Reply to comments by referee 2

Referee comments in black.

Replies in blue.

Suggested changes to text in italic green.

Katharopoulos et al presented a study that estimates posterior emissions of f-gases over Switzerland. In this study, the meteorological fields were driven using COSMO model with 7 (C7) and 1 km (C1), with an improved turbulent scheme for C1. Source-receptor relationships were obtained using FLEXPART model. Prior emissions were spatially and temporally distributed considering different assumptions, which resulted in a set of scenarios. In general, the results are consistent and in agreement with the literature. Authors claim that the uncertainty on baseline concentrations and spatial distribution of prior emissions have more impact on posterior estimates. However, the authors must explain and clarify some issues first. Also, there are some minor issues to be solved.

We would like to thank the referee for the detailed comments and overall favorable evaluation of our manuscript. We have addressed all questions in the following and modified the manuscript accordingly.

Line 55: explain why is harder with Eulerian models

We added the following text explaining why inverse modelling with Eulerian models may require additional efforts.

"In contrast, deriving a source-receptor relationship from an Eulerian model either requires an adjoint version of the model, a finite-differences approach including multiple perturbed forward simulations or employing ensemble methods (e.g., Brasseur and Jacob, 2017), all of which come at a higher computational cost especially in situations with small amounts of observational data."

Line 65: It is general knowledge of the complex terrain of Switzerland, however, it would be appreciated if the authors can provide some numbers.

We modified the section ending with the citation of Schmidli et al. (2018) and added more information on their findings and some general statements concerning the Swiss mountain topography.

" … . Specifically for the typical Alpine topography it could be shown that valley wind systems of the major Alpine valleys like Rhone, Rhine, and Ticino, which have typical valley with of 4 to 8 km and, hence, cannot be sufficiently resolved at 7 km model resolution, are much better captured at 1 km model resolution (Schmidtli et al., 2018). Other smaller scale valleys remain too narrow to be properly resolved even at 1 km resolution. Another important feature of the Swiss Plateau is the flow channeling between Alps

and Jura mountains. With a distance between those two mountain chains of approximately 50 km this channeling is generally resolved at 7 km already."

Furthermore, we come back to this point when discussion model performance in Section 3.2 following line 546, pointing out improvements in model performance in the C1 a-priori simulations, which we mostly attribute to improvements in flow representation in complex terrain.

"*A general performance improvement of FLEXPART-COSMO-1 versus FLEXPART-COSMO-7 can already be seen in the a-priori simulations (see supplement Table 2), where it is solely due to improvements in the transport description/flow in complex terrain, as emission distribution and baseline values were the same for both sets of simulations.*"

Line 71: "Thus, a potential decline of fossil fuels will possibly see increased emissions from refrigerants" Can you explain the logic?

As we explained in the sentence above, tackling HFC emissions is often thought of as the low hanging fruit when it comes to reducing greenhouse gases since HFCs are currently much less connected to the energy sector, which is responsible for a large fraction of global anthropogenic CO2 emissions. However, when moving away from burning fossil fuel in the heating sector by broadly introducing heat pumps, a large demand on refrigerants will result, some of which will eventually find their way into the atmosphere.. We slightly revised the sentence to make sure the connection with the heating sector becomes clearer.

"*Even if they do not play a significant role in the energy system now, they are increasingly used in heat pumps and air conditioners. Thus, a potential decline of fossil fuel use for heating may possibly see increased emissions from HFCs used as refrigerants.*"

Line 190: How did you aggregate the observations?

The aggregates represent simple 24-hour mean values of all available observations within a given day. No further filtering by time-of-day was applied. We deemed this reasonable, since we recently showed that with the higher-resolution transport model we are able to capture the diurnal variations of trace species at the Beromünster tall tower reasonably well (Katharopoulos et al., 2022). As mentioned, we also ran sensitivity inversions with 3-hourly observations without seeing significant differences to 24-hourly observations. Hence, for the sake of computational costs we decided for 24-hour observations. Information on the aggregation method was added to the text.

"*To run our inversions, 24-hourly (3-hourly for sensitivity inversion) mean values were produced from the available observations of the above-mentioned sites.*"

Line 197: Regarding REBS, can you mention other methods? Is it possible to use Thin Spline for instance? Wood, S.N. (2003) Thin-plate regression splines. Journal of the Royal Statistical Society (B) 65(1):95-114.

We added an additional sentence on alternative methods after describing the details of the REBS approach. In general, differences between these purely observation-based approaches are not very large. Other approaches that use additional trajectory information may differ more strongly but obviously depend on a transport model. Thin-plate regression splines could be an additional option, however, the combination of a smoothed curve fit with a rejection/weighting scheme as provided by REBS seems to be the most promising approach.

"Other statistical methods for baseline estimation have been applied to greenhouse gas observations (e.g., Thoning et al., 1989; El Yazidi et al., 2018) some of which use additional transport model information (trajectories or footprints) to select background sectors (O'Doherty et al., 2001). Differences between estimated background conditions are often small or limited to certain events or situations. There is no consensus which of these methods is most robust under all circumstances."

Line 221: Is Flexpart capable of read and run with meteorological fields from ICON?

We are currently developing a version of FLEXPART for direct use of ICON output. This development is well advanced and we expect this version to become available to the ICON community towards the end of the year 2023. No changes to the text.

Line 244: A mention to Hysplit seems appropiate. Stein AF, Draxler RR, Rolph GD, Stunder BJ, Cohen MD, Ngan F. NOAA's HYSPLIT atmospheric transport and dispersion modeling system. Bulletin of the American Meteorological Society. 2015 Dec 1;96(12):2059-77.

Indeed, thank you for pointing out this lack. We added the reference to the revised manuscript as follows:

"Nowadays, FLEXPART (Stohl et al., 2005; Pisso et al.,2019), and other LPDMs like NAME, HYSPLIT and STILT (Jones et al., 2007; Stein et al., 2015, Lin et al. 2003), are utilized for a large variety of tracer transport problems …"

Line 309: It is not very clear, but it seems that to reduce computation cost, cells with lower sensitivities are aggregated. However, higher prior fluxes in the same cells can produce convolved emissions which may be not neglected. Comment, please.

By aggregating to larger grid cells away from the observations, we tackle two problems. One is the reduced sensitivity, which would create many state vector elements with very little individual impact on the observations (although, as pointed out, this may be different for some high emission cells). Second and probably more important, is the smoothing of transport model errors, which grow with distance from the release location. By using larger grid cells at larger distances, transport errors tend to be smoothed out and the impact of individual mismatches (like missing a large distant source by a few kilometers in the simulated footprint) will be reduced. The following information was added:

"The reduced resolution grid serves two purposes. On the one hand, it reduces the number of state vector elements, removing many elements with very little sensitivity. On the other hand, it helps to smooth out transport model errors, which tend to grow with distance from the point of observation."

Line 410: A recent publication states that SF6 seems appropriate (Hu et aL., 2023)

Hu, L., Ottinger, D., Bogle, S., Montzka, S. A., DeCola, P. L., Dlugokencky, E., Andrews, A., Thoning, K., Sweeney, C., Dutton, G., Aepli, L., and Crotwell, A.: Declining, seasonal-varying emissions of sulfur hexafluoride from the United States, Atmos. Chem. Phys., 23, 1437–1448, https://doi.org/10.5194/acp-23-1437-2023, 2023.

Thanks for mentioning this article. We added the reference to the discussion of SF6 priors.

Line 446: Can you scale priori emissions according to temperature and season? Furthermore, for these emissions, using roads as a proxy seems more appropriate rather than the presented methods. Can you comment on that?

We did not scale the a priori emissions by some temperature proxy, but allowed the inversion to pick up any seasonal variability. Similarly for the spatial distribution of the emissions. We allowed the inversion to optimize it and discuss the result in terms of the spatial distribution of the road network, in section 3.2. No additional text added to the manuscript.

Line 453: Do you believe that scaling prior emissions with temperature by season would improve your posterior estimates? Please, comment.

From previous inversions on the same spatial scale but for CH4 and N2O we know that the inversion is very much able to pick up seasonal and monthly variability even if the a priori does not contain any variability. Hence, we did not think it necessary to prescribe such scaling for the HFCs either for which we expected smaller seasonal variations than for N2O emissions, which are strongly driven by soil processes and, hence, temperature and soil water. We added the following to the manuscript:

From previous inverse modelling of Swiss CH4 and N2O emissions (Henne et al., 2016; FOEN, 2022) we know that the inversion was able to realistically pick up seasonal variability even if the a priori emissions did not include any variations in time.

Line 477: A significant test, such as wilcox or mann-whiteney is required.

In line 476 we changed the use of "no significant spatial differences" to "no large spatial differences", since we agree that no statistical test confirms this observation.

Lines 487-491: Can you comment other methods?

We assume that this question aims at different methods for baseline optimization in general. We added the following text to the manuscript to cover such methods:

"Estimating the baseline concentration purely from observations and optimizing it by site may not be the best solution to the baseline problem. Alternatively, baseline observations and transport model information can be used to reconstruct a spatially and temporally resolved baseline concentration at the domain boundaries from which, again with the transport model information, a baseline concentration for

each site and time can be sampled (e.g., Manning et al., 2021; Hu et al., 2023). Baseline concentrations at the domain boundary may then be included as part of the state vector."

Lines 580-583: Paragraph with only two phrases. Each paragraph must have at least three phrases.

In the revised manuscript, we combined these two very short paragraphs to one paragraph.

All figure uses too much blank space. It would be better remove longitudes and latitudes and use white space, leaving only latitudes at left side and longitudes at bottom side. In this way, the figures in the middle, would be without coordinates leaving more space to see the figures.

We thank the referee for this suggestion. We adjusted the amount of white space in figures 2, 8, 10, and 11, where this issue was most prominent.

Additional References

Hu, L., Ottinger, D., Bogle, S., Montzka, S. A., DeCola, P. L., Dlugokencky, E., Andrews, A., Thoning, K., Sweeney, C., Dutton, G., Aepli, L., and Crotwell, A.: Declining, seasonal-varying emissions of sulfur hexafluoride from the United States, Atmos. Chem. Phys., 23, 1437-1448, doi: https://doi.org/10.5194/acp-23-1437-2023, 2023.

El Yazidi, A., Ramonet, M., Ciais, P., Broquet, G., Pison, I., Abbaris, A., Brunner, D., Conil, S., Delmotte, M., Gheusi, F., Guerin, F., Hazan, L., Kachroudi, N., Kouvarakis, G., Mihalopoulos, N., Rivier, L., and Serça, D.: Identification of spikes associated with local sources in continuous time series of atmospheric CO, CO2 and CH4, Atmos. Meas. Tech., 11, 1599-1614, doi: https://doi.org/10.5194/amt-11-1599-2018, 2018.

Manning, A. J., Redington, A. L., Say, D., O'Doherty, S., Young, D., Simmonds, P. G., Vollmer, M. K., Mühle, J., Arduini, J., Spain, G., Wisher, A., Maione, M., Schuck, T. J., Stanley, K., Reimann, S., Engel, A., Krummel, P. B., Fraser, P. J., Harth, C. M., Salameh, P. K., Weiss, R. F., Gluckman, R., Brown, P. N., Watterson, J. D., and Arnold, T.: Evidence of a recent decline in UK emissions of hydrofluorocarbons determined by the InTEM inverse model and atmospheric measurements, Atmos. Chem. Phys., 21, 12739-12755, doi: 10.5194/acp-21-12739-2021, 2021.

O'Doherty, S., Simmonds, P. G., Cunnold, D. M., Wang, H. J., Sturrock, G. A., Fraser, P. J., Ryall, D., Derwent, R. G., Weiss, R. F., Salameh, P., Miller, B. R., and Prinn, R. G.: In situ chloroform measurements at Advanced Global Atmospheric Gases Experiment atmospheric research stations from 1994 to 1998, J. Geophys. Res., 106, 20429-20444, doi: 10.1029/2000JD900792, 2001.

Thoning, K., Tans, P., and Komhyr, W.: Atmospheric Carbon Dioxide at Mauna Loa Observatory 2. Analysis of the NOAA GMCC Data, 1974–1985, J. Geophys. Res., 94, 8549-8565, doi, 1989.