Atmos. Chem. Phys. Discuss., 13, C8954–C8960, 2013
www.atmos-chem-phys-discuss.net/13/C8954/2013/

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.

Anonymous Referee #1

Received and published: 8 November 2013

This manuscript illustrates an approach to estimate source sector contribution to PM2.5 by combining well known source and receptor based methods. The source based method is the application of the CMAQ photochemical transport model using DDM sensitivity coefficients to estimate the impacts for a wide range of source sectors. The receptor based approach is a traditional Chemical Mass Balance (CMB) receptor model that uses source fingerprints of trace metal compounds to differentiate chemically speciated measurements into various source groups. The source based approach estimates of sector contributions (CMAQ DDM) are adjusted based on source sector contributions estimated with CMB using 24-hr average CSN speciated measurement

data. Ultimately, source sectors with very poorly estimated primary PM2.5 emissions (such as the dust sector) in the 2002 National Emission Inventory (NEI) are reduced through ambient based constraints.

The authors start from an interesting premise that seeks to utilize strengths of both systems to better estimate source contribution to PM2.5. Photochemical model approaches are powerful in that collinearity is never an issue and they can attribute secondarily formed pollutants, but the estimates are only as good as the underlying emissions inventory and host model representation of physical and chemical processes in the atmosphere. Receptor models using routinely measured speciated PM2.5 (e.g. CSN, IMPROVE) are severely limited by collinearity issues (many sources have similar emissions profiles) and rarely attribute the bulk of the measured mass to specific sources as most is generally secondarily formed.

With regard to the general approach, I have several concerns. It is not clear that the limitation inherent with CMB in apportioning secondary PM2.5 mass does not translate to a similar limitation in this "hybrid" approach. Since the receptor modeling (CMB) used to adjust the CMAQ DDM source estimates only can resolve sources that emit primarily emitted PM2.5 this approach only really changes CMAQ DDM source sectors that have primary PM2.5 emissions. Based on presented Tables showing contribution as a concentration, sectors such as confined animal operations ("livestock") that emit ammonia and biogenics that emit nitrogen oxide and VOC do not substantively change after the "refined hybrid approach" is applied.

This approach does well for sources that are grossly overestimated due to emissions inventory errors such as the fugitive dust sector. However, that could be achieved by simply applying model performance bias ratios against the model predicted species to obtain similar results without a back-end receptor model. This approach may have promise as a diagnostic tool to identify parts of the emission inventory with overestimated primarily emitted PM2.5. A major limitation is this approach does not seem to make meaningful adjustments where sectors do not emit primary PM2.5 and since

contributions go down after hybrid method application it also may not work well where emissions are underestimated. I encourage the authors to test how robust this tool is with extra diagnostics such as excluding a known source from the photochemical model or making a systematic adjustment to a well characterized category such as EGUs and see how well the hybrid system "corrects" the perturbation.

The current description, formulation, and presentation of results gives the appearance that the hybrid approach somehow "improves" sector estimates that are entirely secondary in nature. Perhaps that is why such a short episode in a winter month was chosen for the evaluation period; to minimize the limitations in the approach that would be even more evident in the warmer seasons when secondary PM2.5 is greater. Why was January 2004 selected? Does it coincide with elevated PM2.5 in the areas selected? Is this method too inefficient to apply for longer periods of time? The authors need to apply this approach to a variety of seasons to include the range of physical and chemical processes important for PM2.5 formation.

The manuscript needs to clearly explain why the authors choose an emissions sensitivity approach to estimate sector impacts rather than photochemical model source apportionment techniques like PSAT in CAMx (ENVIRON, 2013) or TSSA in CMAQ (Wang et al., 2009). Source apportionment for PM2.5 has been available in CAMx since version 4.5 of CMAQ was released (which was used for this study) and TSSA was implemented in a version of CMAQ contemporary with version 4.5. Receptor models like CMB, PMF, and UNMIX provide information about a specific measurement and do not provide information about how that measurement would be different if emissions were zeroed out from large sectors because chemistry would fundamentally change especially in nonlinear systems like PM2.5 nitrate. It seems to make sense to match a photochemical model source apportionment approach with receptor model source apportionment as opposed to matching a source sensitivity approach (e.g. DDM, brute force emissions changes) with source apportionment. Please provide some discussion in the manuscript that supports why a source sensitivity approach better matches the

type of information obtained from a receptor model source attribution approach.

The author's choice of first order sensitivities using DDM is not supported. First order DDM sensitivities best approximate a small perturbation in a linear system, which is the reason higher order sensitivities were later included in the method (HDDM). PM2.5 nitrate is not a known linear system and would seem to need higher order terms and possibly interaction terms. The reference provided to support the use of first order DDM sensitivity coefficients to represent PM2.5 impacts is to (Hakami et al., 2004), which is a comparison of small perturbations in ozone precursors to DDM estimated ozone. An evaluation of the ozone DDM system is not a substitute for the PM system.

Finally, I am very concerned by the author's choice of an outdated photochemical transport model to illustrate their approach. CMAQ version 4.5 (most notably for this work) has outdated PM chemistry, particularly related to organic aerosol. CMAQ version 4.5 was released in September of 2005. CMAQ 5.0 was released in February of 2012 and version 5.0.1 in September of 2012 with versions 4.6 and 4.7 released in the years between 2005 and 2012. The use of emissions based on the 2002 NEI is also puzzling. Updated national emission inventories have been released for 2005 and 2011 and have improved on many sectors, including the "dust" category.

The authors need to work within a modeling system framework that is relevant and contemporary. That means using a photochemical model version that is a recent representation of the state of the science, a recent emission inventory, and applying the photochemical model for at least an entire calendar year (preferably a recent year or a meaningful period of time that coincides with elevated PM2.5 or a special field campaign). While not a requirement, it would be preferable to use 12 km sized grid cells rather than 36 km and employ a vertical representation from the surface to at least 100 mb, which is how most ozone and PM2.5 photochemical model applications for regional and urban scales are currently applied.

More specific comments follow.

Abstract.

I disagree that this approach accounts for emissions uncertainties. The authors provide an estimate of uncertainty for each emissions sector but the method itself does not estimate these. I would agree that all photochemical model applications must be evaluated with observations as noted in the abstract (regardless of intended use) but the abstract seems to suggest this hybrid method must be done for every source attribution approach as the “evaluation”.

Introduction.

Please provide some references to support the sentence at lines 11-15 of 26659. Since this paper is about source apportionment it is important that the authors specifically acknowledge the existence of photochemical model source apportionment ((ENVIRON, 2013; Fann et al., 2013; Kwok et al., 2013; Wagstrom et al., 2008; Wang et al., 2009)) and provide some explanation why this approach would not be better suited to be matched with a receptor based source apportionment model.

Methods.

The authors need a much more contemporary model and model application. An advantage of using one of the more recent CMAQ releases is that they include most crustal species which would require fewer assumptions about speciation in the post processing steps. Support for ignoring higher order sensitivities for nonlinear systems like PM2.5 nitrate formation is needed. Do the first order sensitivities add up to the bulk estimate? How are the authors able to show full mass closure in the Tables showing percent contribution when lateral boundary inflow is ignored? It would likely be small for a winter month but bigger than many of the small categories chosen for this analysis.

On page 26665 at the bottom of the page, the authors state they are ignoring higher order DDM terms based on (Hakami et al., 2004). This paper only evaluated first and higher order terms for ozone and provides no such evaluation for PM2.5! Other

papers by the same authors suggest higher order terms are needed to approximate a 50% change in NOX on modeled ozone (Hakami et al., 2003). Here, the authors are assuming far beyond 50% reductions as sensitivities are being used to approximate model response when 100% of the emissions for a particular sector are not included in the simulation.

Results.

The comparisons with other receptor model studies should only be included for Atlanta since they are both on similar (the same actually which is good) time scales. The others should not be included unless the authors apply their approach for at least an entire year otherwise it makes little sense to compare January 2004 with studies that looked at contributions over multiple years.

Table 3.

It is peculiar that this approach doesn’t result in substantially better model performance. PM2.5 nitrate performance seems to be worse using the refined approach. Are the authors sure this method should be mandatory for people using source based attribution methods?

Table 4.

The “refined” total absolute contribution (as a concentration) always seems to be lower or the same as the original estimate. Does this approach work best in situations where the model is clearly overestimating primarily emitted PM2.5 and not as well when the model is missing a sector or grossly underestimating the primarily emitted emissions for a sector? That seems like a very important aspect of this that needs explanation and discussion.

Table 5 is redundant with later Tables.

Figure 1 is very difficult to read. Please just include the areas and monitors used in this analysis.

Table S8 provides emissions totals for each category. PM10 should not be on this table as it is not relevant to this paper.

References

ENVIRON, 2013. User's Guide Comprehensive Air Quality Model with Extensions. ENVIRON International Corporation, www.camx.com.

Fann, N., Fulcher, C.M., Baker, K.R., 2013. The recent and future health burden of air pollution apportioned across 23 US sectors. Environmental Science & Technology.

Hakami, A., Odman, M.T., Russell, A.G., 2003. High-order, direct sensitivity analysis of multidimensional air quality models. Environmental Science & Technology 37, 2442-2452.

Hakami, A., Odman, M.T., Russell, A.G., 2004. Nonlinearity in atmospheric response: A direct sensitivity analysis approach. Journal of Geophysical Research-Atmospheres 109.

Kwok, R., Napelenok, S., Baker, K., 2013. Implementation and evaluation of PM2. 5 source contribution analysis in a photochemical model. Atmospheric Environment.

Wagstrom, K.M., Pandis, S.N., Yarwood, G., Wilson, G.M., Morris, R.E., 2008. Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model. Atmospheric Environment 42, 5650-5659.

Wang, Z.S., Chien, C.J., Tonnesen, G.S., 2009. Development of a tagged species source apportionment algorithm to characterize three-dimensional transport and transformation of precursors and secondary pollutants. Journal of Geophysical Research-Atmospheres 114.

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