We thank the anonymous reviewer for their helpful comments – we have made several changes to the manuscript in response to their suggestions outlined in red below.

This study describes the characteristics of regional aerosol over the Southeast during the summer of 2013. Through comparisons with the GEOS-Chem model the paper aims to explain the distribution, speciation, and seasonality of PM and AOD in the region. The study provides some new insights into aerosol sources in the region and the August-October transition in concentrations. However, the text over-stretches in some interpretation, and leaves open some key questions. Here are some major issues that the authors should address/correct:

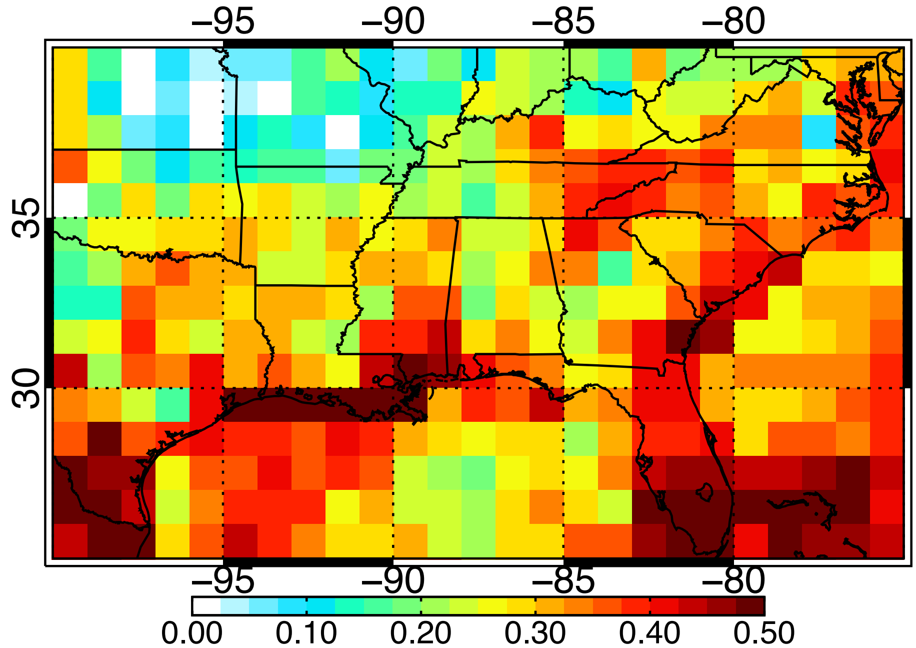
1. The GEOS-Chem model aerosol simulation used in this study is very different from previous published versions (meteorology, ML heights, resolution, emissions, injection heights, chemical mechanism especially with respect to sulfate and OA formation). In order to interpret the results, and particularly comparison with previous GEOS-Chem studies, this study should provide some context for how these changes impact the PM simulation, and where possible (e.g. the impact of changes to sulfate and SOA formation, as well as ML heights) some "before" and "after" comparisons. It's not clear from the manuscript whether the ability of the model to capture PM concentrations in the Southeast in 2013 is a result of the extensive model modifications and if so, which factor(s) are most important.

We have added quantification of the major changes and their impact on the PM simulation in Section 2.

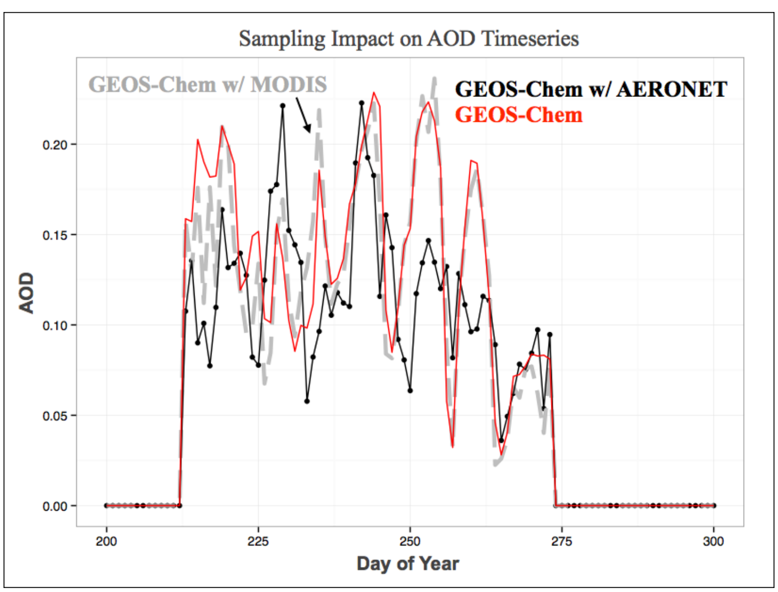
2. Figure 10 and Section 6: The figure shows that the model substantially underestimates MODIS AOD (factor of 2?) in the summer in the SEUS (a seen in Figure 13). This bias should be quantified and discussed in the text, particularly in light of the closer agreement in surface PM and extinction discussed previously (i.e. closure is not achieved, statements on page 17675, line 21-23, page 17676, line 9-10 and all similar statements in the text should be removed). This comparison appears to be in line with the previous results of Goldstein et al., 2009 and Ford and Heald., 2013. The statistics in Figure 10 suggest that both MODIS and GEOS-Chem are both biased low (NMB = -16% vs NMB = -18%) compared to AERONET, whereas the top-left panel of Figure 10 MODIS appears biased high compared to AERONET, not low. This should be resolved. (In addition the sentence on page 17672, line 25-26 is not supported by this analysis).

The model underestimates MODIS AOD by 28% averaged over the Southeast US during August and September 2013 (Figure 10). This is shown below in the difference plot and the information has been added into the text.

GEOS-Chem Percent Low Bias Relative to MODIS AOD
August-September 2013



We have moderated our discussion on closure as suggested by the Reviewer. We have added text to explain the apparent discrepancy between the statistics shown inset on Figure 10 and the map. In particular, the statistics compared to AERONET are calculated only when there are collocated and corresponding data for both AERONET and MODIS, whereas the map shows the spatial average for all available data during the mission. This impact of sampling time and location on regional mean AOD is illustrated further in the figure below using GEOS-Chem. The red line shows pure GEOS-Chem output, the black and gray lines when sampling at the available MODIS and AERONET retrievals respectively.



3. The plots and data do not support the conclusion that this model captures the seasonality in AOD in the Southeast. Figure 4 shows ~4-fold increase in observed AOD from winter to summer; whereas the model increase is at most a factor of 2. The text should be extensively revised accordingly, particularly in Section 7 and 8 and the abstract.

We have moderated the text and expanded on the discussion in Section 7 on the underestimate of the seasonal cycle. Our main point however is to focus on using the model to understand how there can be no seasonal cycle in surface PM, but a strong seasonal cycle in AOD.

4. It appears that a highly simplified/tuned non-volatile SOA simulation provides a more reliable simulation of observed OA concentrations and variability than has previously been achieved in field campaign comparisons. What are the implications of this? Does this study suggest that SOA is non-volatile, and models should eliminate the use of partitioning theory and NOx-dependent yields?

This is an excellent point – we have added commentary to the text as to the implications of the simplified OA parameterization, and added a sentence to the abstract.

Minor Comments

1. Two recent studies (Nguyen et al., ES&T, 2015; Xu et al., PNAS, 2015) have suggested important OA formation mechanisms for the SEUS. How do these relate to the current simulation (are these mechanism included in GEOS-Chem?).

These OA formation mechanisms are not explicitly considered in the GEOS-Chem simulations presented in this study. We now reference Marais et al. (2015) for a more mechanistic GEOS-Chem simulation of OA including consideration of the above references.

2. Page 17656, line 25-26: This sentence should be removed as the manuscript does not support the argument that variation in PBL height is responsible for the seasonality in AOD. (The analysis of Section 7 suggests that the variation in PBL height leads to the simulated seasonality but does not quantify this effect. Furthermore the simulated seasonality underestimates the observed seasonality by a factor of ~2).

Sentence has been removed.

3. Page 17660, lines 27-28: Please clarify - aren't "aqueous aerosols, or cloud processing" included in the sulfate simulation in GEOS-Chem?

We have removed this statement, which was indeed confusing.

4. Page 17661 line 19-page 17662 line 2: This paragraph is confounding. The authors discuss how SOA yields depend on the fate of RO2, but have assumed that the yield is constant under all conditions, despite their statement that both low-NOx and high-NOx

regimes being equally important in this region. This seems like a major limitation of the model simulation, but the implications are not discussed. What conditions do the fixed yields represent and does this represent a lower/upper limit for SOA formation in the region?

This is indeed a limitation of the work and we now refer to Marais et al. (2015) for a more mechanistic GEOS-Chem simulation including different SOA yields in the two regimes.

5. Page 17662, lines 17-18: How does the GEOS-FP meteorology compare with GEOS-5 or MERRA with regards to ML heights? What is the impact of the correction of the ML heights on AOD and PM2.5 simulated in the region?

This information has been added to the text in Section 2.

6. Page 17665, lines 17-19: Is the GEOS-Chem simulation compared to these observations in these studies? If not, please justify this statement.

The comparison numbers of the model to the observations from these studies have been added to the text.

7. Page 17665, lines 27-28: If the trend in OC is driven by a decrease in anthropogenic emissions, why is the downward trend only significant in summer in this analysis?

We don't speculate in the paper on the factors driving the OC trends in summer or winter because we don't feel that our OC simulation is sufficiently mechanistic for this purpose. Again we defer to Marais et al. (2015), which discusses the issue of OC response to long-term trends in SO_2 and NO_x emissions.

8. Page 17667, line 1-2: "these small inconsistent biases may not be significant." – a 20% bias does not seem all that small. Please remove or justify this statement.

Sentence has been removed.

9. Figures 5 and 6 seem inconsistent, particularly with respect to concentrations in the 2-4km altitudes. Figure 6 shows that the median model concentration of sulfate is ~2 times lower than observed aboard the SEAC4RS aircraft, whereas Figure 5 shows much better agreement for mean sulfate. Similarly, median model OA appears lower than observed. The authors should comment on the differences between means and medians and/or choose a consistent approach to their analysis. In light of Figure 6, the statement of page 17668 line 28 seems over-stated.

We thank the reviewer for pointing out the inconsistency for sulfate. Median values are shown in both Figures 5 and 6. However, in Figure 6 the observed vertical profile for sulfate shown is for the SAGA measurement, not the AMS measurement as stated in the text. The figure has been updated accordingly and there are no changes to the conclusions stated in the text (which has also been moderated in tone). The OA values are consistent between Figures 5 and 6.

10. Figure 8: The relationship shown with this cloud of points is not very convincing, and thus this analysis does not seem particularly useful. I recommend removing the figure and shortening the discussion.

The figure has been removed and the text has been revised accordingly – more detailed follow up work on this phenomenon will be explored in Silvern et al. (in prep).

11. Section 6: Why is CALIOP not included in this analysis? It may inform the differences between the CRDS and HSRL, and could provide context for comparing 2013 with previous years. This seems like a major gap in the analysis.

CALIOP data are sparse and interpretation is difficult. We chose not to use them.

12. Page 17673, lines 14-24: Clarify that this mechanism is not included in the current simulation (Figure 12 could be misleading).

Clarification added to the text.