

Interactive
Comment

Interactive comment on “Modeling meteorological and chemical effects of secondary organic aerosol during an EUCAARI campaign” by E. Athanasopoulou et al.

Anonymous Referee #2

Received and published: 18 October 2012

This paper presents results of the implementation of a 1-dimensional (C^*) volatility basis set (VBS) scheme into the COSMO-ART coupled radiation/meteorology/chemical transport model run during a period defined by ground measurements during the EUCAARI campaign in May, 2008. The paper includes comparison of the model outputs with AMS data of OA concentrations and a relatively comprehensive set of sensitivity analyses to explore the influence of different model assumptions on simulation outputs (aerosol loading, radiative forcing, etc.) . The work appears to be carefully conducted and I certainly think it will be of interest to ACP’s readership.

Below I identify several issues that I would like to see addressed or clarified before

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the manuscript is published in its final form. I do not anticipate this requiring further modeling, simply clarifications and discussion of some limitations of the approach.

In general, I found some of the writing in the paper quite difficult to follow, often due to poor word choice or improper grammar. I would recommend careful copy editing for clarity and readability before resubmission. This observation is echoed by the other referee, who asks for clarification on some very basic/fundamental points concerning the model implementation.

Major issues:

Like the other reviewer, I found the description of the SOA mechanism (Tables 1 and 2, P21822-21823) to be quite unclear and poorly documented and I have various questions concerning its implementation. I'm assuming the products with a postscript 'P' are the semivolatile products of reaction (e.g. OLTP) with bin-wise yields given in Table 2? The 'k' value given in the manuscript dictates the gas-phase reaction of the semivolatile vapors associated with SOA production, does that k also dictate the reaction rates of the SOA precursors? Finally, the VBS C* distributions in Table 2 are given with no references – from where are these derived?

The authors chose a particular implementation of a VBS modeling scheme for implementation in their model, but there are others possible. The model chosen includes a 1-bin shift in volatility per generation of oxidation and a 7.5% increase in mass. Other modeling studies have tried alternate implementations [Hodzic et al., 2010]. It should be acknowledged that the mechanism is primitive and uncertain (and in fact somewhat arbitrary) and that there may be large sensitivity to choice of parameters.

Related to this, there is much evidence that a simple 1-d VBS cannot represent the multigenerational chemistry that SOA undergoes, as it will continually add mass while actual atmospheric oxidation will form less-volatile products via fragmentation reactions [Donahue et al., 2012; Jimenez et al., 2009; Kroll et al., 2011]. It would be good to include some discussion of the limitations of the 1-d approach and where it could

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potentially be improved.

The discussion of HOA/POA and OOA/SOA are somewhat confused and unclear in the manuscript, and this leads to some erroneous statements. HOA (“Hydrocarbon-like OA”, not Hydrogenated as stated at the top of P21817) and OOA are AMS factors and do not directly correspond to POA and SOA. HOA has been tied to primary combustion emissions (using tracers like CO and BC) and not necessarily to POA as it would be recognized in your model emission inputs. The fact that this model does not treat POA as semivolatile misses the fact that some components often counted as POA in models (SVOCs associated with combustion emissions) can contribute substantially to the OOA mass once they evaporate and oxidize in the atmosphere [Jathar et al., 2011; Murphy and Pandis, 2009; Pye and Seinfeld, 2010]. The fact that you are not treating semivolatile/aging POA is clearly stated, but I think discussions of and distinctions between HOA and POA (e.g. P 21823, L9-11; P21833, L5-7) need to be clarified and this identified as a potential shortcoming. HOA appears to be the fraction of POA from combustion engines and similar sources that is initially in the aerosol phase at ambient conditions, as measured by the AMS, and will likely not age substantially (as stated) – however, SVOC vapors often measured as POA in source testing will age and contribute to OOA.

P21834, L17-27: How confident are you in the parameterization of cloud cover and the feedbacks between aerosols and cloud cover? Are the changes in average and spatially-resolved forcing due to changes in cloud cover actually significant relative to uncertainty in model output? Some discussion of this would be helpful to put these results in context.

Minor Issues

P21822, L2 – ‘saturation concentration’ – this usage doesn’t make sense to me, as this should not be affected by ‘a pre-existing internal mixture of aerosol compounds’. Unless you’re talking about the C^* of a given bin being dictated by the properties of

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the mixture of specific compounds that get lumped in the bin? In any case, this is not clear. . .

P21822, L23 – ‘deprive the model’s capability’ – not clear.

P21823, L26- ‘volatility concentration’ is not a meaningful term, maybe ‘effective saturation concentrations’?

P21824, L13 – ‘large increase in aerosol direct radiative forcing’ – won’t this typically cause a decrease (increase in the negative direction?)

P21824, L14-15 – It hasn’t been made clear at this point why this is an ideal period to model as you haven’t described it. You should qualify this statement with a brief description of the period.

P21824, L25 – OA isn’t ‘supposed to’ do anything, ‘observations suggest that OA reaches a stable oxidation state’

P21828, L6 – ‘Section 4.4’ should read ‘Section 4.3’

P21829, L23-24 – Reading this I expected Fig. 3 will include results from other models. You should clarify that you’re just comparing domain averages here.

P21830, L2-3- ‘differences. . . are not significant’ The difference between PMCAMx and COSMO-ART for domain average OA seems to be about 30%, is this not significant?

P21835, L18-23 – Probably a good place to also mention/discuss the potential for ‘missing’ contributions from the boundary (e.g. the intrusion of African OA observed for May 19-21).

P21836, L1-16 – You make comparisons between the ‘base’ SORGAM model (Scenario 6) output with other model configurations at the different sites, but results are only shown for Cabauw. Why are these simulations not shown (but only summarized in text) for the other sites?

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P21836, L23 – PMCAMx also includes aging of POA (SVOCs), correct?

P21839, L1-2 – How do ‘nonurban sources’ contribute to the model if they are not explicitly included in the model?

P21840, L2- ‘biogenic activities’ should be ‘biogenic precursors’ or ‘biogenic emissions’

P21841, L23 – ‘is now eradicated from COSMO-ART chemistry’ – this is an overstatement. It appears that the addition of this SOA ‘mechanism’ improves predictions of inorganic aerosol concentrations.

Table 5 – I found this table very confusing at first – can this be reformatted? It could also be deleted as most of these values are included in the text.

Figure 7 - It would be interesting to see change in OH expressed in a meaningful unit – perhaps molecules/cc?

All multi-panel figures – it is helpful to have a short label (e.g. indicating the city or date) next to the panel letter to make it easier for the reader to quickly identify.

References

Donahue, N. M., J. H. Kroll, S. N. Pandis, and A. L. Robinson (2012), A two-dimensional volatility basis set – Part 2: Diagnostics of organic-aerosol evolution, *Atmos. Chem. Phys.*, 12(2), 615–634, doi:10.5194/acp-12-615-2012.

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