

**Review of paper “Oligomer formation in the troposphere: from experimental knowledge to 3-D modeling” by Lemaire et al.**

This study investigates the formation of oligomers from biogenic secondary organic aerosol (SOA) using two different approaches in the chemical transport model, CHIMERE; one with a first order kinetic process and another with a pH-dependent parameterization. Formation of SOA is still the focus of recent atmospheric research and oligomerization processes are not well known yet. This work therefore would contribute to the efforts in modeling organic aerosols in the atmosphere. There are however, some important issues to be addressed. I would recommend it to be published after some revision.

**Main comments:**

Authors applied the model with both parameterization to a domain covering western Europe during a summer period and compared the oligomerized SOA in both cases. There is no discussion about the model performance for the studied period. They should add some model performance evaluation for this specific application of CHIMERE. In this way it would be possible to see how the two approaches affect the final SOA or at least OA concentrations with respect to measurements.

More discussion is needed about the differences between the approach used in this work and oligomer formation within the VBS formulation as described by Trump and Donahue (2014).

Meteorological model: Authors should mention which MM5 version was used. Horizontal resolution of MM5 was given as 54 km (page 9236, lines 3-7). The resolutions of CHIMERE domains on the other hand, were 0.5 deg x 0.5 deg and 0.23 deg x 0.20 deg. Was MM5 used with another projection, not lat-lon? Was MM5 horizontal resolution for the mother domain same as the CHIMERE mother domain? Authors must describe how they generated meteorological fields for the nested domain with a resolution of 0.23 deg x 0.20 deg.

Emissions: Resolution of anthropogenic emissions is very coarse (0.5 deg.). Do biogenic emissions have the same coarse resolution? Maps showing biogenic emissions (isoprene, monoterpenes, sesquiterpenes separately) during the modeling period should be shown in order to understand the locations of BSOA precursor emissions. Fig. 8b (SOA from monoterpenes) and Fig. 9 (right) (SOA from monoterpenes and isoprene) suggest that isoprene and monoterpene emissions have the same distribution (SOA concentration increases at the same locations due to addition of isoprene emissions). Is that true? Authors should discuss the influence of horizontal resolution of biogenic emissions on the distribution of biogenic SOA produced from isoprene and monoterpenes.

The number of biogenic precursors was given as 5 (page 9234, line 25), but the following list has 6 names (isoprene, alpha-pinene, beta-pinene, limonene, ocimene and humulene). The focus of the study is on monoterpenes.

However, humulene is a sesquiterpene. It is not very clear whether BiBmP refers to the sesquiterpene surrogate. Does Fig. 9 include the sesquiterpene BSOA? As it is given in the paper, BSOA from monoterpenes means from alpha-pinene, beta-pinene, limonene and ocimene. What about BSOA from humulene (a sesquiterpene)? Authors should explain how they treated sesquiterpenes in their formulations.

The particle size range was given as 10-40 micrometer (page 9234, line 16). If this is correct, then it means only the coarse fraction was modeled. Authors should explain it and give the particle size range the results refer to.

### **More specific comments:**

Fig. 2: Two figures g and h are missing.

Fig 3: Upper panel: Two red points for BIA0D on the blue line have to be described.

Fig. 4 When is the “daily maxima”? Highest oligomers are over the Adriatic Sea. In Fig. 6 d however, it looks very different at 05 UTC.

Fig 6: The choice of 05 UTC to analyze precursor and oligomer concentrations has to be explained. Most of the biogenic emissions are higher around noon and boundary layer develops during late morning hours.

High precursor concentration and deliquescent aerosol with low pH favour oligomerization. Precursor concentrations are very low over the Mediterranean but oligomers are high - just because of low pH? Is such a low pH (around 1, Fig. 6 b) reasonable? This needs to be discussed more clearly.

In Fig 6b, pH seems to be 3.5 around the Benelux area which has the highest NH<sub>3</sub> emissions in Europe and therefore acidity is expected to be neutralized. Is it reasonable to have such a low pH in that area?

Fig. 7: Location of the grid cell for the time series should be given

Fig 8 d and e: Relative concentrations show highest contribution of oligomers over the seas. Authors might consider to show absolute contributions.

Page 9236, lines 8-11: Authors must describe how the boundary conditions for the European domain were generated.

Page 9240, line 6: replace “.. could not considered” with “.. could not be considered”

Page 9242, eq 3: H<sub>i</sub> instead of H<sup>i</sup> as in line 18

Page 9242, lines 23-25: indicate the Eq. 4 at the end of the sentence.

Page 9242, line 26: (set .... Robinson et al. (2007))

Page 9246, line 9: ..smoother gradients (Fig. 5)

Page 9246, line 11: ..previously. (remove (Fig 5))