Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-625-AC3, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temperature-dependence of aerosol optical depth over the southeastern US" *by* Tero Mielonen et al.

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Received and published: 7 October 2016

Response to the comments of the anonymous Referee 3

We thank the referee for the evaluation of our manuscript. We have taken the constructive comments into account to improve the manuscript. Our replies to the general and specific comments are given below.

The manuscript by Mielonen et al. attempts to answer the question of whether the observed temperature dependence of aerosol optical depth over the southeastern US is driven by BVOC emissions versus aqueous phase production above the boundary layer. They attempt this partitioning via the combined use of remote sensing obser-

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vations and a coupled climate-chemistry model. Such use of a model of this type is warranted for the problem due to the complex interacting sources of aerosol which makes causality difficult to infer through purely empirical means. I find the argument compelling, but also that the manuscript could be strengthen by greater clarity in explaining its approach and why the evidence presented adds up to a coherent storyline.

Thank you for these observations. We will clarify the text regarding the approach and improve the storyline.

Much of the empirical argument rests on the idea that different sulfate dynamics in different time periods lead to different behavior in the aerosol load. One issue here is that those time periods are not treated uniformly; in figure 1 these are before versus after 2008, whereas in figure 4, the late period also include 2002 and 2003. It isn't totally clear why this switch was made. I realize this is discussed on page 10 first paragraph, but I don't follow the logic for why it was handled this way.

The reason for the discrepancy between the observed and modeled time periods with low anthropogenic emissions is due to the imperfection of the modeled regional meteorology and deposition of aerosols. Therefore, the modeled AOD levels and SO4 concentrations in 2002 and 2003 are smaller than observed. This will be clarified in the text.

In assessing temperature sensitivities, the manuscript needs to be specific about what timescale it is working on. Figures 1-4 are seasonal means, figure 5 is monthly mean, and figure 6 is diurnal-scale. What is the justification for jumping around like this, and what processes will be dominant at the different timescales?

The main goal of the paper is to study seasonal means but due to the small amount of summers we decided use monthly averages to show the change in the modeled AOD due to different aerosol sources (Figure 5). The usage of seasonal averages produces similar fits and does not change the conclusions, thus we decided to show the monthly results because the larger number of points gives us a more reliable fit to the data. The

purpose of Figure 6 is to show how temperature dependent biogenic emissions affect AOD levels and how there is a delay between the emissions and the increase in AOD. Therefore, these results were shown with the diurnal-scale.

For the modeling part of the argument, the crucial point is whether the no aqueous phase experiment accurately captures the essence of the hypothesis put forward by Ford and Heald. It is not clear to me that it does, so I think a stronger explanation and justification needs to be made of what mechanisms this experiment tests, and why this experiment is accurately representing that hypothesis.

We agree with the reviewer that a more thorough explanation will improve the manuscript. In the revised manuscript we will discuss the Ford and Heald (2013) hypothesis as well as the SOA production mechanisms in our model runs in more detail.

In the introduction we stated that "Goldstein et al. (2009) detected an exponential dependence between the temperature and AOD in the southeastern US, and hypothesized it to arise from enhanced natural BVOC emissions on warmer days. However, a subsequent study over the same area revealed that the previously observed correlation is predominantly associated with the occurrence of aerosols above the surface layer and the authors hypothized the source to be increased aqueous phase aerosol production (Ford and Heald, 2013)."

Our description of the hypothesis put forward by Ford and Heald (2013) in the MS was somewhat inaccurate. Ford and Heald (2013) compared the satellite observed and modelled vertical extinction profiles, and showed that their GEOS-Chem model lacked a source of aerosols in the lower troposphere. While they discussed the possibility that this source is related to aqueous phase oxidation of VOCs, the lack of correspondence between cloud cover and aerosol loading in the satellite data prompted them to mainly state that "the oxidation of biogenic VOCs, whose emission peaks in summertime, is required to explain the observed aerosol enhancement above the surface layer."

In the revised manuscript we will give a more accurate description of the Ford and

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Heald (2013) hypothesis, which we now see as being related to lack of biogenic SOA source in general, with a possibly significant involvement of aqueous phase SOA production. We will also present our modelled monthly AOD values compared to those from satellite retrievals (Fig. 1 below). This data, along with the AOD data shown in figure S12, shows that the model captures well the yearly averaged AOD in the region without a systematic low bias. Therefore our model does not seem to be missing a significant source of aerosols that would affect AOD. Furthermore, the figure below shows that trend and magnitude of summetime AOD is captured better by the model including biogenic SOA production (control run) than the model run without biogenic SOA. However, our modelled wintertime AOD appears to be too large in all the model runs, probably related to our use of yearly averaged non-biogenic emissions that are likely too high during the winter and too low during the summer. These issues will be discussed further in the revised manuscript.

We will also discuss further the details of our new representation of gas phase SOA production, that is based on volatility basis set -representation of Donahue et al. (2011) augmented with volatilities determined by chamber experiments (Kokkola et al., 2014). Here we would like to note that the new gas phase SOA production scheme in our model is distinctively different than that applied by Heald and Ford (2013) in GEOS-Chem. Furthermore, we will discuss in more detail the applied aqueous phase SOA production scheme in aqueous aerosols. This scheme does does not greatly contribute to total AOD in the studied region (Fig. 4 in MS), so the improved trend in modeled summertime AOD (as seen in the figure below) is mostly due to gas phase SOA production, also represented in the "no aqueous phase" experiment. Therefore, our main message with respect to Ford and Heald (2013) results is that the missing biogenic SOA component can be largely explained with the use of an improved gas phase SOA scheme. This is in line with the results by Kim et al. (2015) who used a newer version of GEOS-Chem and found that the model was able to reproduce the observed vertical distribution of aerosol mass and no additional aerosol source above the surface layer was needed. However, our results do not categorically rule out the

possibility of significant aqueous phase SOA production due to the lack of modelled SOA production in cloud droplets.

page 9, line 14: do you mean non-anthropogenic AOD anomalies?

Yes, we do. Thank you! This will be corrected.

page 9, lines 29 and 30: these p values are completely meaningless. the relevant test of a model at this level of complexity should not be that it performs better than a completely uninformed null model, but rather that it performs better than some informed but much simpler model. delete this or else come up with a much better null model that you argue the full model has better skill than.

We agree with the reviewer and we will remove the p-values.

Also the sentence at the end of this paragraph doe not logically follow.

We modified the sentence as follows:"Based on these comparisons, the modelled values agree reasonably well with the observations, which gives confidence that the model results can be used to illuminate the mechanism behind the observed phenomena."

references: note that Ford and Heald 2013 is discussed in the manuscript but does not show up in the bibliography

Thank you for pointing this out! We will update the bibliography.

References

Kim, P. S., Jacob, D. J., Fisher, J. A., Travis, K., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., Jimenez, J. L., Campuzano-Jost, P., Froyd, K. D., Liao, J., Hair, J. W., Fenn, M. A., Butler, C. F., Wagner, N. L., Gordon, T. D., Welti, A., Wennberg, P. O., Crounse, J. D., St. Clair, J. M., Teng, A. P., Millet, D. B., Schwarz, J. P., Markovic, M. Z., and Perring, A. E.: Sources, seasonality, and trends of southeast US aerosol: an integrated analysis of surface, aircraft, and satellite observations with the GEOS-Chem chemical transport model, Atmos. Chem. Phys., 15, 10411-10433, doi:10.5194/acp-

15-10411-2015, 2015.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-625, 2016.

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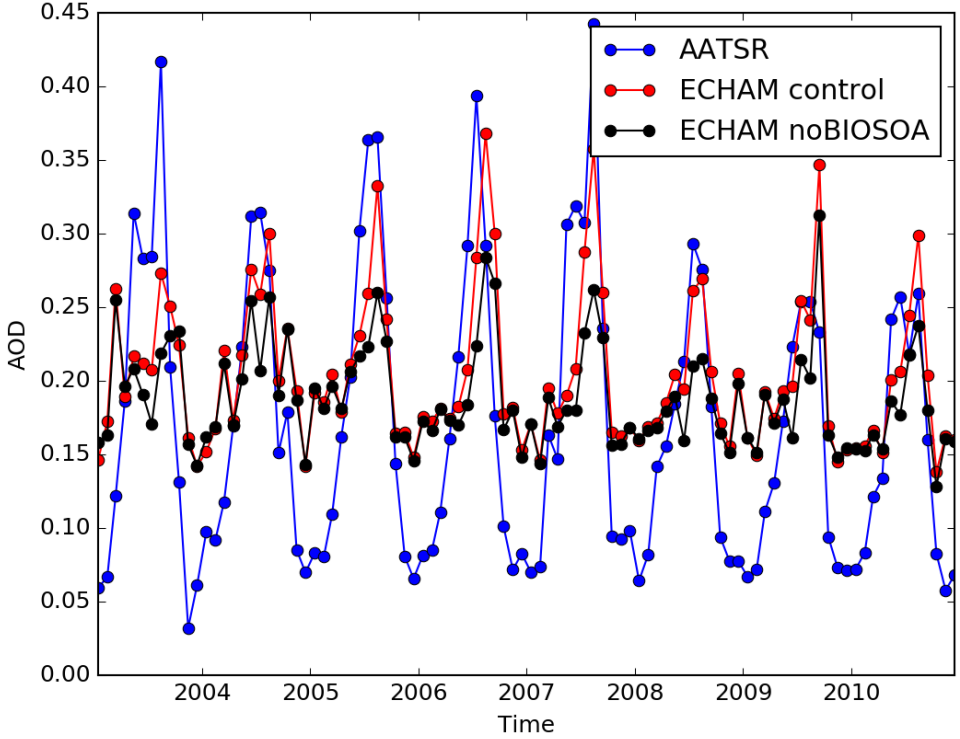

Fig. 1. Monthly averaged AODs for the years 2003-2010 over the southeastern US based on AATSR observations and the CONTROL and noBIOSOA model simulations done with ECHAM-HAMMOZ.

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