

“Secondary organic aerosol formation during June 2010 in central Europe” by Langmann et al. This study presents a modeling case comparing the thermodynamic-kinetic to the pure kinetic approaches and compares predictions to measurements at a site in France in June 2010. First of all, I concur with the comments from the first reviewer. While the study is useful, the manuscript has general lack of clarity. I recommend major revisions with careful accounting of my several detailed comments below before the manuscript can be considered for publication.

1. Abstract: I disagree with the last sentence based on results shown in the manuscript. I do not see a huge improvement in modeled SOC aerosol mass concentration with the thermodynamic-kinetic approach compared to the pure thermodynamic approach. Figure 7 shows small differences in number concentrations of accumulation mode particles for aged air masses. Please also see my comment number 10 below. In addition, any comparison for SOC mass should include LV-SOG in both approaches. Including LV-SOG in just thermodynamic-kinetic approach, but not in the pure thermodynamic approach is not a consistent comparison, since LV-SOG is formed by aging of semi-volatile organic vapors.
2. P 26764 Lines 1-5: Prior to Jimenez et al. 2009, the aging of organic vapors producing less volatile organic vapors was implemented in 3D model by Robinson et al. [2007] and Shrivastava et al. [2008]. Please include reference to these studies too. In addition, Jimenez et al. 2009 also demonstrated the concept of fragmentation reactions which may lead to increase in volatility of organic vapors. Recently, Shrivastava et al. [2013] demonstrated that including fragmentation reactions in a 3D model improved model-measurement agreement compared to mechanisms that neglect fragmentation reactions. The one-way decrease in volatility of organic vapors is less realistic based on several studies [e.g., *Cappa and Wilson, 2012; Chacon-Madrid and Donahue, 2011; Chacon-Madrid et al., 2010; Kroll et al., 2011; Lambe et al., 2012*]. The authors should at least acknowledge the limitations of ignoring fragmentation reactions. In the study of Shrivastava et al. [2013], the role of fragmentation (considered only in gas-phase) was chiefly to reduce the amount of condensable organic gas precursors. In the consideration of threshold values on page 26768 (Lines 1-5), it should be further discussed that fragmentation also reduces the amount of condensable organic vapors. I suggest that the authors cite the relevant recent studies (e.g. Shrivastava et al. [2013]) in addition to Jimenez et al. [2009].
3. Another major point which the authors ignore is the importance of oligomerization reactions. Shrivastava et al. [2013] have discussed how rapid particle phase oligomerization reactions could transform semi-volatile organics to low volatility compounds, and showed that this transformation causes large spatial and temporal distributions in predicted SOA loadings. Given the potentially large impact, some discussions are needed as to how would oligomerization reactions influence the modeling approach proposed by the authors.

4. P 26768 L14-16: Treating LV-SOG nucleation without its condensation to H₂SO₄ nuclei is clearly unrealistic. This could be a pedagogical case, however, the authors should consider adding the H₂SO₄+SOG nucleation similar to Metzger et al. for completeness. In addition, the authors should better describe their replies to the first reviewer comments clarifying how they treated nucleation of LV-SOG in the manuscript. On my first read of the author's replies, I thought they were considering 2 different nucleation pathways which were mutually exclusive. On reading the manuscript I realized this was not the case. Their nucleation pathway includes H₂SO₄+LV-SOG+H₂O, but they neglect interactions between LV-SOG and H₂SO₄.
5. P 26772 last paragraph: The description is very confusing. Also the caption to Figure 6 says Model simulations correspond to those shown as light blue line in Figure 4. Do all model simulations shown in Figure 6 correspond to the light blue line in Figure 4? But line blue line in Figure 4 corresponds to 1% aging. Figure 6 shows both 1% and 10% aging. I also do not understand how light blue line for 1% aging shown in Figure 6a generates more semivolatile SOA than 10% aging (red line). The last paragraph on page 26772 says the semi-volatile SOA is sum of modeled MV-SOC and SV-SOC. Does this mean that semi-volatile SOA is lower in the 10% aging case (red line in Figure 6a) because it is moved to the low volatility (shown in Figure 6b)? If this is the case, please explain this better.
6. Since the authors describe SOC and OC results please indicate what factors were used to convert from OM to OC? Most models predict OM rather than OC.
7. Table 1: If the authors are not using the saturation vapor pressures to describe MV-SOC and SV-SOC what is the utility of Henry's law coefficient shown in the table? Is it used only in their dry and wet scavenging parameterizations? Please explain.
8. P26774 Lines 10-15: Why do fresh air masses show more particles in the Aitken mode using the pure thermodynamic approach? The curvature effects should cause less particle growth of nuclei to the Aitken mode in this approach compared to the thermodynamic-kinetic approach.
9. Why are the differences in the pure thermodynamic and the thermodynamic-kinetic approach much so small for aged air masses? The authors suggest that a faster growth occurs in the thermodynamic-kinetic approach during aging. Would these differences increase if the aging rate from SV-SOC to LV-SOC were increased?
10. Lines 20-22: I find this statement very less useful: "Generally, the thermodynamic-kinetic approach can more realistically reproduce observed data than pure thermodynamic-kinetic approach in particular in fresh air-masses". The model resolution of most global models is even coarser than the 0.5 degrees used by authors. This implies that air masses are already aged in these coarse grid models. In that case the author's results imply no large improvement in predicted distribution of CCN relevant accumulation mode particle numbers by using the

thermodynamic-kinetic as compared to the pure equilibrium approach. Am I missing something here? Please explain.

11. P26775 first few lines: Why is nucleation of increasing importance with increasing height in the thermodynamic-kinetic approach? My understanding is this approach is based on LV-SOG which is nearly insensitive to decrease in temperature with altitude that favors condensation of semi-volatile organics. Please elaborate.
12. Going back to first few lines on page 26765: The authors say that different to Yu et al. (2011), they consider the contribution of anthropogenic SOC to SOC aerosol formation. But in their sensitivity tests (e.g. in Figure 4) they only increase biogenic VOC emissions by a factor of 5. What about the uncertainty due to anthropogenic SOC emissions? Also in the conclusions please comment on the relative importance of anthropogenic vs. biogenic SOA in their study.

References

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