

Interactive comment on “A global perspective on aerosol from low-volatility organic compounds” by H. O. T. Pye and J. H. Seinfeld

Anonymous Referee #1

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This paper uses a global chemical transport model to investigate the effects of partitioning and aging of low-volatility organic compounds on predicted organic aerosol loadings. This is the first study of this type done on a global scale. The paper treats POA using absorptive partitioning theory and a two product approach. Semivolatile vapors are allowed to react to generate SOA. The model also treats SOA production from oxidization of IVOCs. Given the significant uncertainties the paper takes a rational approach to scaling emission inventories and treatment of chemistry. Sensitivity studies were also performed to identify important uncertainties. Results are compared with those of the traditional model (non-volatile POA) and observations. The base version of the revised model predicts lower surface OC concentrations. Increasing the SVOC emissions improves model performance compared to measured OC levels. The paper is well written and addresses a topic of interest to the readers of ACP.

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I recommend the paper be published after the authors address the following comments

Wintertime performance – The base version of the revised model underpredicts US wintertime OC levels. The paper investigated a number of sensitivities, but one that they did not consider was enthalpy of vaporization. The model uses a value of 42 kJ/mol for all organics based on work of Offenberger work to support it, but a value this small is hard to physically justify (see, e.g., Epstein et al. EST 2009). Presumably the wintertime model predictions are sensitive to this value and doubling it into the physically realistic range might substantially improve model performance in the wintertime.

The paper discusses that doubling SVOC emissions essentially closes the wintertime model gap. How does that change affect the summertime performance?

Model measurement comparisons – I was disappointed that the paper did not use naphthalene as an intermediate. For anthropogenic sources they are scaled with benzene. Is this correct?

I was also confused in the derivation of the beta parameter (equation 9). It seems like you are linking the IVOC emissions to the NVPOA emissions? If one repeats this sort of analysis with a phenol or other IVOCs in the Schauer emission profile how variable are the estimated IVOC emissions?

The Shrivastava wood smoke volatility distribution is not well constrained at atmospherically relevant concentrations. This issue was raised in section 4.2. It seems that details of this distribution may be important for the wintertime predictions. For example, assigning the lower volatility component of shrivastava a C^* of 10 as opposed to 20 $\mu\text{g}/\text{m}^3$ (at 298 K) may not alter the fit over the range of the source test data but could have a significant impact on model performance?

Is the fact that anthropogenics account for half of the IVOC inventory an present more detailed model measurement comparisons. The only comparisons were for the winter absolute OC levels in the US (Fig. 7). I understand that the authors focused their

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model evaluation on the winter period to reduce uncertainties associated with biogenic SOA, but more summertime comparisons would also be informative. Comparisons beyond absolute OC levels might be useful as well. For example, Shrivastava et al. (JGR 2008) used OOA/HOA ratios. Fossil versus contemporary carbon seems to be another. Section 3.5 discusses these data but does not quantitatively compare the model predictions to the data.

Scaling Schauer et al. emissions data (page 4088). The paper uses a scaling factor of 0.27 to scale up POA inventories for gas phase SVOC. It appears that this is based only on the speciated SVOC measurements? Is that correct? The paper states that this is a conservative lower bound estimate. What fraction of the total SVOC vapor emissions were speciated? Is there any reason to think that this fraction might be biased low? Can the SVOC UCM be used to bound the range of missing SVOC?

I was confused on how the IVOC emissions were generated. The paper states that IVOC emissions are distributed like naphthalene (page 4091). However, my impression from reading the paper was that for biomass burning the emissions are scaled with artifact of how it was constructed (i.e. using naphthalenes)? My concern is that the argument in section 3.5 that "These observations lend support to the hypothesis that SVOCs contribute significantly to ambient OA and that scaling up SVOC emissions may be more justified than scaling up IVOC emissions" seems circular.

At a number of points the paper mentions the need for additional constraints, but only briefly discusses those in the final paragraph of the conclusion section. Given the challenges associated with simulating OA, it would be good to expand if the paper could be specify some directions for the community could head to help resolve these issues.

How was deposition of IVOC gases treated, i.e. hydrophobic or hydrophilic?

Naphthalene may be a reasonable IVOC for biomass smoke, but it is less clear for urban/mobile source dominated emissions which may be dominated alkanes.

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