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AMTD

4, C2986–C2997, 2012

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***Interactive comment on* “Characterization of atmospheric aerosol in the US Southeast from ground- and space-based measurements over the past decade” by E. J. Alston et al.**

E. J. Alston et al.

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Dear Referee #2,

On behalf of my co-authors and myself, we would like to thank you for your time in reviewing our manuscript. We appreciate your insightful comments and suggestions. We believe in addressing your comments our paper will become stronger and have a better appeal to the audience of this journal. We have carefully and thoughtfully considered every point you made. We will address your comments in the order given.

Major Comments:

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comment #1: The paper is thorough although not particularly well written. Reviews of prior literature are given but are given generally cursory treatment. Gupta and Christopher (2008) treated the same subject for much of the same period, but no discussion of the conclusions of that work are given. This topic has been widely published over the last 5–10 years and many of the papers have given little or no insight to the aerosol processes which correlate haze and mass. This paper often makes comments about the possible reasons for some observed correlation or behavior, but doesn't provide any assessment in depth to resolve the behavior. This is likely to weaken the impact of the work and place it in the larger panoply of suggestive relationships without any conclusions.

Response: First, we would like to reiterate at the scientific merit and timeliness of our research: –

We analyzed 10 years (2000–2009) of daily satellite data from three different sensors (8 years for one sensor) and ground-based observations of aerosols in the Southeastern U. S., which has not been previously done for this specific region. We used the most recent version of Level 2 satellite retrievals which has various improvements. – characterized the seasonality of regional aerosol signal in all datasets addressing similarities and differences, as well as the potential factors involved. –

We demonstrated the presence of decreasing trends in satellite AOD and ground-based $PM_{2.5}$ that implies the brightening over the study region during the past decade. We agree with the referee in that there have been a number of papers published on relating satellite AOD to $PM_{2.5}$ through linear regression analysis for air quality application. Yet, that is not this paper's intent. We are not seeking to use AOD as a proxy measurement for predicting $PM_{2.5}$, which is generally the goal of papers we cited in our introduction (e.g., Gupta and Christopher, 2008 and Zhang et al., 2009). We modified some text in Introduction to clarify this. Additionally, we define our region to be much smaller in scale compared to other published work for this region, and we do so to try to isolate the effects of Atlanta's urbanization against a relatively cleaner background.

Furthermore, we revised and expanded the text with discussion of our findings to pro-

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vide more in depth interpretation to the extent possible.

Referee comment #2: This reviewer is particularly perplexed by the observed trend in surface PM_{2.5} in Georgia from the surface samplers and no statistical trend in the AOD data. This must have a reason. Conjecture on the dominance of humidity in the AOD retrievals is one reasonable explanation in that the high haze events are dominated by high humidity and mask decreases in mass (or particle number), but other reasons (increases in burning and elevated smoke, for example) are possible but are not fleshed out. Could the authors not assess the number of fires over the decade from Fire Radiative Power (FRP) measurements?

Response: We expanded our discussion of possible factors that might affect the differences in the seasonal cycle and trends seen in AOD and PM records, addressing the potential influence of transported smoke events, organic aerosol layer aloft suggested by Goldstein et al. (2009), relative humidity, and differences in measurement techniques.

We also re-calculated the trends in Table 1 on a per year basis. Our statistical analysis incorporates testing of the slope and error terms as well. These re-calculated values are statistically significant for the MODIS Terra anomaly dataset and PM_{2.5} anomaly datasets.

Respectfully, we believe that assessing the influence of prescribed fires over the past decade is outside the scope of this research as that could be a possible publication in itself. Such an analysis will necessitate the use of a chemical transport model to predict smoke loading and areal coverage from FRP products. A recent work by Zhang et al. (2010), for instance, demonstrated significant discrepancy between previous studies caused by uncertainties in biomass burning emission, and modeled meteorology and smoke transport.

Referee comment #3: Trends are often masked by the treatment of the statistics. For example, the authors know that 2007 was an anomalous year with a major fire burning

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for much of the summer. Why not remove that anomaly in the trend analysis? Clearly, much of the AOD in any year in the southeast is fire related and you need to keep the average behavior, but a single event (or perhaps even 2σ events) could be removed to make the trends more robust. There is a technique known as “Winsorizing” which has long been used in trend analysis to remove outliers which dominate the slope of a trend. The authors might consider that.

Response: An additional statistical analysis was performed to test the influence of “outliers” (i.e., outside 2σ). We found that removing outliers has a negligible effect on the trend since only one-to-two points were filtered out in the 10-year monthly mean record of each data set. All filtered points were related to the 2007 wildfire event. We modified the text to clarify this point.

Referee comment #4: I have another concern about bias. The PM data at the surface is highly focused on Atlanta, with almost half of the samplers in that region. Yet, the AOD from the satellite should not have that bias. What would the authors have seen with more coastal surface monitors. Their results show elevated AOD near the coast. Is that real or an artifact of the satellite retrieval. Without concomitant PM measurements, it is impossible to determine whether there is bias here.

Response: Unfortunately, we used all of the available coastal monitors in Georgia. We believe the elevated AOD near the coast is an artifact of the satellite retrieval, specifically the transition between the land/ocean surfaces. We provide references that speak directly to this issue in the text, but we revised the sentence to make it more apparent why those citations are used as they address land/ocean bias in coastal areas. We suggest that a bias could be present in the statistics for both MODIS and MISR, but given the current availability of data it would be difficult to prove.

Minor Comments:

Referee comment #1: Pg 7560 line 14: there is “good agreement”. $R=0.64$ is hardly “good agreement” since only about 40% of the variance is explained. Thus my concern with qualitative statements of “good” or “bad”. There is correlation.

Response: The qualitative statement has been removed in the revised text.

Referee comment #2: Pg 7561 line 7 “Samoa”. Pg 7561 line 23 “each sites trend slope” is very poor grammar. Next line “each sites respective region” as well. You need to use apostrophes to help us out figuring out what you mean.

Response: The spelling and punctuation has been corrected.

Referee comment #3: Pg 7562 line 4 “geographic region that has been studied” grammar.

Line 13: showing a decrease. . . of what? 19% as well? How much of a decrease?

Line 14: “distinctly characterized”. Really? No other part of the US has this aerosol mixture?

Response: We changed “that” to “which”. According to Figure 15 of the EPA report as cited within the text, the SE is the only region with a high sulfate and organic carbon signature. Other parts of the Eastern U.S. have smaller organic carbon footprints and larger nitrate components.

Referee comment #4: Pg 7563 line 19: “Our earlier work, used probabilities of AOD” Why the comma? And what are probabilities of AOD? Probability AOD exists or doesn’t exist?

Response: We removed the comma. In Alston et al. (2011) we calculated the number of AOD occurrences that fall within the different AQI categories. From these occurrences of AOD per AQI category we calculated probability density functions that then provide information regarding the AOD/PM_{2.5} relationship. The sentence is meant as a summary of the main take-away from the publication, and for more details about that methodology we refer readers to that publication.

Referee comment #5: Pg 7564 line 14: Sect. should be spelled out.

Line 18: ten years record» the ten year record

Line 24: there are 18 sites, you used 12. . . seven are in Atlanta. What is wrong with the other six sites?

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Response: Section has been spelled out, and the spelling of years has been corrected. We excluded the other six sites because their datasets did not encompass the time period we are considering or had data records that were incomplete where a majority of the year had missing data. We have text already included in that paragraph about our exclusion criteria.

Referee comment #6: Page 7565 line 17: Atlanta's. Line 19: "We have three subsets for each PM_{2.5}." You mean that you subdivided the data into three groupings: all of Georgia, Atlanta and the remainder.

Line 23: Given the repeat . . . not taken. You need to describe this process in more detail. How did you fill in data on a 6-day repeat cycle– Did you linearly interpolate between times? How do you justify this in terms of aerosol spatial and temporal scales in Georgia?

Response: Yes, the PM_{2.5} data record was split based on geographic criteria as explained in the manuscript.

The fill values referenced in text are place holders to ensure that the PM_{2.5,FRM} dataset is the same length as the PM_{2.5,TEOM} dataset. The fill values (e.g. NaN) do not factor into any calculations, and no spatial/temporal linear interpolations were done. This section was modified to clarify the methodology.

Referee comment #7: Pg 7566 line 7: "5–15 min after" is not correct for Terra. Line 14: Nominal resolution? The resolution of MOD04 is 10x10km. The resolution of Terra is as small as 250 m. You need to be careful about which product you are describing. But you don't use data at that resolution. On Page 7567 you say you aggregate to 0.25 (about 30 kilometers) for MODIS and less for MISR. Your box is not Georgia. It is 30–35N x 80–85W which contains half of South Carolina and much of Alabama. Yet you only use Georgia ground monitors. This significant mismatch draws the work into question.

Response: We misspoke, the 5–15 minutes is usually true for MODIS onboard Terra. A correction was made in the text to denote which timeframe refers to which satellite.

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Also, we added which specific products we used to this paragraph. As for the different aggregations, this was only done for the spatial analysis (Fig. 6). The remainder of the study uses the nominal Level 2 aerosol product as listed in this data section.

Yes, the lat/lon box does cover portions of other states, our focus was really primarily on Georgia and impacts of the Atlanta metropolitan area on the rest of the region that we have broadly defined to encompass the entirety of Georgia. By limiting our focus to mostly GA, and still following our data guidelines that each station must have at least 5 years of data, there were two stations in Alabama and eight in South Carolina that were not used. The primary driver behind limiting the regional scope of this analysis is to control the number of point sources (largely Atlanta) and their impacts. The stations in SC would not likely be affected by metropolitan Atlanta, and some of these stations are closer to the metropolitan Charlotte area, which would introduce additional bias into our analysis. As for the two stations in AL, the same argument holds though in this case the stations would likely be influenced by the metropolitan Birmingham area, which has its own unique variability and sources. As such, no $PM_{2.5}$ measurements were used from either Alabama or South Carolina.

Referee comment #8: Pg 7568 , line 5–6: MISR has higher AOD than AQUA MODIS. Not in Figure 2A.

Line 12: an increase of 10 to 18 $\mu g/m^3$ is “modest”. Seems like a factor of two in my calculations (approximately).

Line 13–14 . . . TEOMs were operated only for 7 years. Did this enter into the removal of 8 monitors (see above).

Line 15: differences in measurement techniques. . . Do you have any evidence that the TEOM data is biased relative to the FRM method? If so, why has EPA not removed this data stream. If you are going to make a conjecture like that, you need to have evidence to defend it. What is the $PM_{2.5}$ correlation between TEOM and FRM from nearby stations?

Line 24: 2007 had anomalously high AOD. You know why. State it here instead of

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spinning this out as a mystery.

Response: That was a mistake. That has been changed to read “Generally speaking, both MODIS sensors have higher AOD than MISR.”

Yes, the $PM_{2.5,FRM}$ did approximately double, we did not want to overstate the case, so we chose to say modest. Also, the TEOM monitors have a shorter operational time than the FRM monitors and that is part of the exclusion criteria (please see above answer).

There have been some published papers that highlight some of the biases between discrete filter based measurements and TEOM measurements. The TEOM measurements are biased low in relation to FRM measurements likely due to loss of volatile organic compounds. But the two measurements are well correlated with each other in this study and in other published works. We have added some references to this section specifically focused on the TEOM/FRM bias. Having said that, it is unlikely that this bias is significant enough to remove this data stream from the EPA’s AirNow system, which is primarily used as a means to provide real-time air quality information, and it also feeds air quality models used across the country. The bias is the likely reason why currently TEOMs are not considered FRM equivalent (to determine NAAQS compliance) for $PM_{2.5}$ mass.

We provided more details about the 2007 wildfire in the section as to not prolong the mystery.

Referee comment #9: Pg 7569 line 19: “Remer et al. (2008).. 1% of the time over land”. What Remer actually said is that 80% cloud cover in a pixel with an aerosol retrieval only happened 1% of the time. This is different than what you suggest. Reread.

Response: The correct context of the reference was added to that sentence. It now reads “Remer et al. (2008) found that on a global scale, AOD in situations with 80% cloud fraction are twice the global mean of AOD values, although this occurred less than 1% of the time over land.”

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Referee comment #10: Pg 7570 line 5 “we do not force our linear regressions through zero”... This is an obvious reason why you do not have a zero intercept but means little.

Line 6: Kahn’s 2010 paper suggests no such underestimation of AOD by MISR. Reread.

Line 12–13: This argument of a difference between sources within the Eastern US and your study needs further argumentation. Is the speciation different in your region? Are there aerosol types not seen in the mid–Atlantic or Northeast? I find this to be a specious argument.

Line 13–14: this is not a sentence.

Response: The sentence relating to the intercept has been removed.

As for Kahn et al. (2009), we respectfully disagree about the suggested lack of underestimate of AOD by MISR. For AOD greater 0.2 or 0.3, MISR AOD values are systematically lower than MODIS AOD over both ocean and land. We added the full context of this reference to this sentence.

Lastly, we are not disputing that the Eastern U.S. has relatively similar aerosol types, however, as we state in the text, by controlling the size of our region and making it smaller we reduce the number of major aerosol contributors, i.e., large cities and their associated effects. That is a point that separates our study from that of Liu and Mishchencko, 2008.

The sentence was corrected.

Referee comment #11: Pg 7571 line 4: Move this argument up to line 24 pg 7568.

Pg 7571 line 18: Your argumentation that Goldstein’s data is biased by using Giovanni or another Level 3 product falls short since you aggregate your own data to a 0.25x0.25 degree grid. It is more likely that the monthly aggregation includes bad data. Giovanni

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was working on tools to remove outliers in that product.

Response: We added a sentence that relates to this argument to the suggested section.

While we recognize that any aggregation has the possibility of incorporating “bad data”, our point is that by using a Level 3 product reduces some of the spatial variability that is still retained at a 0.25 x 0.25 degree grid. It is not a bad thing to use Level 3 data, but it has to be used with some caveats. One of them being that it can possibly lead to overly simplistic conclusions especially about the spatial heterogeneity. We regularly use Giovanni for analysis, and we were co-authors on an AGU presentation using results from Giovanni.

Referee comment #12: Pg 7573, line 9: “does not have this peak”. It appears to in Figure 7B. What are you referring to?

Pg 7573 line 12: determination of trend in highly seasonal data requires care. You have implemented a “Pearson” type solution which aggregates months together and looks at the anomaly from the mean. Not a bad first approach. You might want to consider more sophisticated treatment (See Sirois and Barrie Arctic lower tropospheric aerosol trends and composition at Alert, Canada: 1980–1995; JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 104, NO. D9, PP. 11,599–11,618, 1999 doi:10.1029/1999JD900077 1999) for pulling trends out of highly seasonal data.

Response: The peak in question refers to Fig. 8, not Fig. 7. Regarding the trend analysis, see reply to comment #3 above.

Referee comment #13: Pg 7574 line 7–10: determining a trend in AOD has been elusive. But you might consider scaling the results to the mean of the anomaly, i.e express the trend in % per year rather than absolute AOD units. It would be more instructive as to whether it is in any way proportional to the percentage decrease in PM.

Response: As requested, we computed the normalized slope of trends (i.e., ratio of

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slope to standard deviation). Values for each data set are shown in Figure 9 in addition to the linear trend lines.

Referee comment #14: Conclusion: There is discussion included in the Conclusion that should be moved up to the body of the paper. Line 2–6 on page 7575 discusses humidity and since this is a major confounding factor between PM and AOD, it should be moved up and discussed in greater detail in the body of the paper.

Response: See our response to comment #2. Briefly, we expanded section 4. Conclusions to include more discussion of our results.

Referee comment #15: Pg 7583 Figure 1: Figure is unnecessarily complicated. Plot on a map, not google earth. I cannot see the labels and they will not reproduce well. Why include inactive monitors?

Response: We made new Fig. 1 with easy to read symbology.

Referee comment #16: Pg 7585: text says that the Terra AOD has higher variability than MISR. This is not apparent from this figure.

Response: I am not sure how to make this point more apparent. Yes, MODIS Terra has higher variability than MISR Terra, but the numbers are quite similar.

Referee comment #17: Pg 7590: Figure 6: No labels on plots to identify A–F. Why change colorbar from 0.4 to 0.3 for difference? The Summer Mean MODIS AOD shows a hot spot right on the Summer 2007 fires. This seems to dominate the entire summer 10 year average. MISR misses those fires entirely.

Response: The labels are have been added to the figure.

We changed the scale because the difference could not be as high as 0.4 as we were trying to show an appropriate scale for each separate sub-figure.

The spring wildfire of 2007 does likely skew the seasonal mean higher, while the effect the wildfire had on the summer mean is not as strong, see Fig. 5 for both Terra sensors. Additionally, MISR does indeed capture the wildfire of 2007 as can be seen through

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multiple figures, Figs. 3a, 5, 7, 8.

We believe we have addressed your issues and concerns; however, if further clarification is needed, we will be happy to provide it.

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