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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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In this paper the authors couple the VBS approach within the COSMO-ART model to assess the effect on multiple processes, including radiative transfer and partitioning of inorganics. The paper reveals very interesting results, specifically with regards to the importance of process coupling and rate constants, for example. The paper is largely caveated appropriately, and is relevant for ACP readers. However there are a few questions that need addressing prior to publication.

Page 21822, lines 28 onwards: Here the description of the gas/particle mass transfer is given. The reference to using the VBS approach is by lumping the condensates into fixed volatility bins that vary according to local temperature. The description goes on to

state that the same gas-particle transfer method is used. Presumably this is dynamic and does not assume equilibrium partitioning?

In table 1 the oxidation reactions are listed for each precursor. I presume the total mass of these compounds (generated through oxidation) is then used to populate the four VBS volatility bins according to yields defined by table 2? If this is the case, and you have the same mass between the VBS version of SORGAM and the 'detailed' version, could it be that the pure component volatility used within the 'detailed' model are the reason for under predictions of mass? If im not mistaken, this is the only real difference in this approach...the bunching of products into specific volatility bins. Is this correct?

Based on the above comment I would also point out that models such as COSMO-ART then display an inherent flexibility to test the importance of certain processes. Whilst I understand the need to use semi-empirical formulisms for correct predictions of mass for ,say, radiative transfer simulations as you have done, this dosnt remove the valuable insights gained from not getting the mass correct using a bottom up approach. This is perhaps a fundamental discussion to be had elsewhere, but it relates to the questions that should at least be addressed in the preceding paragraph.

Page 21823, line 4: 'During each time step SOA species become less volatile by one order of magnitude'. This specific assumption needs justifying. Are you assuming that the condensed material is oxidized by OH or the vapour phase? Can you really justify the order of magnitude drop in volatility across the entire volatility bins? More detail is required. For example, do the condensing organic compounds effectively build up over-time as their volatility reduces constantly?

Section 3: Page 21825, line 5: 'the differences in OA concentration predictions between the SORGAM (scenario 3) and the VBS approach (base-case).' Just to clarify the point made above, the SORGAM model numerics are equal, the range of condensates, their vapour pressures and reaction rates are not?

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Page 21825, line 28: An enthalpy of vaporisation of 30 kJ/mol is low. What is the sensitivity to this? It would be useful to check more recent empirical relation by Epstein et al (2009): Epstein, S. A., Riipinen, I., and Donahue, N. M.: A Semiempirical Correlation between Enthalpy of Vaporization and Saturation Concentration for Organic Aerosol, Environ. Sci. Technol., 44, 743–748, doi:10.1021/es902497z, 2009.

Conclusions: 'The inability of SORGAM to treat chemical oxidation of organic matter..' This needs stating earlier on in the paper as it may address questions posed above. 35% isn't terrible, is this averaged? I would expect a difference of 35% to be rectifiable with a revisit of composition dependent parameters. Has this been performed? Mechanistic models can underestimate by at least an order of magnitude due to insufficient carbon flux in the system.

Minor comments: Table 1: What non highlighted species does this refer to? This isn't clear from just the table caption.

Abstract: Last sentence: please consider revisiting the grammar. In the line 'while the condensation upon pre-existing, SOA-rich particles..' it should be made explicit what condensation you are referring to.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 21815, 2012.