

Interactive comment on “An evaluation of global organic aerosol schemes using airborne observations” by Sidhant J. Pai et al.

Anonymous Referee #2

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The paper evaluates two organic aerosol modeling schemes on a global scale. The use of the results of a series of campaigns in different parts of the world is a major strength of the paper. The authors do attempt to gain insights about the ability of the model to properly simulate different processes and do manage to reach a number of useful conclusions. However, the paper has a number of serious weaknesses that need to be addressed before it can be accepted for publication.

Use of ground measurements. It is not clear why the authors restrict their model evaluation to airborne measurements when there are also ground measurements available in the same areas (US, Europe) for the simulated periods. This is especially problematic because they conclude that the simple modeling scheme is especially attractive for health studies. They also suggest that based on the airborne measurements both

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schemes appear to overpredict OA in the lower troposphere. This comparison (at least with the IMPROVE and EUCAARI measurements) should be added to the revised paper.

Treatment of POA. It is not clear how the POA emissions and the POA atmospheric chemistry are treated in the two models. Some major issues:

(a) Why is the POA emission rate (based on Table 2) in the simple model 21.8 Tg/yr and in the complex model 55.4 Tg/yr? Based on the paper there is a 27 percent increase of the emissions while these numbers suggest a 150 percent increase.

(b) What is the assumed saturation concentration of the POA in the complex model? What is its enthalpy of evaporation?

Anthropogenic SOA. How is this simulated in the complex model? Why is so much less than in the simple model? The production of SOA from IVOCs in the complex model is not described well. There is little information about how the IVOC emissions (shown in Table 1) have been estimated and the corresponding yields used. The sentence regarding the use of naphthalene as a proxy (lines 169-171) does not clarify this issue.

POA lifetime: Line 306. This is quite difficult to understand and it appears to be counterintuitive. The information about the POA treatment is limited. For example, the saturation concentrations of the two products used (lines 152-153) are not given. Also the conversion of the EPOG to the OPOG is not explained and the corresponding parameters for the reactions and volatilities are missing. Finally, there is no information about the removal of the corresponding organic vapors (e.g., assumed Henry's law coefficients). The authors should explain physically how a 27 percent increase in emissions (with some of them in the gas phase) leads to a more than 50 percent increase of the burden of particulate POA.

Lifetimes of OA components: A wide range of global average lifetimes (from 3 to

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11.5 days) is predicted. This is rather surprising for particulate matter components that should have similar size distributions. A discussion of how removal is parameterized and an explanation of these unexpected differences among different components are needed.

Uncertainty of measurements. The authors site a 34-38 percent uncertainty of the measurements that they use. However, this is not taken in the evaluation of the two models. Could the model errors and biases be due to the corresponding measurement errors? Could most of the R^2 difference from unity just be due to these errors? These issues should be addressed and taken into account in the corresponding conclusions. For example, if one of the model was perfect but the measurements had a 35 percent uncertainty what would be the R^2 between measurements and predictions?

Averaging times. The evaluation of any model does depend on the timescale investigated. This timescale appears to be 10 min. If this is the case, it should be mentioned in all graphs, tables but also in the text. An analysis at different timescales (say 1 hr and campaign average) could provide some additional insights. This would allow the authors to include all their measurements (avoid excluding the highest values).

Model performance statistics. The discussion is based on R^2 and normalized mean bias (NMB). I think that the errors (both absolute and normalized) should be included in the analysis. For example, they appear to be quite useful in the discussion related to Figure 12.

Role of enthalpy of evaporation. Given that a lot of the measurements were collected at relatively low temperatures the predicted partitioning of the semi-volatile OA components will depend on the assumed enthalpy of evaporation. It is not clear which values are used in the present work. The sensitivity of the conclusions of the paper to these assumed values should be discussed.

Underestimation of OA. Is it still appropriate to say (abstract and introduction) that models significantly underestimate observed OA concentrations in the troposphere?

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The most recent studies including both models here tend to overestimate OA levels. Some additional discussion of the recent OA modeling work is needed here.

Sources of bias. The complex model tends to overestimate OA levels. It is not entirely clear which attributes of this model (at least compared to the simple one) are contributing to this tendency for overestimation. Is it the isoprene SOA that is included in different ways in the two models? Something else? Some additional discussion is needed.

Sesquiterpenes. It is not clear how the models treats SOA from sesquiterpenes. In Figure 1 this pathway is missing. In line 168 they are mentioned as a source of SOA. The yields used are surprisingly low at least for the simple model: they are equal to those of the monoterpenes, despite the much larger size of these molecules. The yields used in the complex model are not clear. The same applies to their importance as SOA precursors in these simulations.

Organonitrates. The treatment of organonitrates requires some additional explanation given that they are implicitly included in the SOA formed in the parameterizations of Pye et al. (2010). Do they represent additional SOA mass or do they describe a subset of the SOA predicted by the parameterizations used? Could this be contributing to the overestimations? Finally, there has been considerable work quantifying the organonitrate PM levels in Europe (Kiendler-Scharr et al., GRL, 2016) during the simulated EURAARI period that is not used here.

Model performance in Europe. The parameterizations used in both models have been based, to a large extent, on US field measurements. The relatively poor performance of both models in Europe during EUCAARI requires additional attention. The comparison with the ground sites would be quite helpful here. What is the source of the high predicted concentrations which are inconsistent with the measurements?

Model behavior during the winter. This is significant weakness of the paper given that similar modeling exercises suggest that the models have significant problems in

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the winter. The authors can use ground measurements to close this gap. They could also change the title of their paper to specify that this evaluation is only for the summer and spring.

Evaluation for CO and other gases. Given the importance of CO for the parameterization used in the simple model, an evaluation of the ability of the performance to reproduce measured CO levels is needed. A similar evaluation for isoprene (in the appropriate regions) would be helpful.

Characterization of the two models. I found the characterization of the two models as simple and complex rather misleading. Their differences are mainly in the processes that they simulate, the emissions that they use, and the parameters that they use. They are by no means a simplified version of the other. While I understand the need for names, I also think that the paper should include a table with the main differences in the two simulations. These differences should be the emissions (Table 1 is confusing right now), the processes simulated in one and not the other, the differences in the simulation of the same process (e.g., assumed effective yields for a given OA level), and the different parameters used. If the authors can attribute the differences in predictions of the two models to simplicity or complexity, this should be done carefully and should be properly supported.

Some additional points

Lines 42-43. This sentence is confusing.

Lines 73-74. The formation of SOA in some of these models was by no means instantaneous. The VOC precursors were required to react first.

Lines 131-132. The names of the species in the code are not needed here.

Lines 141-143. The use of the term yield is rather confusing. Does this mean for example for fires that the assumed SOA emission is 1.3 percent of the emitted CO? Something else? In Figure 1 it appears that CO is producing SOA making this even

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more confusing.

Lines 142-143. Please explain the biogenic sources discussed here. I am assuming that this means isoprene and the monoterpenes. Referring to Figure 1 could help.

Lines 147-150. Please explain what do these tracers represent and how their use allows the independent adjustment of model parameters.

Table 2. I find this table confusing. For example, the four POA emission rates do not add up to the total. It also mixes actual emission inputs with estimates from steady-state calculations. May be the information could be clearer if it was replaced by two tables.

Line 202: The 54.3 Tg/yr of POA emissions in the simple model cannot be found Table 2.

Figure S1 should probably be moved in the main paper. If needed it can replace of the figures with the vertical profiles.

The conclusion regarding the overestimation of the aqueous uptake of isoprene should be added to the abstract.

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