Atmos. Chem. Phys. Discuss., 9, C2059–C2063, 2009
www.atmos-chem-phys-discuss.net/9/C2059/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling organic aerosols during MILAGRO: application of the CHIMERE model and importance of biogenic secondary organic aerosols" *by* A. Hodzic et al.

Anonymous Referee #2

Received and published: 21 June 2009

This paper presents an interesting study which demonstrates the capabilities and weakness of an air quality model when applied to the extremely difficult problem of organic aerosol. The paper does have a number of weaknesses, as discussed by referee #1, and below. On the other hand, the paper is unusual in presenting comparisons for a number of pollutants and meteorological drivers, and for consideration of emissions uncertainties, and the authors are to be commended for doing a thorough job in this respect (too many papers discuss just OA, and one never knows if the model is capable of reproducing other pollutants, or if the emissions are just plain wrong).

The more substantial problems are:

1. The authors find that anthropogenic and biomass burning emissions are 'reasonably captured', since their modeled POA match the measured HOA reasonably well. It can be argued that this agreement is a bad sign, since Robinson et al. (2007) and others have suggested that the 'emissions' of POA will quickly evaporate on dilution. This problem is not mentioned at all in the text, and needs to be discussed.

2. The methodology introduced in section 5.5 to compare column integrated SOA seems flawed to me. The authors simply multiply observed ground-level SOA (OOA) by the PBL depth, assuming a uniform concentration. This makes no sense to me, since there will obviously be variations in the vertical concentration, and anyway there are large uncertainties with even the observed PBL depth. Indeed, p12216 says that these uncertainties amount to several 100m. I cannot understand how the proposed methodology is an improvement over the use of excess CO for example.

3. The authors use a low-NOx parameterization of isoprene. This seems like a bad choice for the Mexico plume. Surely the high-NOx schemes for isoprene would have been more suitable?

4. The discussion of isoprene, with the back-of-the-envelope calculation given on p12240, confused me. Doesn't the coarser scale-run of the model cover the domain shown in Fig. 1, cover all the relevant distance scales? Why the discussion of 150km transport?

5. I agree with Ref #1 that when plots are given as an average over all stations, many difficulties (or good performances!) can be hidden. Given that the focus of this paper is on the OA results from just a few stations, I would have preferred to see comparisons of the modeled O3, NO2, etc. for these (T0, T1) stations. The "all-station" plots could be relegated to the supplementary material as background information on overall model performance.

6. I would also have brought the Figures on OH and OOA into the main text - they are directly relevant for the discussions of the T0, T1 data-sets, and it is rather unusual to

have the possibility of checking the OH from a model.

7. Finally, it would help the reader to have a table of emissions for both the fine and course domains, putting the various anthropogenic and biogenic sources in context.

Smaller comments:

The discussion frequently mixes SOA and OOA terms for observations. Although the concepts are similar, the AMS measures OOA.

p12209, line 17. The Dockery et al reference is now 15 years old - find something more recent if the evidence is indeed 'growing'!

p12209, line 23. 'most of which' is water soluble - be more specific, is this 51% or 99% ?

p12210, line 14. Total OA mass can't be measured by AMS; only the fine fraction.

p12211 and elsewhere. The Hallquist review article is 2009, not 2008 for ACPD. Also, this paper is now accepted for publication in ACP. This paper also contain some more recent references for aspects (e.g. aqueous processes) discussed from line 25 onwards.

p12212, re Song et al. (2007). The authors remarks are correct as such, but the Song study only applies to very fresh POA. Any aging will quickly allow partitioning to POA.

p12212, Lines 16. I would also say that OA measurements have suffered from the lack of chemical speciation, or of marker info (14C, etc.)

p12213. The Hildemann et al ref is missing from the reference list

p12213, lines 23 onwards. It would be good to put this small BSOA contribution in the context of results from other cities. Is Mexico city unique, or typical?

p12215. Give a brief indication (in % terms) of the level of agreement of AMS instruments and what "were consistent" means.

p12218, line 20, explain acronym 'TBO'.

p12219, line 16. Here the authors state that most SOA is not very hydrophilic. How is this consistent with the earlier statement that 'most' SOA is water soluble?

p12219, line 23. Why was NO2 chosen as the model for OA deposition? Wesely's scheme has several organic species which would seem more similar in character.

p12221, The model setup described here suggests a nesting ratio of 7:1 was used. The normal recommended procedure for MM5 is 3:1. Why wasn't an intermediate nest used?

p12222, I was amused to see a blog given as a reference, but the website did indeed contain proper data and descriptions. Still, I wonder how long-lived such a reference can be. Is there no other document which can be referred to?

p12223. What about emissions of CO?

p12244 says that the comparison highlights the need for more complex parameterizations for air quality models. I don't see how adding complexity to a system where one doesn't know the basics can improve things?

p12227. The model has some problems with the wind-field. This is likely inevitable, but I wonder how well MM5 captures surface features of the Mexico-city urban area - was any investigation made of the sensitivity to z0 for example?

p12230, line 12. Don't say 'correctly simulated', use 'adequately simulated' or simulated reasonably well. I never expect to see a 'correct' simulation from a model!

p12247, line 4. I would remove the reference to the Hodzic et al. (2009) ACPD paper unless it really makes it to ACPD within the life-cycle of the current manuscript. Who knows what will be accepted for ACPD?

p12248, Conclusions:

I didn't understand conclusion (1). How can a slight underestimate of PBL height give weaker dispersion?

Conclusion (II) is too vague - quantify.

Conclusion (III) is also vague - avoid words like "reasonably reproduced", "no significant bias" ... give the numbers.

Conclusion (V). The modeled BSOA is part of the base-model simulation, so what is that reduces the model overall bias?

Conclusion (XI). Ref #1 has more to say on this, but when saying that "we have identified one of the important missing processes", then let the reader know which one. This is conclusion XI, so quite a few processes have been discussed already.

Table 2. very confusing.... why are so many different parameters given - they are all related. Use consistent parameters! Also, give references for the values used.

Table 4 is very small. Also, the equation for RMSE should have the 1/N outside the root sign.

Fig. 1 - show where the small domain fits into the larger domain.

Figures - general. Why is the solid line used for the model results? This suggests an over-confidence in the model compared to the measurements.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12207, 2009.