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Interactive Comment

Interactive comment on "Sources, seasonality, and trends of Southeast US aerosol: an integrated analysis of surface, aircraft, and satellite observations with the GEOS-Chem chemical transport model" *by* P. S. Kim et al.

Anonymous Referee #1

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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.

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).

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.

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

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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?

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?).

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).

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

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?

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?

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

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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?

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.

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 $\sim$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.

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.

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.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 17651, 2015.