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Interactive comment on “Fine particulate matter source apportionment using a hybrid chemical transport and receptor model approach” by Y. Hu et al.

Anonymous Referee #3

Received and published: 11 December 2013

1 Overview

The manuscript by Hu et al. introduces a hybrid source apportionment approach that utilizes information from Eulerian chemical transport modeling and spatially distributed emissions inventories to improve receptor-model factor analysis. The attempt to merge these two generally unique approaches to constraining sources is interesting and relevant. Overall, the manuscript would benefit from clarify a few aspects as detailed below.

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2 Comments and questions

- Most of the discussion of the results is related to the comparison of the factor contributions before and after optimization. However, it isn't clear from the optimization results themselves how much of an improvement the rescaling provides. For several species, the bias seems to have gone from positive to negative, but they are still significant. Can the authors focus more on this? Was data that was not included in the optimization used as part of the assessment of the refined model estimates? Figures 2 and 3 are useful in this regard, but I'm not sure about the statement " χ^2 is reduced by 98% on average." Average over what? χ^2 is a single scalar number – its reduction should be absolute. It's also not clear what is plotted in Fig 3 – perhaps the individual residuals themselves?
- It seems evident that photochemical modeling and the a priori information of spatially distributed emissions inventories can be used to improve RM. However, what wasn't really made clear was how the proposed approach would be an improvement over full Bayesian CTM-based optimizations. What is added by using the CTM information in the context of receptor modeling?
- 26676.27: It wasn't intuitively obvious to me why the hybrid method would find lower secondary contributions than RM methods – could this be explained?
- 26674.14: The text points out that SM source apportionment would benefit from using measurements. Photochemical models are indeed subject to uncertainty; they can be improved using data assimilation. But this is widely known to begin with, as evident by the large body of work using such approaches, so I'm not really sure what is being concluded here.
- 26674.25: Some of the species being discussed here (NH_3 , nitrate) have very nonlinear model responses, so this doesn't seem to fit with the earlier claim that

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- higher order sensitivities are negligible. Has truncation of the 2nd order sensitivity terms from Eq. 5 really been justified for these aerosol species? They will certainly be sufficient for the minimization aspect, since L-BFGS just needs the local gradients, but it might be a bit of a stretch for the source attribution results.
- 26661.11: Application of SM approaches is actually quite an active field, for example there are large communities using CTMs to constrain sources of trace-gases based on remote sensing observations. There are definitely uncertainties involved with these approaches, but I wouldn't necessarily call their application limited.
 - 26671: What is the theoretical basis for expecting that the optimal solution comes from a regularization parameter value such that the two terms of the objective function are equally balanced? Would standard methods for estimating Γ (such as an L-curve technique) be more suitable?
 - 26662: It seems that discussion of the sensitivity calculations is inserted a bit awkwardly into the boiler-plate model description. Maybe it would fit better a bit later, after finishing the description of the meteorology and emissions used to drive CMAQ?
 - The data presentation leaves a bit to be desired – most results are presented in long tables, or supplemental tables. Can the sector information be visualized in some way?
 - 26665.9: I thought, based on 26664.10, that the initial and boundary conditions were being ignored.
 - 26658: suggest “demonstrating” rather than “revealing”
 - Figure 2: Could the text in the figures (axis, legends, etc.) be made larger? They are a bit hard to read.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 26657, 2013.

ACPD

13, C9896–C9899, 2013

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