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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Potential contribution of semi-volatile and intermediate volatility primary organic compounds to secondary organic aerosol in the Mexico City region" *by* A. Hodzic et al.

Anonymous Referee #1

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This paper summarises the results of a comparison exercise of a chemical transport model with atmospheric measurements of organic aerosol performed during MILAGRO in the Mexico City Metropolitan Area. The key theme of this paper is the use of the volatility basis set approach and the inclusion of schemes for the simulation of intermediate and semivolatile species (in the form of two published parameterisations and a reference run that does not include this). The authors find a general improvement in model accuracy when it is compared with AMS data, specifically the outputs from PMF analysis. However, various shortfalls remain, which are discussed.

This is a very topical paper and deals with an important subject, as there is currently

a major effort into improving the general predictive capability when it comes to organic aerosols in the atmosphere. There are a variety of different approaches that can be taken and each will require careful and critical evaluation against real-world data in order for the science to advance. The MILAGRO study is an excellent opportunity for this, as it had many sampling platforms and a number of groups performing modelling exercises independently of one another. I would say this work is important and ultimately will probably deserve to be published in ACP, but regrettably, the paper in its current form suffers from a number of serious shortcomings (detailed below). Because some of these problems relate to the interpretive side of the paper, I feel it prudent that this paper should be published only after major revisions and subsequent re-review.

General comments:

The most immediate problem with this paper is one of clarity. There is a general overuse of confusing acronyms and terms that a reader unfamiliar with this work would find rapidly overwhelming. For example, the first paragraph of page 661 required several reads before I could make sense of it. But even once the terms had been established, the remainder of the paper became frequently abstract and often left me no wiser as to the scientific case being established. Given the number of important and very specific quantities being dealt with in this paper, many of which have already been defined in other works, it would be wrong to request that these be changed. Instead, I would recommend that the important quantities be summarised in a table or a series of bullet points in the introduction, defining the acronyms and also giving a concise description of their physiochemical properties that the reader can refer back to.

The second major problem is a lack of focus in how the model treatments are being tested. The authors should be clear and consistent in what metrics they are using to test the models when comparing with observed data (for instance, was the % accuracy derived using linear regression, a ratio of means, or something more elaborate like orthogonal distance regression?). The authors do not pay nearly enough attention to the evaluation of how well temporal variability is captured, giving correlation statistics

in only a handful of places. If this work is to present a thorough evaluation of different model treatments (i.e. REF, GRI and ROB), it should be consistent as to which tests it is performing and present all of the results for each test more systematically.

This leads onto a more general conceptual issue as regards evaluating model performance. If REF gives a better result for an r2 test than GRI and ROB (as reported in section 3.2.3), then this would surely imply that it could give the best overall result if tuning was used. Granted, this wouldn't tell us anything about the underlying chemistry and may not lead to an overall skill improvement, but it does throw into question whether the GRI and ROB treatments are actually improving the model or whether they are getting the right answer for the wrong reasons. This issue must be discussed and discounted if the key conclusions of the paper are to be reached.

A major ambiguity when it comes to comparing outputs with AMS data is the issue of rotationality within PMF. It is possible that different solutions within the PMF analysis could lead to radically different assessments of both accuracy and temporal variations, but this topic gets little more than lip service in the current manuscript. As it stands, this is very inadequate. No information is given as to the reasons behind the choice of the number of factors or whether rotations were explored (e.g. by using the 'fpeak' parameter) and this should be documented. References to other papers or supplementary material would suffice, but this issue must be specifically summarised either way. Ideally, model outputs should be tested against alternative solutions to test how robust the comparisons are to this ambiguity.

Specific comments:

Please consider changing the title. The casual reader, who would probably not be aware of the terminology would be instantly turned off before they even got as far as the abstract. I would recommend changing it to something along the lines of 'An evaluation of atmospheric aerosol models incorporating treatments of intermediate- and semivolatile organic components' or something, as this is far more descriptive of

the work being presented.

I found the conclusions section a bit rambling and it left me slightly baffled as to what the core thesis of the paper is. If the authors are to make the case that the GRI or ROB treatments offer a genuine improvement in model performance or scientific understanding, they should state how the evidence shows this more clearly and concisely. If the authors are making specific recommendations for future model development, these and the evidence supporting them should also be made clearer.

I consider figure 12 to be potentially misleading and of questionable merit. The plot it recreates, figure 2 in Volkamer et al. (2006), was designed to highlight the gap in fundamental mechanistic understanding when it comes to SOA formation. In presenting the graph as an updated version of this figure, this could give a reader the false impression that the gap in understanding has been shown to have been closed by this work. However, the volatility basis set approach effectively makes up for the shortfall in the inventory of precursors and reaction routes by using semi-empirical parameterisations, so the lack of explicit understanding of the fundamental processes remains. It should also be pointed out that Johnson et al. (2006) were effectively able to achieve an observed/modelled ratio of unity through tuning, so using the runs featured in the Volkamer et al. (2006) plot as a basis for comparison with 'traditional' approaches is very unfair. The authors should by all means show that their approach delivers realistic mass concentrations (this, after all, is a key theme of the paper), but I would be strongly against the inclusion of figure 12 as it is currently presented.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 657, 2010.