

## Review of “Description and Evaluation of a Secondary Organic Aerosol and New Particle Formation Scheme within TM5-MP v1.1”

In this study the authors present and evaluate their implementation for new particle and secondary organic aerosol formation from semi and extremely low volatility monoterpene and isoprene oxidation products. The new implementations expand the functionality of the M7 aerosol module within a global chemistry transport model (TM5-MP). The authors explain the new functionalities in detail, including the updated biogenic emission inventory, the chemical conversion to SVOC and ELVOC, the new particle formation and SOA formation approaches. After introducing the changes, the authors compare the extended M7 scheme with an older version, by comparison of the simulations themselves, but also including observations. Observational datasets to evaluate the global model are surface organic mass concentrations (IMPROVE and EMEP networks), number concentrations on the surface (EBAS), satellite and ground based AOD products (MODIS, AATSR, AERONET).

The chemical conversion from isoprene and monoterpenes to SVOC and ELVOC in form of lumped species is not novel, but the extension of M7 functionalities can be applied for a number of global chemistry models and thus makes a fair contribution to the aerosol model development in general. The manuscript is generally well structured and I recommend that the manuscript should be accepted for publication after major revisions where the authors carefully addressed my comments.

### **General comments:**

The TM5-MP model description lacks the spatio-temporal model resolution used for this study. In the evaluation and conclusions, the time resolution becomes clear. In section 2.4 the simulation period, and later in the text also the horizontal resolution, are mentioned. For the reader it would be easier to get the full information on spatial and temporal resolution, such as the time period for the simulations either right at the beginning in Section 2.1 or in Section 2.4 in a more coherent way.

In Section 2.3 the earlier version of TM5 is described and it is stated that it uses biogenic monoterpene emissions from MEGAN v1 and does not include isoprene emissions for SOA formation. In contrast, the new version calculates SOA from isoprene and monoterpenes, based on MEGAN v2 inventories. In section 2.4 the authors describe the simulations to be compared, one simulation using the older version “OLDSOA” and a second simulation for the same time period using the new formation schemes “NEWSOA”. The difference between these two simulation configurations are not only the novel SOA and new particle formation schemes, but also the changed biogenic emissions plus, in the old scheme only monoterpene-derived SOA was included. For disentangling the contributions from the updated biogenic emissions from the contributions by the extended SOA treatment, I would suggest to add sensitivity simulations. I wonder how does the NEWSOA scheme perform compared to OLDSOA while using the same MEGAN v1 emissions? Also, how do NEWSOA and OLDSOA compare when both using MEGAN v2 emissions, even if OLDSOA does not form isoprene-derived SOA? I would not expect the difference in emissions shown in Figure 2 apply in a linear way to SOA formation. In Section 3.1 the last paragraph points into the direction. Additional sensitivity simulations with both emission inventory versions would help to understand what

difference is caused by the changed emissions and what difference is related to the novel SOA formation pathways.

In section 2.3.2 the production of ELVOC and SVOC is described. In the introduction, the authors do mention low volatility compounds (LVOC), but do not explicitly state here why they assume two products of the isoprene and monoterpene reactions (ELVOC and SVOC) and leave out the possible LVOC products. It would be helpful for the reader to know the reason for this decision.

Table 2 nicely shows the SOA budget together with some literature values. In this study, the authors differentiate between monoterpene-derived SOA and isoprene-derived SOA, a distinction which cannot be found in the literature cited in Table 2. A study focusing on isoprene derived SOA could add some more information on how to interpret the values for isoprene-derived SOA. Please consider comparing to the global isoprene-derived SOA values in Table 4 given in Stadtler et al. 2018 (<https://doi.org/10.5194/gmd-11-3235-2018>). I did not find a comparable study for monoterpene-derived SOA on a global scale, but it might also exist.

In Section 3.3 the authors start to compare the OLD SOA and NEWSOA simulations to observed particle number concentrations (Figure 8). Although this kind of scatter plot is a common evaluation method, the comparison shown here does not include any error or uncertainty indication, either for the observations, nor for the statistical error caused by comparing grid cells to point measurements. Swall et al. 2009 (<https://doi.org/10.1016/j.atmosenv.2008.10.057>) describe and discuss the incommensurability problem arising from comparison of point measurements to grid cell values. According to Swall et al. even if a perfectly performing model and measurements without observational error, the scatterplot will not show modeled and observed values on a one-to-one line. The same holds true for Figure 14 and comparison to AERONET stations, Figure 10 in comparison to IMPROVE stations and Figure 11 in comparison to EMEP stations. The authors should consider including information on uncertainty derived from the comparison approach and, if available, the observational errors. A model error would be nice-to-have, but I dare not to ask for that.

In whole Section 3 the description of the results and comparison is blended with the discussion of the model errors and limitations. The authors should consider editing the subsections in Section 3 such that the description of the results is separated from the discussion and interpretation of the model strengths and weaknesses. A clear strength of this study is the evaluation with a variety of observations, but it makes the manuscript difficult to read if the interpretation and discussion is mixed into the comparisons to the old model and observations. The authors should consider to collect all the discussion points of NEWSOA schemes' limitations and advantages and to write them into a single "Discussion" Section in a concise way.

Out of curiosity, why was it not possible to include isoprene and monoterpene oxidation by NO<sub>3</sub> in this study? As you mention, it limits the current model performance, but should be considered in a future study. I wonder what reason was prohibiting to implement this additional oxidation reaction in the NEWSOA scheme.

Technical corrections:

In the abstract line 11 – 14 it is written “Compared to the old scheme, the new scheme is increasing the number concentrations across the observation stations while still underestimating the observations. The total aerosol mass concentrations in the US show a much better seasonal cycle and removal of a clear overestimation of concentrations.”, which is confusing the reader. First the authors speak of “still underestimating” but the next sentence reads “removal of overestimation”. Please make it clearer here.

In Line 34 there is some number in the word “vegetation” “vegetat589527ion”.

Line 431 and 432: Two sentences in a row starting with “However”.

In Figure 13 the plot labels indicate [NEWSOA-OLDSOA], but according to the caption of Figure 13 the annual mean difference of NEWSOA and MODIS (a), NEWSOA and AATSR (b) are shown. Please change the label of the plots.

Line 504 it says “However, compared to OLDSOA the absolute bias across all stations increases in NEWSOA by 0.007 to a low bias of 0.003”, should the verb be “decreases”?