**Atmospheric
Chemistry
and Physics
Discussions**

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 its 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. Sensitivity studies are performed to identify important uncertainties. Results are compared to predictions of the traditional model (non-volatile POA) and observations. The surface OC concentrations predicted by the base version of the revised model are substantially lower than observations. Increasing SVOC emissions improves model predictions of measured OC levels.

The paper is well written and addresses a topic of interest to the readers of ACP. 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 the work of Offenberger, but a value this small is hard to physically justify (see, e.g., Epstein et al. EST 2009). The wintertime model predictions should be sensitive to this value and doubling it into the more physically realistic range might substantially improve model performance.

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

Model measurement comparisons – I was disappointed that the paper did not present more extensive model-measurement comparisons. The only comparisons were for the absolute OC levels in the US during the wintertime (Fig. 7). I understand that the authors focused their model evaluation on the winter period to reduce uncertainties associated with biogenic SOA, but more summertime comparisons would also be informative. For example, does a large model measurement gap also exist for total OC in the summertime? Furthermore, more extensive comparisons that consider parameters in addition to absolute OC levels would be useful too. For example, Shrivastava et al. (JGR 2008) compared their model predictions to OOA/HOA ratios inferred from measured AMS data. Fossil versus contemporary carbon would be another parameter to consider. Section 3.5 of the paper discusses C14 data but the discussion seemed qualitative.

Using Schauer et al. emissions data to scale inventories (page 4088). The paper uses a scaling factor of 0.27 to scale up POA inventories to account for gas phase SVOCs. It appears that this value is based only on the speciated wood smoke SVOC measurements of Schauer? Is that correct? How representative is Schauer et al. fire

place burning profile to the sources that dominate global sources of POA? The paper states that this is a conservative lower bound estimate. What fraction of the total SVOC vapor emissions were speciated in the Schauer study? Is there any reason to think that this estimate of missing SVOC vapor emissions 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 was that for biomass burning the emissions are scaled with CO, using naphthalene as an intermediate. For anthropogenic sources they are scaled with benzene. Is this correct?

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

The Shrivastava et al. wood smoke volatility distribution is not well constrained at atmospherically relevant concentrations. This issue was raised in section 4.2. For example, assigning the lower volatility component of the Shrivastava two product fit a C^* of 10 as opposed to 20 ug /m3 (at 298 K) likely will 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 artifact of how the inventory was constructed (i.e. using napthalenes)? I am concerned 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 information to constrain the model, but only briefly discusses what those might be in the final paragraph

of the conclusion section. Given the challenges associated with simulating OA and the additional parameters introduced by this new approach, it would be good if the paper could discuss in more detail some directions that the community could head to help resolve these issues. For example, what types of ambient data (beyond absolute OC concentrations) could be used to help assess the performance of different models?

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

The authors argue that napthalene is a reasonable IVOC surrogate for biomass smoke, but it is less clear that is a good surrogate for mobile source dominated urban emissions which may be dominated alkanes.