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Geoscientific Model Development Discussions

Interactive comment on "Oligomer formation in the troposphere: from experimental knowledge to 3-D modeling" *by* V. Lemaire et al.

Anonymous Referee #3

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General comments:

I read the manuscript with great interest. The paper addresses an important subject, the formation of biogenic secondary organic aerosol (SOA) by oligomerization. Laboratory studies have shown that this process could be a substantial formation pathway for aqueous condensed species. The authors compare and discuss two different modeling approaches, one with a first order kinetic process description and the other one by a pH-dependent equilibrium parameterization. Both approaches are implemented and applied in the atmospheric chemistry transport model CHIMERE for a two-month period in summer 2006. Unfortunately, no in situ measurements of oligomers are available to evaluate the approaches or enable further improvements. However, it should be possible to analyze the impact of the two approaches on the concentrations of SOA or

the total organic matter in comparison with measurements. In the study, the influence and sensitivity of several model parameters are investigated for different conditions. The results show a strong dependency on the selected approach. The irreversible first order kinetic description leads to the oligomerization of about 50% of the biogenic SOA mass. The pH-dependent equilibrium approach shows a broader range of impacts, which strongly depends on the surrounding conditions and the possibility for the process to be reversible. The paper contributes to a better understanding in modeling organic aerosol in the atmosphere.

I agree with the two other referees and recommend the paper for publication after revision, which takes into account the specific comments given in the reviews.

Specific comments:

1) The spatial and temporal variability of the biogenic precursor emissions is essential for the interpretation of the SOA concentration fields and, hence, for the discussion of the approaches. Therefore, the authors should give a short description of the applied MEGAN scheme and the used land use data set. Furthermore, it is recommended to include maps of the main biogenic emissions.

2) The authors point out that the particle pH is restricted to a range of values between 2 and 6. The lower limit is set to 2 due to numerical reasons. What are the numerical problems? How often and where was the lower pH limit reached? Can you comment this, please!

3) As mentioned above the authors should try to compare the modeled concentrations of SOA (or total organic PM) with measurements?

4) The figures g and h in Fig. 2 are missing.

Interactive comment on Geosci. Model Dev. Discuss., 8, 9229, 2015.