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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inversion of long-lived trace gas emissions using combined Eulerian and Lagrangian chemical transport models" *by* M. Rigby et al.

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We thank the reviewer for their constructive comments, which we respond to below. Reviewer comments are italicized, author responses follow each comment.

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.

We thank the reviewer for this positive assessment.

1 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,l23 and extend the current discussion to the following points:

We agree with this comment, and have created a new section.

What is the final benefit of the combined system over a purely regional scale emission estimate based on the Lagrangian model only?

The primary benefit of the proposed approach is that it allows emissions estimation on global, continental and local scales simultaneously. Regional estimates using only Lagrangian models are generally limited only to the local scale. Some authors have presented global estimates using Lagrangian models only (e.g. Stohl et al., 2009), however, assumptions about boundary conditions to each sub-region must be made, whereas boundary conditions are implicitly solved for in our method. Of course, the major draw-back of the proposed approach is that two models must be used. We feel that these benefits have already been articulated in the introduction. However, we now also include further justification for our approach in the new discussion section.

How do the emission estimates compare to other top-down estimates (e.g. Vollmer et al., 2009; Kim et al., 2010)?

Qualitatively similar results to these studies (and others) are obtained. For example, all studies find higher emissions for all major East-Asian countries than the EDGAR 2005 values. However, significant national-scale differences exist. We feel that this reflects the sparse measurement network (and a likely under-estimate in the uncertainties of each inverse method, e.g. due to modeling error). These comparisons are discussed in the new section.

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?

This is an interesting question, which we have investigated. However, coherent signals are difficult to find. For example, even a potentially simple change, such as a 2% increase in the AGAGE measurement scale does lead to higher national emissions on average, but can lead to a decrease in some countries (presumably due to some compensation for increased emissions from larger emitters). We feel that the paper would not greatly benefit from a discussion of these changes, since it is not central to the development of the 'combined sensitivities' approach. However, it will be important for future applications. We note that this 'ensemble' approach has been used to investigate the influence of assumptions used in the inversion by previous authors (e.g. Bousquet et al., 2011)

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. Further- more, it would be interesting to see how these performance parameters improved (did they?) from the priori to the posteriori emissions.

Three model performance parameters are now tabulated for the prior and posterior models (RMSE, bias and correlation). Improved performance was found following the inversion at every site (an improved model-measurement mismatch over all sites is required in a Bayesian inversion).

2 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?

The reviewer is correct that the method used to determine the extent of the local regions could be improved. The choice of the rectangular boundaries was chosen "by eye", to encompass areas of relatively high footprint and significant emissions. It would indeed be more elegant to chose regions based on the footprint multiplied by the prior emissions field. However, in practice, such a system requires that the locations of emissions and sensitivities within unusually shaped boundaries must be stored and manipulated in the code. This additional level of computational complexity is not thought to add much value to the work, since it is not of central importance to the methods presented in the paper.

3) Determination of HLE,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 HLE 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 HLE,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?

The LPDM domains are regional, however the reviewer is correct that they are very much larger than the 'local' region domains in the paper. There is indeed the chance that parcels may leave the domain and re-enter it. We have actually accounted for this possibility by making the 'powerful reactant' domain 10 degrees larger than the 'local' domain, thereby allowing the air in the Eulerian model to leave and re-enter before being destroyed. However, we did not explain this in the original submission. We thank the reviewer for pointing this out, and have included a line in Section 2.4 explaining this.

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?

Our assumption in the paper is that both sets of meteorological analyses are correct. This is clearly not the case, and is a limitation of this and almost all other 'top-down' modeling efforts. In future applications of this method, it is clearly preferable to perform an ensemble of inversions using multiple meteorological data sets in both models. However, having said this, it is not immediately clear to me that using two different meteorological drivers necessarily induces additional uncertainty into the calculations. One model is largely determining local-scale processes, while the other is determining large-scale transport. Even in single models, different parameterizations of various processes may induce different errors at small scales, compared to the larger scales.

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?

We have clarified this in section 2.2: "Sensitivities to emissions from each continent (North America, South America, Europe, Asia, Africa, Oceania) are shown, excluding emissions from four local regions described in Section 3."

5) Fig 2b: I understand that the blue curve is added to the red curve to give to total influence from the local region HLE. 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.

We have added a line to the figure caption saying: "The blue line has been added to the red line to show the overall sensitivity due to this grid cell (Equation 4)."

6) p14700,l15: What was the backward integration time? Were particles terminated *when leaving the local domain (see above)?*

The backward integration time was 13-days. We have clarified this in Section 3: “36,000 particles were tracked in the NAME model for 13-days prior to each measurement, with a time-resolution of 3-hours."

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.

This is because there are no estimated emissions in the EDGAR database for these countries. The inverse method that we employed scaled the available prior information, and therefore emissions cannot be determined where no prior information exists. An alternative inverse method could be used that, for example, solved for an offset to be added to each region, rather than a scale factor. This would then allow non-zero emissions to be solved for in countries like Slovakia. However, we preferred the approach used in Section 3 in this instance, since it retains the relative EDGAR emissions distribution within each region. We also note that it is certainly possible that there are very low (or zero) emissions from these countries.

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?

This is indeed an interesting point. There are at least two possible reasons for this. Firstly, the error in the AGAGE measurement scale (which was changed in some of the sensitivity runs) leads to changes in the local emissions. Secondly, the fact that we obtain different emissions for different averaging periods and numbers of regions suggests that the way in which we have calculated the model-measurement uncertainty

may not be entirely consistent for each case, and/or that the assumption of independent (uncorrelated) observations may be more or less appropriate for each sensitivity run. Whilst these factors clearly have a significant influence on the derived emissions, we do not feel that the paper would greatly benefit from a more detailed investigation of these sensitivities. In future ‘application’ papers, these considerations will be crucial.

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.

These numbers have been made available in the supplement.

3 Technical comments

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

5 days. This has been added to the figure caption.

References

Bousquet, P. et al., 2011. Source attribution of the changes in atmospheric methane for 2006-2008. Atmospheric Chemistry and Physics, 11(8), pp.3689-3700. Available at: http://www.atmos-chem-phys.net/11/3689/2011/

Stohl, A. et al. (2009), An analytical inversion method for determining regional and global emissions of greenhouse gases: Sensitivity studies and application to halocarbons, Atmospheric Chemistry and Physics, 9(5), 1597-1620

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 14689, 2011.