Atmos. Chem. Phys. Discuss., 13, C11984–C11992, 2014
www.atmos-chem-phys-discuss.net/13/C11984/2014/

Atmospheric Chemistry and Physics Discussions

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

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Received and published: 6 February 2014

Reply to Referee #3

Comment: 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.

Response: Thanks.

Comment: 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 "X2 is reduced by 98% on average." Average over what? X2 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?

Response: An indication of the overall accuracy of the adjustments can be found using the weighted least square error of all species (i.e. X2 as expressed in Eq. 13).

On this regard, our scaled results showed that the refined X2(c,adj) (Eq. 13 with obtained Rj) are much smaller than the corresponding initial X2(c,base) (Eq. 13 with Rj equal to 1) as seen as scatter dots in Figure 3 (each dot represents one pair of X2(c,base) and X2(c,adj)). The correlation between X2(c,adj) and X2(c,base) reveals a 98% reduction on average from X2(c,base) to X2(c,adj). However, this overall reduction doesn't mean every individual species have better performances. Note that several elements with very low ambient concentrations (e.g. near the measurement uncertainty) were found to have slightly deteriorated agreement. On the other hand the X2(c,adj) represents the remaining prediction error. As the referee observed "for several species, the bias seems to have gone from positive to negative, but they are still significant". However, the magnitude of overall error, X2(c), decreases, and instead of most species being biased in one direction, there is a mixture of positive and negative biases. This is expected as there are likely errors in the source profiles, i.e., source profile fractions for some species are high, while others are low. Thus, even if we perfectly estimate the source impact (which we do not), some of the species estimates

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will be low, some will be high. What is important is that the overall error and bias are reduced. We will add further discussion on this particular aspect of the results.

Comment: 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?

Response: As the referee has pointed out, our hybrid method is in fact a RM framework utilizing CTM information. It can be applied to "correct" source apportionment results obtained from any CTM-based method. There are many CTM-based source apportionment datasets out there that need to be "corrected". These multi-year spatially-distributed source apportionment datasets resulted from various CTM applications over the years (http://www.epa.gov/heasd/research/cdc.html). These applications were conducted with different models and different NEI inventories for different years. The hybrid method is a quick and efficient solution for improving source impacts estimates of existing datasets.

Comment: 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?

Response: As noted in the sentence, in this simulation the results tended to find lower secondary contributions vs. RM methods, but this was not always the case (e.g., Chicago and Pittsburgh). This result is time period and location specific. For the case at hand, the simulated nitrate from CMAQ tended to be biased low in the base simulation, and the approach used here will adjust the nitrate upwards to better match the observed value, but will not force it to match the observation. Looking at eq. 14, the second term on the right hand side penalizes adjusting the impact based upon the estimated uncertainty in the emissions (e.g., of NOx in this case). Given that estimated emissions of NOx from power plants are viewed as well estimated, and emissions from

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mobile sources are not as uncertain as say, dust emissions, this term will limit the adjustment of impacts from those sources more than many other sources. Typical RM methods do not have a similar term. If the base simulation had a high bias, the hybrid approach will adjust the impact down, but still not to match observations exactly.

Comment: 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.

Response: We are modifying the sentence to say "This further supports that SM source apportionment results should be evaluated using measurements".

Comment: 26674.25: Some of the species being discussed here (NH3, nitrate) have very nonlinear model responses, so this doesn't seem to fit with the earlier claim that 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.

Response: We agree that it would be more accurate to include the higher order sensitivities in approximating source attribution results, especially for sectors that contribute to nitrate and ammonium. In fact our hybrid method, as designed, can add source impact portions estimated from using higher order sensitivities when they are available. We will change the corresponding discussions to reflect this. As noted in reply to Referee #1, multiple studies have assessed the use of first and second order sensitivities when conducting source apportionments.

Comment: 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

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these approaches, but I wouldn't necessary call their application limited.

Response: We will change this sentence to "Due to these uncertainties and the required level of effort, SM approaches are not as widely used as RM methods for conducting PM2.5 source apportionment".

Comment: 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 Gamma ïĂă(such as an L-curve technique) be more suitable?

Response: We based this expectation on L-curve plots of the values of the two terms in the objective function that we calculated for a series of Gamma (as in Eq.14) values. As an example, the following L-curve plot (Figure 1) is made from the case of January 4, 2004 at the Atlanta CSN site.

The next plot (Figure 2) shows that the prediction error (X2Ci) is kept relatively small when constraining the second term (X2Rj) just off of the optimal Gamma, i.e. by using a slightly larger Gamma.

Comment: 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?

Response: We will move the discussion of the sensitivity calculations to a later section. Along with the discussion of the sensitivity calculations for approximating source-impact estimates, we will add a discussion of other CTM-based source apportionment methods as well.

Comment: 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?

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Response: We tried stacked bar charts instead of tables to show the source impact estimate results. However, since we have 33 sectors, the stacked bar charts became too crowded and it was very difficult to read an individual sector's contribution. However, we will add a stacked bar chart to present results for a reduced number of sectors such as 13 sectors.

Comment: 26665.9: I thought, based on 26664.10, that the initial and boundary conditions were being ignored.

Response: On page 26665 we describe the general formulation of source impacts estimation by using DDM-3D calculated sensitivity coefficients. Initial and boundary conditions can be treated as "sources". In our case study for January 2014, we found the impact of initial and boundary conditions to be very small. While they are inherently included in the model simulations, we do not modify their impact via the hybrid method. We will rearrange the order of these descriptions to clarify the issue.

Comment: 26658: suggest "demonstrating" rather than "revealing"

Response: We will change the wording as suggested. Thanks.

Comment: Figure 2: Could the text in the figures (axis, legends, etc.) be made larger? They are a bit hard to read.

Response: We will make Figure 2 more readable.

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

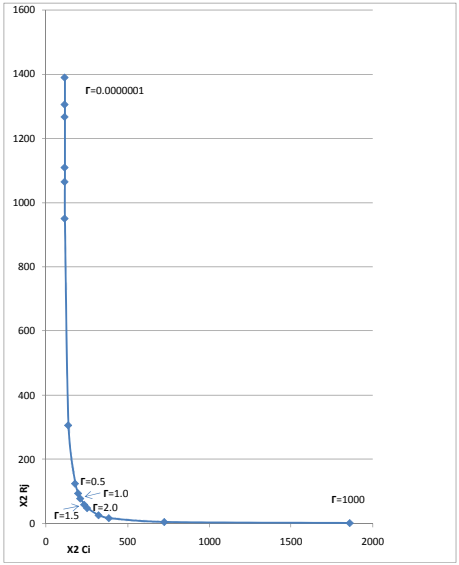

Fig. 1.

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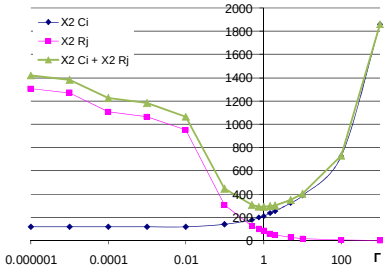

Fig. 2.

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Equations:

$$\chi^2 = \sum_{i=1}^{N} \frac{\left(c_i^{obs} - c_i^{base} - \sum_{j=1}^{j^{CIM}} (R_j - 1) SA_{i,j}^{base} \right)^2}{\sigma_{c_i^{obs}}^2} \quad (13)$$

$$\chi^2 = \sum_{i=1}^{N} \left[\frac{\left(c_i^{obs} - c_i^{init} - \sum_{j=1}^{j^{crw}} (R_j - 1) SA_{i,j}^{init} \right)^2}{\sigma_{c_i^{obs}}^2 + \sigma_{SR_i^{crw}}^2} \right] + \Gamma \sum_{j=1}^{j^{crw}} \frac{(\ln R_j)^2}{\sigma_{\ln R_j}^2} \quad (14)$$

Fig. 3.

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