

Review of "Inversion of long-lived trace gas emissions using combined Eulerian and Lagrangian chemical transport models" by Rigby et al.

General

The study presents a very promising, novel method for global and regional scale emission estimates, combining large scale emission sensitivities obtained from a Eulerian model and regional scale emission sensitivities obtained by backward runs of a Lagrangian model. The methods are described in detail and the application to SF₆ is demonstrated. The manuscript is well structured and well written, but lacks further discussion of the obtained results and envisaged improvements compared with previous methods. After extending the discussion and incorporating some further specific comments the manuscript should be well suited for publication in ACP.

Major comments

1) Extend discussion of results: While the authors present the applied method in great detail, they neither provide a detailed discussion of the obtained SF₆ emissions nor of the expected benefits of the combined inversion in contrast to a purely regional or global scale inversion. I suggest to insert a new section ("Results and Discussion") after p14703,123 and extend the current discussion to the following points:

- What is the final benefit of the combined system over a purely regional scale emission estimate based on the Lagrangian model only?
- How do the emission estimates compare to other top-down estimates (e.g. Vollmer et al., 2009; Kim et al., 2010)?
- A number of sensitivity tests (inversion ensemble) were introduced on p14703. Their results are summarised as uncertainty ranges. It would be beneficial if the authors could comment on any kind of systematic conclusions that can be drawn from these sensitivities. For example do emissions generally increase/decrease if the data averaging period is changed?
- Model performance: Fig 8 presents time series of observed and simulated SF₆ mole fractions. However, no statistical (e.g. correlation, bias, RMS, etc.) estimates of the model performance are given. It would be good if such estimators would be included in the figure or an additional table. Furthermore, it would be interesting to see how these performance parameters improved (did they ?) from the priori to the posteriori emissions.

Minor comments

2) Selection of local regions: The extent of the local regions "was chosen based on the average footprint and the extent of significant emissions, as predicted by EDGAR" (p14699,23f). While there is more information given in the supplement on how local regions were aggregated, there is no further information on how the general extent of the local regions was determined. Looking at the local regions in North America and Western Europe that indicate quite low sensitivities towards the east and south-east as compared to the Asian region, I was wondering why the authors did use rectangular local regions. Wouldn't it make

more sense to cut these regions according to the shape of the footprints and the priori emission distribution right from the start?

3) Determination of $H_{LE,EUM}$: If I understand correctly the model domain of the LPDM is not the same as the local regions but extends globally. What was done with particles that left the local regions? Where they allowed to re-enter the same local region at a time further back in time? If that was the case then it might happen that certain parts of H_{LE} were estimated twice. Think for example of emissions in the south-west of your northern hemispheric local regions in the case of a high pressure system west of the domain. Such emissions would, in the forward runs, leave the domain on the southern or western boundary but possibly recirculate outside the region and re-enter it further north, finally reaching the observation site. If I understand correctly this situation would be accounted for by the Eulerian simulations and the difference between the realistic and the "powerful reactant" run would attribute such cases to $H_{LE,EUM}$. At the same time your Lagrangian model would see those emissions as well, if particles are allowed to leave the local region and re-enter it at a later time. Does this happen in reality? Was this accounted for? And if not what might be the possible influence?

What is the effect of using two different kinds of meteorology that drive the transport in MOZART and NAME? Might this create situations in which sensitivities are either accounted for by both models or even missed by both?

4) Section 2.2 and Fig 4: I did not find a definitions of the non-local regions. Could they be displayed in Fig 4 to show their extent?

5) Fig 2b: I understand that the blue curve is added to the red curve to give to total influence from the local region H_{LE} . However, I don't think this is well indicated in the figure caption. It might also help to display the contributions of the individual terms by filling the areas below the curves. This also to distinguish part b from part a, where the individual contributions are not added up.

6) p14700,115: What was the backward integration time? Were particles terminated when leaving the local domain (see above)?

7) Fig 5, 6: I did not find an explanation why emissions from North Korea and Slovakia were not estimated. From the footprints in Fig 4 I see no reason that those areas should be excluded due to low residence times.

8) Fig 7, 1: It is interesting to note that for countries residing in the local regions the uncertainty estimate for the posterior emissions is smaller (blue bars) than the uncertainty estimated from the sensitivity tests (black bars) and the other way round for countries mainly outside local regions. How can this be understood?

9) Fig 7, 2: Since the numbers displayed in Fig 7 are of great interest for other researchers and authorities, I would appreciate if they could be repeated in a Table, so that they can be readily accessed. In this context it would also be necessary to give the fraction of national emissions covered by the local regions and in order to be comparable to other studies an extrapolation to country total emissions.

Technical comments

10) Fig.8: What is the temporal resolution of the displayed data?