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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.

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Response to the comments of the anonymous Referee 2

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.

My overall impression is that the authors do not present any new scientific insight. They discuss the Goldstein et al., 2009 paper but not many of the more recent papers (only Attwood et al., 2014; Ford and Heald, 2013; Hidy et al., 2014) that have looked at this region in detail or that present findings from the SAS/SOAS/SENEX/SEAC4RS

campaigns in 2013 (yes, I know this satellite product is only to 2012 but the findingsare relevant to the region, see Hidy et al. (2014) for comparison of 2013 to previous decade of observations).

Here, we respectfully disagree with the referee. The aim of our researcher was to estimate the radiative effect of biogenic emissions in this region and how it depends on temperature. This has not been estimated before. We concentrated on the results by Goldstein et al. (2009) because it was the only publication we could find about radiative effects of biogenic aerosols in this region. However, we found that their radiative effect calculation had some shortcomings and they did not provide any uncertainty estimates for it. Furthermore, they predicted higher regional summertime aerosol levels due to increasing VOC emissions in warmer temperatures but this has not been observed because anthropogenic emissions have reduced and they have a stronger influence on AOD. Therefore, we feel that our research contains scientific insight that has not been published before. 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.

However, we agree with the referee that our literature review has been too limited and we will revise it accordingly. We also thank the referee for the helpful list of publications from this region.

While they do use a new AOD product, it is not compared to other AOD datasets used in previous studies of the region, and the authors rely on correlations of anomalies without a thorough description of processes that I think are better discussed in other papers. My recommendation is therefore to reject.

The purpose of the study was not to validate the AATSR AOD product because it has already been extrenally validated (see Kolmonen et al. (2016) and Holzer-Popp et al. (2013). However, we did some comparisons with the MISR retrievals over the studied region and these results can be included in the supplement. The comparison between

monthly averaged Level 3 MISR and AATSR AOD products are shown in Figures 1 and 2 below. As the Figure 1 show both instruments exhibit similar seasonal cycles but the summertime AODs in the AATSR retrieval tend to be larger than the corresponding MISR values. This overestimation is also visible in the scatterplot (Figure 2) but the retrievals are very well correlated (R^2=0.92). As we use anomalies in the actual study, the bias in the AATSR AOD does not affect our conclusions.

The referee is right that the aerosol processes are described in more detail in other papers. However, we are using satellite retrievals and a climate model to estimate the magnitude and climatic significance of the effect biogenic emissions have on AOD in this region. These tools do not enable detailed analysis at the process level but they are well suited for our goals. A more detailed discussion of the aerosol processes in this region will be added to the manuscript.

I have several more specific points that I think should have been addressed and some statements that I found confusing. In particular, this specific AOD product from AATSR is relatively new (the Kolmonen et al., 2016 paper was published earlier this year). Therefore, I would have appreciated a greater description and validation of the product over this particular region. The Kolmonen et al. (2016) paper gives global validation statistics for the full time period (using binned AOD), so I'm unsure if there are any regional differences. The authors present anomalies of regional, summertime average AOD, which makes it hard to get a feel for the data. The authors should have shown some comparison with AERONET measurements and MODIS or MISR, specifically over the southeastern US.

As we stated above, the purpose of the study was not to validate the AATSR AOD product. However, we did some comparisons with the MISR retrievals over the studied region and these results can be included in the supplement. The comparison between monthly averaged Level 3 MISR and AATSR AOD products are shown in Figures 1 and 2 below. As the Figure 1 show both instruments exhibit similar seasonal cycles but the summertime AODs in the AATSR retrieval tend to be larger than the corresponding

MISR values. This overestimation is also visible in the scatterplot (Figure 2) but the retrievals are very well correlated (R^2=0.92). As we use anomalies in the actual study, the bias in the AATSR AOD does not affect our conclusions.

Additionally, I would like to have known: (1) How many observations went into each summer AOD calculation and what is the spatial distribution of these observations (the Kolmonen paper says observations are scarce, maybe available every 3-4 days at mid-latitudes)?

There is no straight answer to this question because we are using the Level 3 data with 1x1 degree spatial resolution while the actual retrievals are done with a resolution of 10x10 km^2. We have the information how many observations were used to calculate the L3 1x1 degree averages but we do not know their spatial distribution. We have not studied the spatial distribution of the L3 data but this can be added into the text along with the information about the number of observations. The referee is right that we do not have AATSR observations from every day but the gaps in the daily values are evenly distributed thus, they do not cause artificial biases into the monthly averages. The sampling is just scarcer than with MODIS for example.

(2) Is there interannual variability in the number or spatial distribution of observations?

The number and spatial distribution of the observations depends mainly on the orbit of the satellite and cloudiness, therefore we do not expect interannual variability. However, we will check this.

(3) How much spatial variability is there in the AOD anomalies and trend in anomalies?

This has not been studied yet but will be done by separating the studied region into three or four smaller areas and doing the analysis for them separately.

(4) Did the cloud filtering impact your results?

Not sure what the referee means with the cloud filtering here. The L3 products are are averaged from the cloud-free L2 products thus, we did not apply any additional cloud

filtering to the analysis.

Finally, I would like to have seen some discussion of the regional AOD (not just anomalies) and its trends compared to the AOD trends shown in other studies, such as Alston et al. (2012) and Attwood et al. (2014).

This kind of a comparison can easily be added to the analysis although, the goal of the paper is not to study temporal changes in AOD but how it depends on temperature. The comparison with the MISR observations (Figure 1) already shows that both retrievals produce similar trends.

These concerns with the satellite product and calculation of the anomalies all make me question the radiative effects calculations, which already have really large uncertainties.

We feel that the referee's concern on the quality of the satellite product used is exaggerated. The AATSR AOD has been externally validated and it compares well with MISR data over the studied region as already shown. The uncertainties in the radiative effect calculations are large because they are based on a limited number of observations with significant uncertainties. For example, Goldstein et al. (2009) and Attwood et al. (2014) did not provide any estimate for the uncertainty of their radiative effect calculations. Although, our uncertainties are large, they are realistic and the reported best estimate gives the most likely size range for the radiative effect in this region.

There was no discussion of aerosol water and how that might impact AOD. Nguyen et al. (2015) showed that there was a significant decrease in aerosol water mass concentrations from 2001-2012 in the southeastern US and Attwood et al. (2014) showed that the decrease in AOD over the southeastern US was due to both reduced mass loading and reduced aerosol hygroscopicity.

The referee is right that aerosol hygroscopicity plays an important role in the AOD and as the composition of the aerosols change so does their hygroscopicity. We did

not discuss this specifically because the satellite observations used do not contain information on the composition of the aerosols. However, when we estimated the "non-anthropogenic" AOD using tropospheric NO_2 data the change in the aerosol hygroscopicity was also taken into account because the higher AOD values were linked with more hygroscopic aerosols and the lower AOD values with less hygroscopic aerosols. Therefore, the change in the chemical composition of the aerosols and their hygroscopicity was taken into account, although indirectly. In the model results the effect of changing aerosol composition and hygroscopicity was naturally also taken into account. Furthermore, this study is not about the temporal change in the southeastern US but about the effect of rising temperature on the biogenic emissions and AOD.

The authors state that anthropogenic emissions are the main driver of AOD levels in the region because they use NO2 columns as a proxy for anthropogenic pollution and NO2 columns are correlated with AOD (for the modeling, they use SO4 mass as a proxy for anthropogenic pollution). Studies have found that a large portion of the aerosol mass is organics (e.g., Attwood et al., 2014; Edgerton et al., 2006; Ford and Heald, 2013; Kim et al., 2015; Xu et al., 2015) with SOA predicted to be 40–90 literature does strongly suggest that anthropogenic pollutants influence SOA formation (e.g., Budisulistiorini et al., 2015; Hoyle et al., 2011; Rattanavaraha et al., 2016; Volkamer et al., 2006; Xu et al., 2015), but I'm not sure this is what the authors are referring to when they are separating out the non-anthropogenic from anthropogenic contribution in the anomaly trends. It could be that decreasing anthropogenic emissions are leading to a decrease in SOA formation along with a decrease in inorganic aerosol mass. This kind of discussion is lacking in the paper. Separating into "anthropogenic" and "non-anthropogenic" may be too much of a simplification for the chemistry in this region.

We agree with the referee that the paper would benefit from a more detailed discussion on the processes affecting the AOD levels and we will add it to the text. The tropospheric NO_2 was used as a tracer for anthropogenic effect on AOD and it includes the effects of primary emissions and the enhancement of SOA formation. With our meth-

ods we are not able to discern the exact aerosol formation mechanisms thus, we have to use a simple separation into “anthropogenic” and “non-anthropogenic” components. However, even with this simple method we are able separate the temperature dependent effect of biogenic emissions from the anthropogenic influence. This has not been done before.

Page 3, line 16-18: Just over land or the whole region?

Just over land. We just used the same domain as Goldstein et al. (2009). This will be clarified in the text.

Page 3, line 21-27: Authors should state the overpass time.

The AATSR overpass time is approximately 10 am local solar time. This will be clarified in the text.

Page 4, line 5-10: Authors should state version of product. Also, how are clouds handled and AOD aggregated with regards to Level 3? It is not clear in the Kolmonen et al (2016) paper as the supplementary section describes clearing pixels and the main text discusses buddy checking in the final product, but I’m unsure of the impact on the Level 3 product.

The version of the product is 1.42. The Level 3 AOD is the average of the Level 2 product. The Level 2 (10x10 km^2 ground resolution) AOD is retrieved from cloud-cleared satellite observations. After the retrieval, the level 2 AOD is post-processed to remove remaining cloud contamination. More information on the cloud screening can be found from the algorithms ATBD: http://www.esa-aerosol-cci.org/?q=products

These will be clarified in the text.

Page 5, line 25: How does a reader see details of schemes in a paper that is in preparation?

This is a good point. We were aiming to submit the manuscript by Kühn et al. right

after this manuscript was submitted but it was delayed. We will provide a more detailed description of the schemes in the text.

Page 9, line 27-28: Unless the authors show that using daily vs. using the satellite overpass time does not have an impact, then it is not the right comparison. There should be diurnal variability in the aerosol mass loading (transportation has diurnal variability, photochemistry during the day, the southeastern US has interesting nighttime chemistry, eg. Xu et al., 2015; Ayres et al., 2015; water uptake is greater at night, e.g. Hidy et al., 2014, boundary layer impacts, e.g. Kim et al., 2015) and the trends may be different in daily vs. hourly, especially if there have been changes in the aerosol formation processes.

Figure 6 in the manuscript presents averaged daily AOD cycles for the summers based on the model results and from that plot it is evident that the modeled diurnal cycle of AOD is smaller than 0.01. Therefore, the usage of satellite overpass times only would not affect the comparison results.

Also, I’m confused by this comment that they only have daily because later (page 10, line 30), they discuss hourly results for their sensitivity simulations. The authors could at least look at one year and see how much difference it makes.

The sensitivity simulations had 3-hourly outputs but the control run was limited to daily values due to limited disk space. As stated above, we do not think that the usage of data limited overpass time would affect our conclusions.

Finally, did the authors also sample the model to the grid and days when the satellite had valid observations? Did this change their results at all?

The model results were not sampled to correspond with the satellite observations because the control simulation only had daily averages and the sensitivity simulations with better time resolution did not include all the aerosol processes. Therefore, we are not able to test how the comparison results are affected by the sampling.

Page 10, line 6: What do the authors mean that NO2 is not included in the model? Do they mean not included in the output? If so, why?

We mean that the model does not have NOx-chemistry and consequently, there is no tracer for NO_2.

Page 10, Line 14-15: This is a bit concerning. Are there no emission factors to account for weekly or diurnal cycles? The authors should at least discuss in better detail what impact this might have on their results. If there is no diurnal variability in anthropogenic emissions, then Figure 6 is misleading. Additionally, did the authors do a sensitivity test where they turned off anthropogenic emissions?

We agree with the reviewer that this is a deficiency in the model. The ACCMIP emission data used as an input for the model does not include daily or yearly cycles and the model does not superimpose any ideal cycles because they might not correspond to actual cycles. We will add more discussion about this into the text. Figure 6 is only used to show the temperature dependent effect of biogenic SOA on AOD and how the biogenic emissions increase AOD with a delay of some hours. These findings are still valid even though the simulations are done with a really simplified anthropogenic emissions. We did not do a simulation without anthropogenic emissions.

Page 10, line 7-12: This is confusing, can the authors just give the percent (normalized mean bias? Or such) rather than “half” or “one third”?

Yes, we can provide percentages instead.

Page 9, line 2-6: Most of the fires in the southeastern US are agricultural fires that do not produce much smoke, but the southeastern US also gets impacted by smoke from wildfires in the western US (as shown in Kim et al., 2015; Washenfelder et al., 2015).

We also used CO data from AIRS as a tracer for biomass burning plumes but we did not see any in clear influence from them. Moreover, the model simulations did not indicate that smoke (regional or transported) had a significant role in the temperature

dependence of AOD.

Page 11, line 1, 9-11: Aren’t biogenic emissions in the model a function of temperature?

Yes they are. The gaseous emissions from vegetation are temperature dependent but the temperature dependence of the formation of biogenic SOA is not not straightforward. 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.

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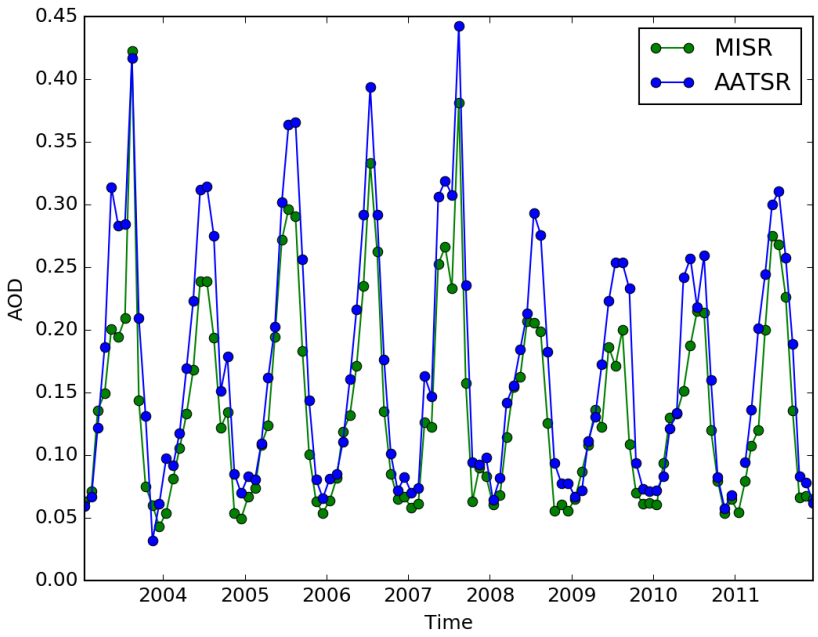

Fig. 1. Time series of monthly averaged AOD over the southeastern US from AATSR and MISR Level 3 products

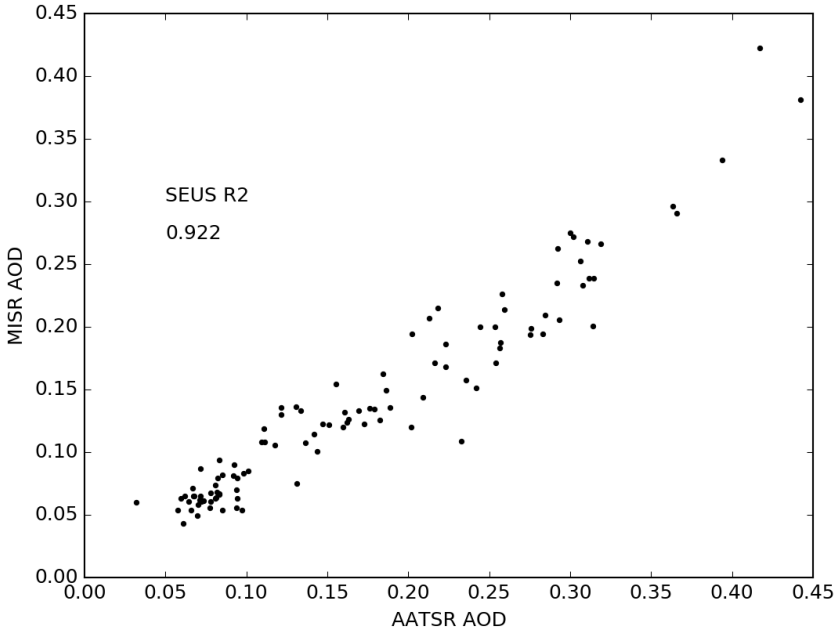

Fig. 2. Monthly averaged Level 3 MISR AOD vs Level 3 AATSR AOD over the southeastern US for the years 2003-2011. The R2 refers to the coefficient of determination.