Response to Referee #2

Interactive comment on "Development of $PM_{2.5}$ source impact spatial fields using a hybrid source apportionment air quality model" by C. E. Ivey et al.

Anonymous Referee #2

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This manuscript seems to be prepared with care. However, it is not clear the substantial contribution to modelling science compared with the previous paper of Hu et al. (2014). Hu et al. (2014) developed a hybrid CTM-RM method to calculate adjustment factors to refine the CTM-estimated source impacts at monitoring sites. This paper simply spatially interpolates the impact adjustment factors using the commercial MATLAB software. If this is the "new method" developed in this paper, it has no enough novelty for publication in GMD, a premier international journal. If the authors can revise the manuscript to highlight what new scientific findings they made by spatially interpolating the adjustment factors (rather than emphasizing on the method itself), it is still possible for this manuscript to be published.

We would first like to thank the referee for the constructive review of our manuscript and his/her note about the care with which the manuscript is prepared. The integration of the Hu et al. (2014), Chemical Transport Model-Chemical Mass Balance Method with kriging and interpolation in time allows both increasing our understanding of the spatiotemporal characteristics of source impacts in the United States, as well as developing accurate source impact fields for health studies. What we have done is linked together multiple modeling approaches, e.g., CMAQ-DDM3D responses are integrated into a chemical mass balance method, which is then linked with spatial kriging and temporal interpolation, thus developing a larger modeling system approach. We use commercially available packages to do the integration because they are very efficient and this also allows other modelers who are interested in using the modeling approach descried here to more readily develop variants (e.g., alternative chemical transport models, optimization techniques, etc.).

In regards to the new scientific findings, in the initially submitted version of the current paper we downplayed the scientific findings that the spatial extension brings, as we emphasized the method, which is the typical focus of GMD. In the revised paper we expand the discussion of its application and develop increased scientific findings (e.g., biomass burning impacts, dust impacts, mobile source impacts). We will also extend the discussion of the use of the method as an approach to provide spatiotemporal fields of source impacts on air quality for use in acute health studies, the main purpose for the development of this modeling approach. We are currently using this approach to develop 8 years of source impact fields for use in epidemiologic health studies. When using the initial CTM-RM method, source impact estimates are available for 1-2 locations in a metro region. After applying the current method to get spatially resolved source impact estimates, we are able to see the spatial variability in 33 source categories at more than 20 grid locations in a metropolitan statistical area (MSA). This allows the spatial variability in sources to be detected through the modeling domain. The spatial extension is necessary for this application, and it is very unique. In the revision, we emphasize the scientific findings that were elucidated in our results, along with the application in health studies.

In addition, there are also some other major comments that should be addressed before publication, as described below. Specific comments:

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(1) Section 1. Introduction: The authors only used 2-3 sentences to describe the previous studies and their shortcomings. I think this part should be significantly expanded.

We extended this section. In particular, we make additional references to previous receptor- and source-oriented apportionment studies and discuss their strengths, limitations and uncertainties. In the initial version, we attempted to make the manuscript as concise as we thought reasonable.

(2) Section 2. Data and method: The configurations of the CMAQDDM modeling system should be described, e.g., the modeling domain, geographical projection, physical and chemical mechanisms, initial and boundary conditions, and emission inventory, etc.

We now provide the additional modeling configuration in the revised version.

(3) Section 2.4 Model evaluation: The authors should explain the objectives of each evaluation method.

We included an explanation of the objectives of each evaluation method Section 2.4:

"The hybrid optimization is directly applied to withheld observations to assess the performance of the kriging model. Concentrations are reconstructed using Eq. 3 and the spatially interpolated adjustment factors. Then the original CMAQ-DDM, directly applied hybrid (CTM-RM), and spatial hybrid (SH) concentrations are compared to observed concentrations at withheld observation locations to evaluate the performance of each method in simulating concentrations. Linear regression was used to assess correlations between observations and modeled concentrations for each method.

In order to evaluate prediction performance in remote locations and in locations independent of CSN, CMAQ-DDM and hybrid concentrations were compared to observations at SEARCH and IMPROVE locations."

(4) Section 3. Results; Section. 4 Discussion: These two sections should be reorganized. Firstly, the spatial extension method should be evaluated before any discussion of the modeling results, so Section 3.5 and 3.6 should be moved ahead of Section 3.1-3.4. Secondly, the discussion section is long and complex, with model performance, modeling results, advantage/shortcomings mixed up; the majority of this section is actually "results" rather than "discussions". I would suggest the author to merge the "results" and "discussion" sections and move most of the "discussions" to the corresponding sub-sections of "results".

We reorganized the results and discussion section so that scientific findings information is located in the in a separate section of the paper. We moved the spatial evaluation before the results section.

(5) Section 3.1, P653 L10-14: The authors should explain why the adjustment factors for specific sources are less than one, near one, or more than one.

The value of the adjustment factor Rj is specified depending on the initial CMAQ-DDM simulation of the source impact. If, after optimization, the Rj value is less than one, then the initial CMAQ-DDM estimate will be reduced to be more consistent with observations. In turn, if the Rj value is greater than one, then the initial CMAQ-DDM estimate will be increased to be more consistent with observations. An Rj value of 1 indicates that no adjustment to the CMAQ-DDM is necessary to improve consistency with observations. We included this explanation in the revision:

"In general, for an R value less than one, the initial CMAQ-DDM estimate was reduced to be more consistent with observations. In turn, for an R value greater than one, the initial CMAQ-DDM estimate was increased to be more consistent with observations. An R value of 1 indicates that no adjustment to the CMAQ-DDM is necessary to improve consistency with observations."

(6) Section 3.1, P653 L14-17: How much is the difference between these two methods?

The difference in the two approaches is the log-transformation of the Rj values before kriging. In one approach, we log-transform the Rj values at the monitors before kriging, and then the kriged values are unlogged before use in reconstruction. In the second approach, we do not log-transform before kriging. In this manuscript we have chosen the second approach, as the additional log-transformation step produces similar results as the second approach.

(7) Section 3.2: This section has the same problem with Section 3.1. Lots of modeling results are shown, but their reasons/implications are seldom illustrated.

We initially placed the reasons/implications in the discussion section. We address the referee's concerns by rearranging the information presented in the results and discussion, as stated in our response to specific comment (4).

(8) Section 3.3: This manuscript focus on the development of source impact spatial fields. However, only the source impacts at the withheld CSN monitors are illustrated in this section.

Section 3.3 is meant to compare kriged results to direct application of the hybrid method. Other sections address performance of the spatial extension, and we feel that section 3.3 brings added perspective of the benefits of the hybrid method. We have reorganized the results section and have expanded the discussion of refined spatial fields in Section 3.2.

(9) A conclusion section should be added. The limitations of this method, except for that described in the last paragraph of the manuscript, should also be summarized. For example, the accuracy of this method still needs to be further improved by optimizing source profiles etc., and this method cannot capture the nonlinearity in the source-receptor relationships.

We included a discussion of CTM and RM limitations in the introduction:

"Several receptor-oriented SA models have been developed to quantify emission source impacts on pollutant concentrations. Each model has its own unique characteristics and associated uncertainties (Balachandran et al. 2012; Seigneur et al. 2000). Schauer and Cass (2000) used organic tracers for source apportionment using the Chemical Mass Balance (CMB) method at two urban sites and one background site in central California (Watson et al., 1984). Their implementation addressed the improper accounting of VOCs from motor vehicle exhaust and wood combustion. Watson, Chow, and Fujita (2001) reviewed several studies that used CMB for source apportionment, and reported that uncertainties in source contributions of VOCs led to uncertainties in impacts from important sources such as off-road vehicles, solvent use, diesel and gasoline exhaust, meat cooking, and biomass burning. The authors also describe several limitations of CMB, including reliance on existing observations and overlooking profiles that change between source and receptor due to factors such as dilution, aerosol aging, and deposition. Maykut et al. (2003) used Positive Matrix Factorization (PMF) for source apportionment at an urban

Seattle, Washington (USA) site with selected trace elements to distinguish combustion sources (Pattero and Tapper, 1994). Temperature-resolved organic and elemental carbon fractions were also used in Unmix to distinguish diesel and other mobile sources but did not lead to significantly different results (Henry 2005). There was also difficulty in distinguishing small sodium-rich industrial sources due to the similarity to the aged marine aerosol source profile.

In an effort to improve the spatial and temporal resolution of SA data and improve source distinction, chemical transport models (CTM) have been adapted to estimate emission impacts on pollutant concentrations. Marmur et al. (2006) conducted a comparison of source-oriented and receptor-oriented modeling results for a winter and summer month in the Southeastern U.S. The brute force method was used in the Community Multiscale Air Quality (CMAQ) model to calculate impacts from mobile sources, biomass burning, coal-fired power plants, and dust. The authors determined that meteorological effects had a strong impact on the temporal variation of CMAQ source impacts, where receptor model results exhibited more day-to-day variability. Koo et al. (2009) used the decoupled direct method (DDM) in the Comprehensive Air Quality Model with extensions (CAMx) to determine the sensitivity of particle sulfate concentration to changes in emissions of SO2 and NOX from point sources; NOX, VOC, and NH3, from area sources, and all emissions from on-road mobile sources (Byun and Schere, 2006; Dunker, 1981, 1984; Napelenok et al., 2006). DDM first order-sensitivities under-estimated the impacts on sulfate concentration when all emissions are removed due to nonlinearities, as compared to brute force method results. Zhang et al. (2012) addressed this issue by calculating second order sensitivities of inorganic aerosols using DDM, which better captured nonlinear responses to changes in emissions up to 50%."

We also included a summary of limitations of the spatial hybrid method in the discussion:

"Spatial hybrid inputs, methods, and results have inherent uncertainties and challenges that are associated with implementation. Input uncertainties include measurement error and challenges are posed with temporal availability and spatial representativeness of concentrations. Emissions inputs for each source are available at different temporal and spatial scales. For instance point source emissions are available at hourly intervals in some cases, while dust emissions are highly variable, both spatially and temporally. Area source emissions are estimated at weekly or monthly intervals and averaged source fingerprints for the primary components of the PM2.5 emissions are used, which removes the consideration of locally-varying source composition. Physical processes in CMAQ-DDM are uncertain as modeling atmospheric behavior is a complex undertaking. Also, first-order sensitivity approaches may not capture all nonlinearities in source-receptor relationships. SH results are also subject to potential systematic bias from the optimization and kriging steps, though our evaluation suggests those biases are minimal"

(10) The figures and tables are not consistent with the citations in the main text. For example, in P653 L8, "Fig. 3" should be "Fig. 2". In addition, some figures and tables included in this manuscript are never cited or described in the main text, and some are cited but not described.

Thanks for catching this. We corrected the table and figure labels and removed those that are not referenced in the manuscript. This comment was also addressed in the review from referee #1.

Technical corrections:

P649, L3-4: "Chemical Speciation Network (CSN)" should be "CSN"

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Yes, CSN was already defined in the introduction. We corrected this in the revision.

P655, L22: How do you determine outlying data pairs?

Outlying data pairs are determined by examining the distribution of the directly calculated R values (mean = 0.837, stdev = 0.478) and the kriged R values (mean = 0.828, stdev = 0.294) at the withheld observation locations. Data pairs were removed if either value was more than six standard deviations from the mean R value. The removed data points (5 points out of 2475) were well outside of this range; hence we concluding that removing them would not alter the correlation. We added this explanation in Section 3.1.

"Outlying data pairs are determined by examining the distribution of the directly calculated R_j values (mean = 0.84, stdev = 0.48) and the kriged R_j values (mean = 0.83, stdev = 0.30) at the withheld observation locations. Data pairs were removed if either value was more than six standard deviations from the mean R_j value. The removed data points (5 points out of 2475) were well outside of this range."

P657, L1 and L4: What is the meaning of N? I deduce that it represents the number of observation sites but it should be explained.

Yes, N represents the number of monitoring locations from each network that were used for evaluation. We changed this to (N = 8 monitoring sites) and (N = 38 monitoring sites) to enumerate the number of monitors from each network that were used for evaluation.

Table S4 and S5: I guess "HYB" should be "SH" because only the latter is described in the main text.

Yes, HYB should be SH. We changed this in the revision.