank the reviewer for the question regarding the flow of Rn information in the dual-tracer inversion. To illustrate this, let's first assume that the CH_4-Rn MDM correlation coefficient (r_{MDM}) is zero. In this scenario, no information from Rn is used to constrain the CH_4 fluxes (nor is CH_4 information used to constrain the Rn fluxes). Thus, the dual-tracer inversion reduces to two separate inversions. As shown in Fig. 8d of the manuscript, the uncertainty reduction for the CH_4 fluxes under this scenario (r_{MDM}=0) is not greater than that achieved in a single-tracer CH_4-only inversion. So, when r_{MDM}=0, the inclusion of Rn provides no additional benefit for estimating CH_4 fluxes.

Now consider the case where r_{MDM}>0, indicating a co-occurrence of large CH_4 and Rn MDMs. If the CH_4 and Rn sources were very similar and their prior flux errors were perfectly correlated, a r_{MDM}>0 would not provide much information on transport model performance. In such a case, a large MDM in both tracers would simply reflect a common bias in both prior fluxes.

However, in reality, CH_4 and Rn sources are very different, particularly in central Europe, which is dominated by anthropogenic sources (as discussed in Sect. 3.1.2 of the manuscript). Therefore, a r_{MDM}>0 primarily reflects shared transport model errors rather than common biases in the prior fluxes. Periods with a large Rn MDM indicate that the transport model is worse, and the r_{MDM}, which is described in the MDM error covariance matrix of the dual-tracer inversion, propagates this information to the CH_4 inversion. As a result, the CH_4 MDM is less reduced during such periods, and the CH_4 fluxes are less adjusted compared to a CH_4-only inversion, which lacks this information about transport model reliability.

The effectiveness of the dual-tracer inversion heavily depends on the Rn prior flux uncertainty. A very high Rn prior flux uncertainty gives the inversion high flexibility to adjust the Rn fluxes and to reduce the Rn MDM. Potential biases in the transport model are compensated by erroneous flux adjustments. However, when the Rn prior uncertainty is low, the inversion has less flexibility to reduce large Rn MDMs during periods of poor model performance. Due to the enforced r_{MDM}, this implies that large CH_4 MDMs are also expected under these conditions, leading to reduced adjustments in the CH_4 fluxes, even if the CH_4 prior uncertainty would allow for more flexibility.

The effectively used Rn information is reflected in the increased uncertainty reduction of the CH_4 fluxes for smaller Rn prior flux uncertainties (cf Fig. 8d of the manuscript). Overall, the magnitude of the Rn prior uncertainty determines whether the Rn information is mainly used to adjust the Rn fluxes (in the case of a high Rn prior flux uncertainty), or to improve the CH_4 flux estimates by diagnosing transport model performance (in the case of a lower Rn prior flux uncertainty).

Finally, given that the uncertainty reductions in our single-tracer inversions are already quite high, the primary contribution of Rn appears to lie not in further reducing the posterior CH_4 flux uncertainty, but in improving the absolute magnitude of the CH_4 flux estimates by correcting for biases induced by the transport model deficits.

We have revised the manuscript to better explain the flow of Rn information in the dual-tracer inversion (l. 499-512).

Reduction of transport-induced bias

The authors motivate their work by citing the impact of transport biases on inversely estimated fluxes, especially the bias in venting the PBL into the free troposphere. However, as they themselves acknowledge, adding ^{222}Rn as a second tracer does not “fix” the transport biases. Following their logic, it’s not clear to me that ^{222}Rn observations would in any way reduce the transport-induced bias in posterior fluxes. If anything, by sampling a biased transport model more, the additional observations would solidify the bias in the flux estimates. If that’s the case, then what is the added benefit of adding a transport tracer such as ^{222}Rn to the inverse model? Palmer et al (2006) benefited from adding CO observations and CO:CO_2 correlations because there were more CO observations than CO_2 observations, and the added correlation could provide information about CO_2 fluxes from CO enhancements. However, that is not the case here, because ^{222}Rn is more difficult to measure than CH_4, so there are not more ^{222}Rn measurements than CH_4. How do the additional ^{222}Rn measurements then mitigate the transport bias effect on CH_4 estimates, that the authors state as a motivating factor? In any case, the metric for checking such a benefit would not be uncertainty reduction, but rather distance from true emissions in an OSSE.

We agree with the reviewer that the usage of Rn in the dual-tracer inversion does not “fix” the transport biases, i.e., it does not directly correct the transport model. However, we disagree with the statement that additional Rn observations would solidify transport-induced biases in the posterior flux estimates. On the contrary, the inclusion of Rn observations helps mitigate these biases by reducing the effective influence of CH_4 observations during periods when the transport model performance is poor, as diagnosed by the Rn MDM. We have tried to explain this benefit from the additional Rn observations, and how they mitigate the transport bias effect on the CH_4 flux estimates in our response to the reviewer's first comment above and in the manuscript (l. 499-512).

We acknowledge that it is challenging to demonstrate the overall benefit of the dual-tracer inversion because the true CH_4 emissions are not known in reality. However, the substantial CH_4-Rn MDM correlations observed at multiple sites across central Europe, combined with the systematic underestimation of both the CH_4 and Rn concentrations at the continental sites, suggests a systematic overestimation of vertical mixing in the STILT model in this region. Accordingly, the dual-tracer inversion results are meaningful; the incorporation of the Rn information leads to a downward correction of the CH_4 flux estimates derived from the CH_4-only inversion. Thus, the Rn information in the dual-tracer inversion is used to reduce the impact of transport model biases on the CH_4 flux estimates, which are not accounted for in the CH_4-only inversion.

We agree that an OSSE can serve as a valuable tool to evaluate the implementation of the dual-tracer inversion, however, it provides limited evidence for real-world applications. The outcome of such an OSSE is highly sensitive to its design, particularly the assumed transport model errors. To ensure the reliability of our method, we have carefully validated the implementation of the dual-tracer inversion through multiple sensitivity tests. Furthermore, in response to a suggestion from the other reviewer, we now present an evaluation of the dual-racer inversion's performance at two independent sites in north-western Germany, which are not used in the inversion (see Appendix B in the manuscript, l. 731-762). The goal of our study is to directly demonstrate the application of the dual-tracer inversion in real-world conditions, to realistically assessing the benefits and limitations of the CH_4-Rn inversion.

References

Gachkivskyi, M., Karstens, U., Fischer, B., Kubistin, D., Müller-Williams, J., Lindauer, M., and Levin, I.: Radon-222 monitoring at German ICOS atmosphere stations, Earth Syst. Sci. Data, 17, 6173–6197, https://doi.org/10.5194/essd-17-6173-2025, 2025.

Karstens, U., Schwingshackl, C., Schmithüsen, D., and Levin, I.: A process-based ^{222}radon flux map for Europe and its comparison to long-term observations, Atmospheric Chem. Phys., 15, 12845–12865, https://doi.org/10.5194/acp-15-12845-2015, 2015.

Levin, I., Karstens, U., Hammer, S., DellaColetta, J., Maier, F., and Gachkivskyi, M.: Limitations of the radon tracer method (RTM) to estimate regional greenhouse gas (GHG) emissions – a case study for methane in Heidelberg, Atmospheric Chem. Phys., 21, 17907–17926, https://doi.org/10.5194/acp-21-17907-2021, 2021.

Maier, F., Falge, E., Gachkivskyi, M., Henne, S., Karstens, U., Kikaj, D., Levin, I., Manning, A., Rödenbeck, C., and Gerbig, C.: How reliable are process-based ^{222}radon emission maps? Results from an atmospheric ^{222}radon inversion in Europe, Atmospheric Chem. Phys., 25, 12779–12809, https://doi.org/10.5194/acp-25-12779-2025, 2025.

Palmer, P. I., Suntharalingam, P., Jones, D. B. A., Jacob, D. J., Streets, D. G., Fu, Q., Vay, S. A., and Sachse, G. W.: Using CO_2:CO correlations to improve inverse analyses of carbon fluxes, J. Geophys. Res. Atmospheres, 111, n/a--n/a, https://doi.org/10.1029/2005JD006697, 2006.