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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 #2

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The manuscript by Y. Hu et al. describes a new approach to estimate source contributions to PM2.5 temporally and spatially based on CMB receptor model and CMAQ chemical transport model. It presents an interesting way to combine strengths of both receptor-based approach and emission-based approach and to give better results of source apportionment of PM2.5 than one model alone. The principles and methodology of the new hybrid approach are formulated clearly and the manuscript has been relatively well organized for such a topic. Under constraint by observations of PM2.5 and its chemical compositions, the new hybrid approach gives much better model simulation results of PM2.5 and species than original CMAQ prediction, and has reasonable

good estimations for contributions of 33 separate sources to ambient PM2.5 at the receptor sites, in which most of the sources are not resolved by receptor model without extra measurement information on unique tracers. The hybrid results from this study are generally consistent with traditional receptor model and also can be used to validate or refine relevant parameters in emission inventory. As a new method of PM2.5 source apportionment combining receptor-based approach and emission-based approach, it merits to be published in ACP. However, more detailed analyses are expected to make it complete and more convincing. In the following, I have a number of comments for the authors to address before publication.

1.The CMAQ performance statistics are well within the normal range of current state-of-the-art CTM's (page 26664, line 5). The simulated concentrations are found to be improved substantially compared to the initial simulation after refining source-impact estimates for major individual components and for most of the elements (page 26673, line 12). But it can be seen from Figure 2, both of the initial simulation and refinement are deviated from observations still quite large except for sulfate. Compared to observation, the original prediction is overestimated, but refinement is underestimated. This deviation might have significant influence on final results of the proposed new hybrid approach. The authors should quantitatively assess the impact of this deviation between model prediction and observation on source apportionment results.

2.Emission inventory and chemical speciation are essential for CMAQ to simulate PM2.5 and to estimate contributions from different sources. It is understood that emission inventory is not well established usually for some sectors, so scale factors are introduced to refine CMAQ estimations. In this manuscript, authors do not provide detailed description about the inventory used in the model. What is the accuracy of the used inventory in general? Is the same set of emission factors and source profiles applied to all cities nationwide, or different cities use different emission factors and source profiles? How to treat temporal and spatial variation for major emission sources? Those information are very important for explaining scale factors and their temporal and spatial variation.

3.Particulate source apportionment technique is available in CMAQ (TSSA) and CAMx (PSAT), which have been applied in some research projects. Because TSSA-CMAQ for source apportionment has the same problem as concentration prediction by CMAQ due to the uncertainty of inventory, the new hybrid approach might provide more reliable results by using observation as constraint. TSSA or PSAT should be discussed in the “Introduction” section or the “Results” section. Of course, it will be nice if the authors can show comparison results between TSSA and hybrid model in some cities during same time periods either from literature or from author’s work.

4.In page 26666, line 18, an effective fij* is defined, which is more or less similar as source profile used in traditional receptor model CMB. The fij* directly accounts for secondary formation of PM2.5 and nonlinearities in pollutant transformations. It is true for secondary aerosol, but not for elements in PM2.5 because there are not much chemical transformation for elements and their mass should be conservative in air if dry and wet deposition processes are not significant. Thus, fij* could be calculated and then be compared with source profiles to validate emission inventory and its chemical speciation. It can also be used to check the uncertainty of CMAQ modeling as well as the scale factors due to transport process.

5.Scale factor Rj is introduced to refine initial source apportionment results by CMAQ under constraint of observed PM2.5 and species. In principle, constant scale factors Rj for the same source should be found without temporal and spatial variation. However, it is partly true in this manuscript. Temporal and spatial diversities of Rj for the same source are still large, as seen in Fig S1 and Table S9. More explanation is needed for this diversity or variation. Is it caused by unified emission factor and source profile nationwide without area specific character, meteorological bias, or model bias? If Rj is mostly related to source uncertainty, I suggest that Rj value should be evaluated quantitatively by using the most recent emission factor and source profile.

6.The manuscript separated the primary and secondary contributions in the aggregated source impacts and merged the secondary portions correspondingly into ammonium sulfates, ammonium nitrate, and secondary organic carbon. It is worthwhile that authors provide some results for source contributions from different primary sectors to secondary species such as sulfate and SOA. 7. The comparisons with traditional receptor model should be during same time period at the same site. In Table 6, this is only true for the Atlanta site while other five sites use literature results in different time periods. For these five sites, it is suggested that authors reanalyze the dataset of same time periods using traditional receptor model.

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