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Comment on acp-2022-118

Anonymous Referee #1

Referee comment on "High-resolution inverse modelling of European CH_4 emissions using the novel FLEXPART-COSMO TM5 4DVAR inverse modelling system" by Peter Bergamaschi et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-118-RC1, 2022

Bergamaschi et al. present a novel inverse modeling framework for estimating European methane emissions. The analysis includes a detailed investigation of the sensitivities and uncertainties in the inverse system, and in general the estimated emissions are shown to be robust. The authors have gone through the additional, considerable effort of performing some of the inversions in different inverse systems, to highlight the advantage of their new approach. The analysis is comprehensive, and addresses all important aspects that the presentation of a new system should address. The system itself is a valuable addition for estimating methane emissions in a time where, as the authors point out, methane has become an important mitigation target.

My most important comment is that while the technical aspects of the analysis are strongly highlighted and generally well-explained, I miss reflection on the context of the inverse system especially in later parts of the manuscript (discussion or conclusion). Therefore, I think it is important to address some additional relevant questions in the manuscript, such as:

- How do the authors plan to use this system in the future? Can the system easily be used to estimate methane emissions over a longer timescale, instead of just 2018? If so, how far back can you go (e.g., availability of observations)? How quickly can it be applied to recent years?
- Will this system be incorporated in existing “emission verification” efforts?
- The authors have focused their uncertainty analysis on one year. If the framework would be extended to multiple years, it is likely that part of the differences between the inverse set-ups will be constant between years (i.e., systematic), and will therefore not impact a trend analysis. If the authors agree, it would be good to add some discussion on this for context.
- In general, the inverse emission estimates suggest some differences between top-down and bottom-up, but uncertainties overlap. Given the wide range of sensitivity tests, can the authors provide more insights into the way forward towards reducing the top-down uncertainties such that these differences can be better understood? To what degree can more modeling efforts help, and to what degree do we need a denser observational network? Related to the previous point, will a trend be better constrained than an

absolute emission estimate?

- Even if total emissions were much more strongly constrained, then still the significant uncertainty in natural emissions will sustain a large posterior uncertainty. Do the authors see any way to address this challenge from a top-down perspective?

Other comments:

- I suggest splitting up the lengthy L60-L108 paragraph.
- From Table 3 I understand that the standard spatial correlation is 100km in the FLEXVAR inversions. However, in the FLEXKf and TM5-4DVAR inversions 200km is used (L300 & L319). Why this difference? Could this partly explain why the FLEXVAR inversions reproduce the observations best (i.e., a less stiff state)?
- L368-374: If I understand correctly, only in-situ observations in the optimal three-hourly window are selected, then these are averaged to one daily value per site. I understand this choice, but do the authors consider that any valuable information is lost in averaging out the high-frequency signal, or in the data that are not in this three-hour window? Additionally, how is the data from discrete air sampling treated, since there is not the same choice for selecting a time window?
- The authors point out that an advantage of the 4DVAR approach is that, for optimization, the emission grid does not need to be aggregated. However, there is still limited information in the CH_4 observations, so that correlation lengths in space and time need to be applied in the optimization. It seems useful to add the effective degrees of freedom that these correlation lengths result in, in addition to the total number of state elements, to compare to the other inverse systems (e.g., near L175).
- The authors calculate posterior uncertainties in inversions different from the reference inversions. Most importantly, the alternative inversions allow for negative emissions. For this to be a valid strategy, the alternative inversions should converge to similar emissions and reproduce observations similarly as the standard inversions. I could not find any confirmation that this is the case. I understand that qualitatively this approach makes sense and the results seem plausible, but I would like to see some evaluation of this aspect in the manuscript (or supplements).
- Fig. 5 contains a lot of information, and takes a while to fully take in. Having everything differentiated with only color does not help this process. Perhaps it could help to use different markers/linestyles for inversions that start from different priors (e.g., circle for E1, square for E2, triangle for E3)? Then, statements as in L562-L564 can be more easily seen. Other ways to adjust the colors, or reduce the number of colors, would be desirable: the light-yellow (INV-E1-O1-S3.1) is hardly visible and some of the greens are indistinguishable.
- I would be interested to see a rough comparison of the computational cost of the different inverse systems. In principle a global TM5-4DVAR inversion is needed for the baseline of the FLEXPART inversions, but this global inversion (as I understand it) only needs to be done once. Once the baseline is determined, are the FLEXPART inversions much faster than the TM5-4DVAR inversions? I expect this to be an important additional advantage of the Lagrangian approach.