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Atmospheric Chemistry and Physics Discussions

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

Tero Mielonen et al.

tero.mielonen@fmi.fi

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Response to the comments of the anonymous Referee 1

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 presents an investigation of the effects leading to the enhanced summertime AOD detected over the southeastern USA by satellite instruments. The authors show that the temperature dependence of AOD is an indication of biogenic aerosol formation by comparing observed trends (of AOD vs. land surface temper-

ature) with modeled data in which biogenic aerosol formation is switched on or off. I have several major issues with this manuscript, as I will motivate below. My recommendation to the editor is to reject the manuscript in its current form.

My main objection to the study is that it does not appear to provide any new insights: a very similar (though more thorough) study has been presented in the paper by Goldstein and co-workers (2009), cited in the manuscript.

Here, we respectfully disagree with the referee for several reasons. First of all, the aim of our manuscript was to study if the results presented by Goldstein et al. (2009) could be reproduced with other, independent observations. Reproducibility is the corner stone of science which means that even published results are not that trustworthy if they have not been independently confirmed. Secondly, our study period covers 9 years whereas the paper by Goldstein et al. (2009) was limited to 6 years. The difference in the time periods reveals how changes in anthropogenic emissions affect the temperature dependency of AOD and this was not covered in Goldstein et al. (2009). Furthermore, their prediction of higher regional summertime aerosol levels has not actualized due to reduced anthropogenic emissions. Thirdly, there are some shortcomings in the analysis by Goldstein et al. regarding the radiative effects of this phenomenon. For example, they do not provide any estimate for the uncertainty in their radiative effect. We'll discuss this more in the specific comments below. Lastly, we were able to remove the anthropogenic contribution from the total AOD and provide a temperature dependent estimate for the radiative effect of biogenic aerosols in this region. This has not been done before.

Second, the statistical analysis does not appear to be very robust. The numbers in the manuscript are somewhat convincing, but the figures show very shaky correlations: the number of points is very small in Figure 1, and although the correlation for the red and blue data sets in the matching Figure S1 in the supplement looks alright, it is unclear how the data are separated, as now some red dots have AOD anomalies <0, whereas two blue dots have an anomaly >0.

We agree with the reviewer that the number of points is small which makes statistical analysis challenging. Therefore, we used Bayesian inference in the fitting. This method takes into account the uncertainty in the variables and the number of points. The fewer points you have, the larger uncertainties you get. Consequently, our uncertainty estimates are large. However, estimates of uncertainty have usually been excluded in previous studies.

Regarding the figure S1, the monthly anomalies in this plot were calculated separately for every summer month (JJA). The red dots represent summer months between 2003 and 2007, while the blue dots are for the summers 2008-2011. Monthly anomalies are not the same as the summer anomalies shown in Figure 1 and therefore, the red dots can have negative AOD anomalies and blue dots positive. For example, for the blue dots this just means that some of the months had larger than average AODs even though the corresponding summers had lower than average AODs.

Third, the results from the model study appear to be rather trivial: it is obvious that AOD due to biogenic aerosols depends on temperature, as this is explicitly parameterized in the model.

It appears that we were not able describe the reasoning behind the model study well enough. The aim was not to see if the AOD due to biogenic aerosols was temperature dependent but to see if biogenic emissions have large enough effect on AOD to explain the observed temperature dependency of AOD. Regarding the parameterizations in the model, the referee is partly right: the gaseous emissions from vegetation are temperature dependent but the formation of biogenic aerosols is not explicitly. The gaseous emissions contribute to atmospheric chemistry and some of them condense on aerosols depending on the atmospheric state and composition. Thus, several processes affect the formation of biogenic aerosols in the atmosphere and the model analysis was used to estimate the magnitude of this aerosol source and its significance over southeastern US.

Fourth, the values of the radiative effects of biogenic aerosols determined from satellite data are about twice as small as their respective errors.

The uncertainty in the radiative effects comes directly from the uncertainties in the fits between AOD and temperature anomalies. With a longer time series the uncertainties could be smaller but the AATSR data are limited to years 2002-2012. However, that is already a significant amount of observations. Several studies do not give any kind uncertainty estimates for their radiate effect estimates, thus we feel that our results are a step to right direction. In the future, these uncertainties can be lowered with longer data sets.

Last: please remember that correlation does not prove causality.

Thank you for pointing this out even though we have not suggested that.

Other comments (page 1, line 1 denoted as P1l1) : a. In addition to NO2 columns as a proxy for anthropogenic pollution, it might be of interest to study formaldehyde and glyoxal columns to obtain more information on VOC emissions. See papers by I. De Smedt or M. Vrekoussis, but also Veefkind et al. (2012, cited in the manuscript), or Penning de Vries et al., Atmos. Chem. Phys., 15, 10597- 10618, doi:10.5194/acp-15-10597-2015, 2015

Thank you for the good idea! We added OMI formaldehyde retrievals to the analysis and the data supports our previous conclusions. The summertime formaldehyde anomalies (2005-2011) are positively correlated with land surface temperature and the non-anthropogenic AOD (see Fig. 1 below). This implies that formaldehyde is linked to biogenic sources (as Veefkind et al. (2012) discussed) and that the non-anthropogenic AOD is linked to biogenic VOCs as our previous results indicated. These results will be added to the manuscript.

b. Why did your model control run not provide hourly output?

We don't have hourly output from the control run due to limited disk space in our dis-

posal. The 8 year simulation produced over four terabytes of data with hourly output even after we removed the southern hemisphere and variables we didn't need.

Instead of performing runs with and without certain aerosol sources, is it not possible to read out the AOD due to each aerosol type separately?

The modeled AOD can be separated for different components such as black carbon, organic carbon, sulphate and sea salt. However, these are only aerosol types and they can originate from various sources. Therefore, we have to do a separate simulation where the studied source is turned off and compare it with the control simulation in order to find out the contribution of this source. This method is also used in the IPCC simulations.

P2l7: "natural unperturbed aerosol" – this is not what you are studying in the south-eastern US. As you mention later on, Goldstein et al. (2009) suggested that SOA are more readily formed in the presence of pollution.

The referee is right and we don't claim that the aerosols over southeastern US are unperturbed.

P7l3: "the anomalies of the regional mean" – What is the reference? Or, to be more precise: how did you calculate anomalies?

The anomalies were calculated by subtracting the average of all the summertime values from each summer average. This will be clarified in the text.

P7l14-15: "NO2 column densities available from OMI (only available from 2005 onwards) were used" – Why did you not use SCIAMACHY data? SCIAMACHY was on the same platform and hence has the big advantage that it not only measured the same time period (2002-2012), but it also measured at the same time of day as AATSR. This is important for species that, like NO2, exhibit a diurnal cycle.

We agree with the referee that SCIAMACHY NO2 data would have provided a better companion for the AATSR observations and naturally we looked into it. We compared

tropospheric NO2 values from OMI and SCIAMACHY for the overlapping years over the studied region and found very little correlation in monthly or summer averages. OMI data has been used over southeastern US before (e.g. Krotkov et al., 2016; Russell et al., 2012) thus, it seemed more trustworthy and we decided to use it instead of SCIAMACHY data.

P9l5-6: "summertime FRP anomalies did not show any correlation with corresponding AOD or LST anomalies" – This would have been very surprising, as the region does not exhibit much fire activity. I'm pretty sure that even transported smoke (from Canada or Alaska) does not play a role, but since a summertime maximum of fires exists there, it would be worthwhile to at least mention transported smoke as a possible source of AOD over the SE USA.

Thank you, we will mention this in the text.

P10l22: "aqueous phase SOA" – What are these?

Aqueous phase SOA refers to organic compounds that form SOA through aqueous phase chemistry, i.e. in our set-up isoprene epoxydiols and glyoxals. This is mentioned on P5l24-25

P11l3-6: "there is a delay (. . .) takes at least several hours" – I do not believe this is quite correct. It takes a while for SOA to form, but it also takes a while for them to be removed, so I expect only a very shallow diurnal cycle (if any at all). In fact, your green line (noBIOSOA 2008) shows a minimum at the same position.

Yes, as Figure 6 shows, the diurnal cycles are shallow in all the simulations and all the simulations have a minimum during midday. The most interesting thing in the figure is how the daily cycles differ between warm and cold summers in each simulation. For the noBIOSOA simulation there is no clear difference in the daily cycle between the summers but the for the simulations with biogenic SOA (noAQSOA, noBB) the difference between the summers increases during night-time. This is caused by the

fact that it takes several hours for the biogenic vapours to condence on particles in a magnitude large enough to affect the total AOD. This will be clarified in the manuscript.

P11l24: "All the values for the equation, except for Srad and phi, were taken from Goldstein et al., (2009)" – Why did you change the cited equation? Goldstein (2009) and Haywood and Shine (1995) both included the local daylength (1/2) multiplied by the solar constant S0, which you substituted by Srad multiplied by phi. The values are almost the same, but it is unclear where your value for Srad comes from.

We changed the equation because the original equation by Haywood and Shine (1995) was designed for global estimates, not regional. Therefore, we modified the equation for regional calculations using incident solar radiation received in southeastern US during summertime (Srad) and the mean daytime value of the secant of the solar zenith angle (phi). Both values were averaged for the studied region and season using tools in the LibRadtran package. It's a lucky coincidence that Srad multiplied by phi is only about 10% smaller than S0 multiplied by daylength. This will be clarified in the text.

P12l29: "overestimating"- underestimating (I'm guessing you mean the model)

Thank you for pointing this out. The sentence was not well formulated. It was revised: "This is significantly smaller than the measurement based DRE estimates which implies that the measurement based estimates are most likely overestimating the biogenic effect."

P13l12: "Anthropogenic emissions are the main driver of AOD levels in this region" – This is only true if you mean that anthropogenic emissions enhance SOA formation. Which, to my knowledge, is still a hypothesis.

We based this conclusion on the correlation between the NO2 and AOD data, thus we are not able to say if the AOD is enhanced by direct particulate emissions or by SOA formation. The data just shows that when there is more tropospheric NO2 (tracer for anthropogenic pollution) AOD is also larger. This NO2-AOD dependence affects AOD

much more than the temperature dependent biogenic emissions. This was clear in both observational and model data sets.

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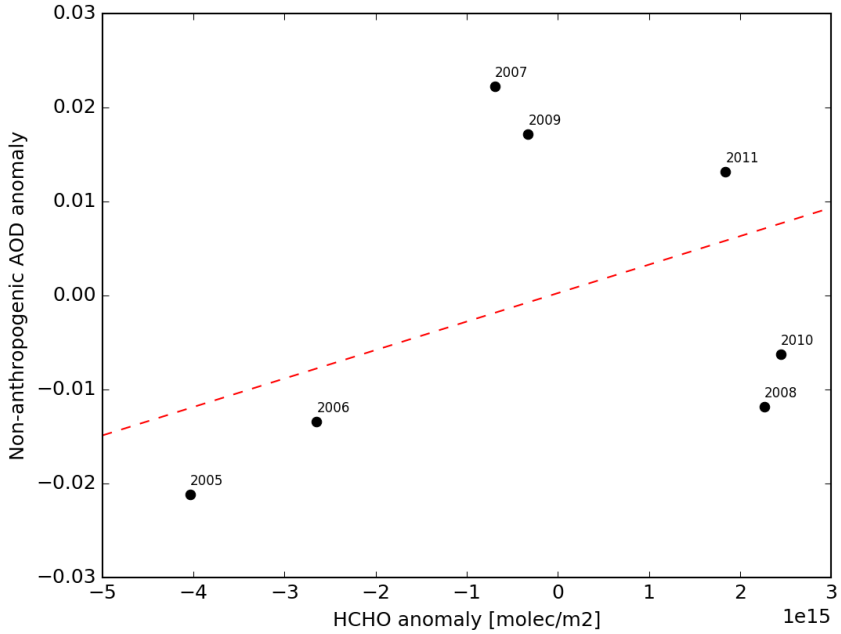

Fig. 1. Summertime anomalies in non-anthropogenic AOD vs. HCHO column, for the years 2005-2011. Non-anthropogenic AOD is based on L3 AATSR AOD and OMI tropospheric NO2 observations. HCHO is from L2G OMI.