Response to Anonymous Referee 1

This manuscript describes changes made to the SORGAM aerosol model, and demonstrates improvements in the ability of this modified module to simulate observe organic aerosol concentrations as measured during the IMPACT campaign in the Netherlands. The suggestions about which parameters are key targets for improvement are likely to be of interest to the readership of ACP, and as such this manuscript is appropriate for publication in this journal, but changes are suggested to improve its readability and usefulness to the atmospheric modeling community.

We would like to thank the reviewers for their comments and questions which have helped us to improve the manuscript. The reviewer's comments are given below together with our responses and changes made to the manuscript.

General comments:

1. This paper needs editing for language. There are numerous grammatical/wording errors throughout. Please find a native English-speaking editor to help you improve the readability of the manuscript. I don't include any copy-editing comments here.

Response:

This paper has now been improved by a native English-speaking editor.

2. In the abstract, model improvements are all described solely in terms of increasing SOA yields to better match field measurements from IMPACT. However, in the text it becomes clear that you are also describing improvements to the description of chamber data, where the previous models OVERESTIMATED rather than underestimated SOA concentration. First, I suggest including this in the abstract/intro, so that the reader is prepared when it comes up on p. 5974 line 8 in the description of a figure. Also suggest including more discussion in the introduction of this - why would field data be underestimated and chamber data overestimated by the same model? Furthermore, it's unclear to me how your new model would reduce SOA under any circumstances –in Figure 7, it looks like every change you made added SOA. Which change is it that reduced SOA in the chamber simulations?

Response:

We revised the abstract as suggested to include the chamber experiments as well.

"... and the updated SORGAM with temperature dependence functions was evaluated

with the chamber data. Good performance was found with the normalized mean error of less than 30%."

And the answers to reviewer's concerns are provided below.

(1) Comment: Why the field measurement of SOA was underestimated while chamber data was overestimated by the original SORGAM model?

Response: There could be a few reasons for the underestimation of the field observation of SOA by SORGAM. For example, some aerosol formation channels are missing in the SORGAM model and the production rates of some precursors are probably underestimated. In this study, we found that overestimation of the chamber data can be corrected by introducing temperature dependent functions of mass stoichiometric coefficients (α) and partitioning coefficients (K) in the two-product model. Therefore, the overestimation of the chamber observed SOA concentrations by SORGAM could be partly due to the overestimation of yield (calculated by α and K) in the temperature regime of chamber experiments.

(2) Comment: Why the simulated SOA concentration by the new model (SORGAM-T) is reduced in the chamber conditions while increased in field conditions compared to the original one (SORGAM)?

Response: This is because of the different conditions between the chamber and field experiments examined herein. These conditions mainly include temperature (T), relative humidity (RH) and the seed aerosol concentrations (M_0).

Of the new model (SORGAM-T), the major improvements compared to the original one are shown in m-xylene and α-pinene as shown in Figure 4. For m-xylene, as investigated in Figure 3, when temperature was low (<293 K), the simulated SOA concentration of chamber experiments by SORGAM-T were higher than those by SORGAM. The simulations by the two models at about 293 K were similar. However, when the temperature was high (~298 K), the simulated SOA concentration by SORGAM was significantly high, so SORGAM overestimated the chamber derived SOA concentration in general. During the field campaign, the temperature was often low (the average as 289 K) so that the simulations by SORGAM-T were higher than SORGAM. For α-pinene, the RH correction was the reason for the higher simulated SOA concentrations by SORGAM-T. In addition, the seed aerosol concentrations (M_0) are lower in field measurements than that of the chamber experiments. According to Figure 3, when M_0 is low in the campaign, the $Y_{updated}/Y_{original}$ is higher.

3. It's not clear to me why the newly temperature-dependent model should include temperature dependence in BOTH K's and alphas. What is the physical explanation for why alpha's would change with temperature? Are these additional parameters justified or could you just be overfitting the available data?

Response:

The temperature dependence of alpha is not very straightforward. In the two-products model, the value of alpha is depends on the specific products combinations according to the dominate reaction pathways in different temperature regimes (e.g. Hallquist et al. 2009). This kind of dependence was also observed in chamber experiments. For example, a negative temperature dependence of alpha was observed in the ozonolysis experiments of α-pinene (Fig. 6a of Saathoff et al., 2009).

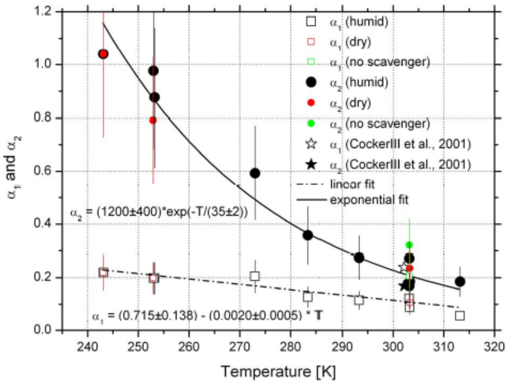

Fig. 6a. Temperature dependence of the mass stoichiometric yield coefficients of the two effective SOA forming reaction products of the ozonolysis of α-pinene.

Therefore, in our new temperature-dependent model we try to fit both K and alpha.

4. Particularly in the case of the 3rd-order polynomial fit of the small temperature range of available data for aromatics, I wonder if you are overfitting a small dataset. The shape of the function in Fig. 3 doesn't seem physical, and the high correlation coefficient reported in Table 4 (0.933) seems suspect in this light. Is there a physical reason to believe a 3rd order fit is appropriate, or could you be fitting noise in the data?

Response:

We agree that a limited effect of overfitting is possible with a cubic polynomial. We also tried a linear and quadratic approach, which performed poor and did not reflect characteristics of the chamber experiment data well enough. On the other hand, we argue that possible effects of overfitting on our model results remain limited. With the present state of knowledge being insufficient to provide a sound basis for alternative fitting functions, we suggest to accept the 3rd order fit function, until further evidence is gained by theoretical results.

5. Does the RH parameterization have any substantial effect? This is introduced but thennot discussed in detail later – did it not change much?

Response:

The effect of the RH parameterization is included in the temperature dependent functions in this study. The total effects of temperature and RH variations are given in SORGAM-T. The specific effect of RH parameterization on the simulated SOA yields can be estimated by equation 4.

We clarified the effect of the RH correction in L 21, P 5969: "The above humidity correction was included in this parameterization as well. Taking α-pinene as an example, at T=293 K and M_0=5μg/m^3, the yield of SOA increased 2.5% at RH=50%, as calculated by Eq. (4)."

6. In the discussion of improved model performance (e.g. last 2 paragraphs of section3.2), I suggest using parallel sentence structure and reporting the same variables for each case. Currently, you sometimes give the NME both before and after changes, sometimes only after. Then, in section 3.3.3, when you describe the use of SORGAM-TIN for the Cabauw data, you describe model improvement in different terms. I suggest being consistent throughout – report all effects of changes in the model in terms of that same NME metric.

Response:

The NME was complemented for both the original and updated model in the section 3.2.

While in section 3.3.3, the concentrations of SORGAM-T, SORGAM-I and SORGAM-N in Fig. 7b were only parts of the simulated SOA (SOA_updated), and the purpose to show them separately is that we focused on the contribution of each improvement. We gave the NME of the SOA_updated, but the NME for each improvement was not necessary.

In general, the figures look great and illustrate the authors' major points well.

8. Fig. 8: Why does isoprene, like NO_3, also add more SOA at night than during the day, even though emissions should be 0 at night?

Response:

The increase of nighttime SOA in the SOA_ISO run could be influenced by several factors. On one hand, the reduced PBL height could enhance the simulated SOA concentrations. On the other hand, the isoprene oxidation does not necessarily stop immediately when the emission is shutting down. That is of course depending on the specific

boundary conditions such as size of the reservoir and also the dilution rate. For Cabauw simulation, the calculated isoprene concentrations and the isoprene oxidation rate were often above zero.

Specific comments:

1. You sometimes refer to the field data as "IMPACT" and elsewhere as "Cabauw" – better to chose one and be consistent.

Response:

It has been revised accordingly. Except for the description of the campaign, the field data are consistently named as Cabauw measurements.

2. The last sentence of the abstract should be removed - this is appropriate for the discussion at the end of the manuscript but not the abstract.

Response:

The last sentence of the abstract has been removed in the revised manuscript.

3. Section 2.2: at end of 2nd sentence, add "with the following exceptions:" to clarify what you did/ didn't change with Hvaps.

Response:

Revised accordingly.

4. p. 5971 line 27: please justify / discuss using the largest experimental value for isoprene, rather than e.g. a mean value

Response:

This value was used in GEOS-Chem (Pye et al., 2010), with the volatility basis set (VBS) method (4 bins), which is different from the two-product method in SORGAM. For the reaction between NO_3 radical and isoprene, α was 0 for the two bins of products with lower saturated concentrations. We chose the value for the third bin with middle volatility, and used it in SORGAM. Comparing to CMAQ using two-product method as well, this value is between α_1 and α_2 for isoprene products of CMAQ, but higher than the mean value.

5. p. 5972 line 22: missing space

Response:

Revised accordingly.

6. p. 5973 lines 9-11: "We limited the form…not higher than 3": Why 3? Seems overfit with such a small dataset over small T range

Response:

Please see our answers to question #4 in general comments.

7. p. 5973 lines 14-15: "…extended to all oxidation pathways…" this assumption deserves some further commentary – how realistic is it?

Response:

This kind of extrapolation may introduce additional errors since the oxidation products from OH, NO_3 and O_3 are different. For aromatics, the major oxidation pathway is reaction with OH, which has been studied in chamber experiments. The extrapolation to NO_3 is therefore just a minor error. For α-pinene and limonene, of which all the oxidation pathways are of comparable importance, the situation could be more complicated. Often the ozonolysis of these two kinds of terpenes were studied in the chamber experiments. The error could be significant in this case.

We estimated the uncertainty of the extrapolation, and added it at the end of this paragraph accordingly: "According to the main reaction pathway of each precursor, the uncertainty of the extrapolation was estimated no more than 24%."

Please also see our answers to question #1 of referee #2.

8. p. 5981 line 11: SORGAM

Response:

Revised accordingly.

9. Table 1: suggest adding lines in between VOC rows – the alignment is confusing now. Also, in the footnotes you say updated parameters are bold, but they don't look bold, and it would be helpful to the reader if they were.

Response:

Revised accordingly.

References:

Hallquist, M.,Wenger, J. C.,Baltensperger, U.,Rudich, Y.,Simpson, D.,Claeys, M.,Dommen, J.,Donahue, N. M.,George, C.,Goldstein, A. H.,Hamilton, J. F.,Herrmann, H.,Hoffmann, T.,Iinuma, Y.,Jang, M.,Jenkin, M. E.,Jimenez, J. L.,Kiendler-Scharr, A.,Maenhaut, W.,McFiggans, G.,Mentel, T. F.,Monod, A.,Prevot, A. S. H.,Seinfeld, J. H.,Surratt, J. D.,Szmigielski, R. and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmospheric Chemistry and Physics, 9(14), 5155-5236, 2009.

Pye, H. O. T.,Chan, A. W. H.,Barkley, M. P. and Seinfeld, J. H.: Global modeling of organic aerosol: the importance of reactive nitrogen (NO(x) and NO(3)), Atmospheric Chemistry and Physics, 10(22), 11261-11276, 2010.

Saathoff, H.,Naumann, K. H.,Möhler, O.,Jonsson, Å. M.,Hallquist, M.,Kiendler-Scharr, A.,Mentel, T. F.,Tillmann, R. and Schurath, U.: Temperature dependence of yields of secondary organic aerosols from the ozonolysis of α-pinene and limonene, Atmos. Chem. Phys., 9(5), 1551-1577, 2009.