

We would like to thank the reviewers for their time and constructive comments. Below we have copied the reviewer comments in plain text and included our responses in bold text.

Anonymous Referee #1 Received and published: 10 March 2015

This paper uses aircraft observations from SENEX and SEAC4RS to investigate what fraction of aerosol mass and extinction in the transition layer over the southeastern U.S. can be attributed to mixing vs. production. It represents a clear and careful analysis of a very nice dataset. However I was disappointed that the study didn't really go the extra step to connect to the big picture question of aerosol sources and seasonality in the southeastern U.S. which served as motivation for the study (i.e. Goldstein et al.). The results beg several questions: (1) how representative was the summer of 2013? Are these results generalizable for the southeast? (2) what is the impact of this highly selective analysis (sub-micron only, afternoon only, no biomass burning, no in-cloud) and how does this relate to average observed aerosol extinction in the region? The AERONET observation of Figure 13 suggest that the sampling may bias this dataset to low AOD, limiting its relevance to mean conditions. While these airborne campaigns provide a limited snapshot (which nevertheless should be mined further to identify the biases associated with the sub-sampling used here), there are longer term ground and satellite observations which should have been used to provide a wider context. Without such work to contextualize the results, the conclusions (line 3150 lines 17 onwards) are overstated.

The reviewer has raised several important points which we respond to in order:

- 1) We agree with the reviewer that adding some additional comparison with other AOD datasets would give the conclusions more context; however, we also would like to limit the scope of this manuscript to focus on the in situ aerosol profiles. To that end, we have added a figure and description in the text comparing AOD from the in situ profiles to AOD measured by AERONET sun photometers at two locations in the SEUS. Both the figure and additional description in the text are copied below.**
- 2) The focused analysis employed here is necessary to 'resolve' the small enhancement in the transition layer. A larger enhancement (one that would significantly increase AOD) would be clearly apparent in the profile of extinction (Fig. 3a). In the case of a large enhancement aloft, the extinction would increase with altitude. Because the extinction decreases with altitude, an analysis limited to profiles during well-developed shallow cumulus convection was needed to resolve the smaller enhancement.**
- 3) Our conclusions about the hypothesized layer of aloft are based only on observations we made during research flights during the summer of 2013. However, our conclusions *suggest* that further study of the seasonality of AOD, meteorology affecting AOD are needed, and that the hypothesis of an aerosol layer aloft should be re-examined. To provide more context for these conclusions we have added references (Alston et al. (2012) and Kim et al. (2015)) that discuss the hypothesis of an aerosol aloft over the SEUS.**

"The altitude-normalized aggregate profiles used in this analysis are drawn from 37 vertical profiles; however, they represent only eight afternoons during the summer of 2013. For comparison and context, Fig. 14 shows an extended time series of 532 nm AOD (level 2 data) measured by AERONET sun photometers [Holben et al., 2001] at the Centreville

SEARCH site and at the Georgia Tech site in Atlanta, Georgia. The Georgia Tech site is in an urban area and is perhaps biased toward larger AOD from urban emissions, while the Centreville site is rural. The sun photometers only report data during cloud-free conditions. Plotted on top of these data from the sun photometers are the AOD from the profiles used in the altitude-normalized aggregate. These data are grouped into the profiles from the SENEX and SEAC4RS studies. Aircraft profiles during the SENEX study did not sample AOD greater than 0.3 while the maximum of AOD observed by the sun photometers was greater than 0.4. Profiles during the SEAC4RS study, although limited in number, cover a range of AOD similar to the sun photometers. Because the majority of the profiles in the altitude-normalized aggregate are from the SENEX study, the aggregate may be biased toward cleaner conditions. The range of AOD observed during summers of 2011-2013 at the Georgia Tech site indicate that the summer of 2013 is not an outlier with AOD higher or lower than typical summers. This is consistent with the analysis of Kim et al. [2015] who has compared satellite measurements of AOD during the summers 2006-2013.”

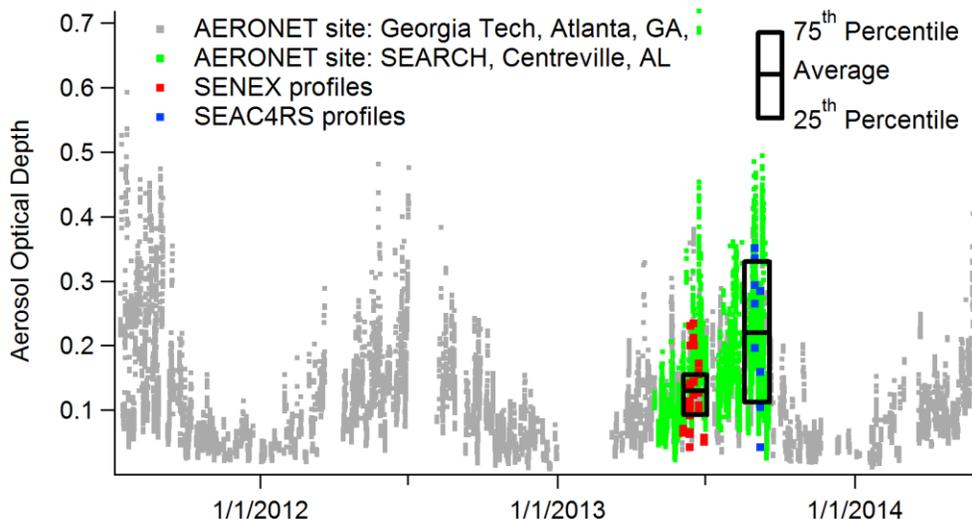


Figure 14: The AOD measured by an AERONET sun photometer in Atlanta, GA (gray) and Centreville, AL (green). AOD from the SENEX (red) and SEAC⁴RS (blue) profiles included in the altitude-normalized aggregate. The black boxes show the average, 25th, and 75th percentiles of AOD from both the SENEX and SEAC⁴RS profiles.

1. Abstract: should state year of observations as well as campaign names

We have added a sentence to the abstract stating the year and campaign names.

2. Page 3129, line 20; page 3130 line 17; page 3150, line 17: Goldstein et al., 2009 linked the seasonality of AOD to biogenic SOA. Ford and Heald concludes only that the observations support a significant summertime aerosol source aloft. They speculate that aqueous sources of SOA or H₂SO₄ from Criegee chemistry are possible sources, so it’s erroneous to suggest that SOA was the hypothesis of this study.

We thank the reviewer for pointing out the lack of nuance in our description of the conclusions of Ford and Heald (2013). To correct this we have rewritten sections of the abstract, introduction, and conclusion to make it clear that the Goldstein et al. hypothesize

a layer of SOA while Ford and Heald hypothesize an aerosol layer that would be composed of organics and/or particulate sulfate.

3. Sections 2.1 & 2.2: Please indicate the collection efficiency of the AMS instruments used and how these two instruments differ on the two aircraft.

We have added the following text to Section 2.1.

“The non-refractory aerosol composition was measured by a compact time-of-flight aerosol mass spectrometer (AMS) downstream of a pressure controlled inlet [Bahreini et al., 2008] and most (97%) of the submicron volume measured by the aerosol sizing instruments was transmitted into the AMS during SENEX. The collection efficiency for the AMS was determined by the composition for each data point using the algorithm described by Middlebrook et al. [2012]. When comparing the volume derived from composition (AMS plus black carbon mass) to the volume measured by the aerosol sizing instruments in the manner outlined by Bahreini et al. [2009], 87% of the aerosol composition and sizing data from the entire SENEX study are within the combined uncertainties.”

We have added the following text to Section 2.2.

“The non-refractory aerosol composition was measured by a high resolution time-of-flight AMS similar to the compact time-of-flight AMS used during the SENEX study. The two instrument differed in the resolution of the mass spectrometer. The higher resolution AMS used during SEAC4RS was operated with a 1 s sample interval. This AMS was operated similar to Dunlea et al. [2009] and also used a pressure-controlled inlet [Bahreini et al., 2008]. The composition-dependent formulation of Middlebrook et al. [2012] was used to estimate the collection efficiency. The AMS sampled aerosol downstream of a HIMIL inlet. (<http://www.eol.ucar.edu/homes/dcrogers/Instruments/Inlets/>). In both the HIMIL inlet and the shrouded diffuser inlet, the sampled aerosol was initially dried by ram heating and then further dried in each instrument.”

4. Page 3134, lines 1-5: state what wavelengths were used for the extinction measurements (here or in table)

We have add sentences to the text stating the wavelength used for the extinction measurements.

“The extinction of dry aerosol was measured at three wavelengths (405 nm, 532 nm, and 662 nm), and humidified extinction was measured at 532 nm. Only 532 nm extinction measurements are used in this analysis.”

5. Table 1 and 2: aerosol extinction is listed as “dry” although the text indicates measurements were taken at 3 RH conditions. Perhaps “dry” is meant to indicate the uncertainty under “dry” conditions? If so, uncertainties for “wet” should also be stated.

“Dry” was intended to indicate that the accuracy was for dry conditions. We have added a line to Tables 1 and 2 stating the accuracy of the extinction measurements at 90% RH. The additional uncertainty at elevated RH is due to uncertainty in the RH measurement (+/-1%) which is translated into an uncertainty in extinction using the kappa parameterization ($\kappa = 0.15$ typical see Fig. 3c) and added directly, because these are likely systematic (opposed to random) uncertainties.

6. Table 1 and 2: Please harmonize instrument labeling: some are given in full (e.g. Aerosol Mass Spectrometer) and some are given as acronyms (e.g. PTR-MS)

We have changed the acronyms in the table to full names.

7. Table 1 and 2: 5th entry should read “Dew Point (RH)” since the accuracy is given in units of dew point, not RH.

We have changed the 5th entry to ‘Dew point (RH)’ as suggested by the reviewer.

8. Section 2.4: would have been insightful for the authors to compare their extinction or total AOD values to other available datasets (CALIPSO, MODIS, or AERONET).

As suggest by the reviewer, we have added a comparison to AERONET AOD at two sites in the SEUS. Please see our response to the overview comments of the first reviewer above.

9. Section 2.4: Did the authors check if the fitted kappa is consistent with the measured composition?

Another manuscript, Brock et al. (in preparation), looks at the aerosol hygroscopicity during SENEX and SEAC⁴RS in more detail and has found that the volume-weighted kappa from calculated from the aerosol composition and the optical fitted kappa agree within uncertainties if we assume the kappa from the organic portion of the aerosol is $k_{org} < 0.1$, which is consistent with other work such as Good et al., Atmos. Chem. Phys., 10(7), 3189–3203, 2010.

10. Page 3136, line 27-28: awkward phrasing “globally and over the SEUS” suggests that the SEUS is not part of the globe. Suggest that this is re-phrased

We have removed globally from the sentence it now reads: “Shallow cumulus convection is common over the SEUS.”

11. Page 3137, line 10-11: Does “mixed layer height” mean top of the mixed layer? Please clarify.

We have changed “mixed layer height” to “the top of the mixed layer” to avoid any confusion.

12. Figure 3 caption (and generally all Figures): please specify which variables are measured and which are calculated.

We have added more description to Figs. 3, 8, 9, and 10 so that it is clear to the reader which quantifies are directly measured and which are calculated from precursor measurements.

13. Figure 4: It would be appropriate to use reduced major axis fitting here. Please specify in the caption/text if this is the case or correct if otherwise.

For this fit we have used orthogonal distance regression (ODR) and have added this detail to the text. Using the ODR fit method is appropriate because both independent and dependent variable are expected to have similar uncertainty and the purpose of the fitting is to determine the relationship between extinction aloft and on the surface.

14. Page 3139, lines 21-22: ion charges missing on nitrate (NO₃⁻), ammonium (NH₄⁺), and sulfate (SO₄²⁻)

In the literature, we have found examples of AMS data presented with and without the ion charges. We normally don't use the charges when referring to AMS data, since those signals can originate from organic species in addition to inorganic salts. For an example of AMS data presented without ion charges see Ng, et al., *Aerosol Science and Technology* 45(7), 770-784, Figs. 7 and 9. Below is a passage from the text illustrating this point. We have added the sentence in italics to inform the reader of the notation.

“The aerosol mass is the total of all ions measured by the AMS, and these ions are typically classified as SO₄, NH₄, NO₃, and OA. The inorganic ions are typically formed by ionization of simple salts such as ammonium sulfate and ammonium nitrate or may be formed from more complex compounds (i. e. organosulfates, organonitrates, and amines) that produce inorganic and organic ions when ionized. *To indicate this complexity, we have omitted ionic charges from the notation (i. e. SO₄, NH₄, NO₃).*”

15. Page 3141, line 14: “virtual potential temperature was constant”. With what tolerance?

We have added the tolerance to the text. The relevant sentence is copied below.

“For individual profiles, the mixed layer height was defined by inspection of each profile as the highest altitude at which the virtual potential temperature (Θ_v) was constant (typical variation in the mixed layer was less than 0.5 K) and there was a reduction in the isoprene concentration.”

16. Section 4.1: Please comment on the impact of neglecting horizontal advection.

We have added the following sentences to the Section 2.5 motivating the aggregation of individual profiles.

“Individual profiles are affected by horizontal advection which couples spatially inhomogeneous emissions to the vertical profiles. Because of vertical wind shear and spatial variability during slant profiles, the vertical layers in an individual profile are not always directly comparable. The aggregation the individual profiles is used to reduce the influence of this variability and resolve the typical vertical structure and mixing over the SEUS.”

17. Section 4.1: Could you comment on how/whether this analysis of the fraction of air is affected by the lifetime of the assumed compound (i.e. CO) relative to the applied species (i.e. aerosols)?

For this analysis, the important time scale is that of atmospheric mixing. In the mixed layer this time scale is approximately one half hour. In the transition layer, the typical lifetime of a cumulus cloud is ~ 1 hour, so the timescale for mixing in transition layer might be a few hours. Regional subsidence (typical velocity = 500 meters /day) mixes air from the free troposphere into the transition layer and mixed layer below. The lifetime of both aerosol and CO are much longer than all of these lower troposphere mixing processes. We could use other long lived species (CH₄, CO₂, non-precipitating H₂O) to determine the fraction of air from the mixed layer, $f_m(h)$, and Fig. 8 shows that the vertical profile of all of these long live species behave in a similar manner.

18. Page 3145, lines 23-24: How does the vertical profile of aerosol mass/extinction differ when including biomass burning?

During the SENEX study, the plumes from several small agricultural fires were transected over the SEUS. These transects intentionally targeted these plumes, and the plumes were transected during level flight legs. During SEAC⁴RS, several large western wildfire plumes were sampled over the western US (in research flights that were not used here) and several small agricultural fires plumes were similarly sampled over the SEUS again largely during level flight legs. Because the analysis presented here only considered the ascending and descending portions of the flights, these biomass burning plumes were *implicitly* not included in this analysis. Hence *explicitly* including/excluding these plumes has no effect on the analysis presented here.

In the course of our analysis, we first explicitly excluded the biomass burning transects from the data from each flight and then extracted the profiles. However we subsequently realized that the biomass burning didn't occur during the profiles. We have modified (copied below) the description of the data filtering in Section 2.5 to indicate this point.

“Transects of biomass burning plumes were identified using tracers such as the acetonitrile mixing ratio, were typically during level flight legs, and were not found in any of the profiles used in this analysis.”

19. Page 3147, line 15: typo: eq (5)

We have corrected the equation referenced in this sentence to Eq. 5.

20. Page 3147, line19-20: The authors might want to note that this approach is supported by their own analysis in Figure 4.

We have added a note that the assumption is supported by the correlation in Fig. 4.

“*Because extinction at the surface and aloft in the mixed layer are well correlated as shown in Fig. 4, the dry extinction is extrapolated as a constant to the surface based on the mean extinction measured in the lowest 200 m of each profile.*”

21. Figure 11: caption missing (caption is for Figure 12).

We have corrected this error. The caption for Fig. 11 follows below:

“**Figure 11: Histograms of the transition layer enhancement ($E(h)$ – see Eq. 4) for several trace gases and aerosol properties. The first column shows conserved species and black carbon: (A) CH₄, (B) CO₂, (C) H₂O, and (D) black carbon mass. The second column shows the aerosol extensive properties: (E) aerosol mass, (F) dry extinction, and (G) aerosol volume. The third column shows the aerosol composition: (H) OA, (I) SO₄, (J) NH₄, and (K) NO₃. The Student's T-test and resulting p-value (noted in each histogram) were used to test if the mean of each distribution was statistically different from zero.**”

22. Figure 12: typo “total sulfur (tS).”

We have corrected the labels on Fig. 12.

This manuscript uses aircraft in-situ data from two different campaigns (SEAC4RS and SENEX) during the summer and early fall of 2013. The aircraft platform has advantages to other platforms (space and ground) in that vertical layers can be probed directly. The analyses presented though representative of a small time period, are methodical and logical. However, these results seem to be disconnected to other research in this area. This reviewer kept waiting to read about how these results tie in (or do not tie in) with other contextual works. The conclusion could potentially be quite insightful, if given the proper context in which to state it. Overall, the work is sound and could be improved by minor changes.

We have made a few changes to the manuscript to provide the reader with more context. First, we have added a figure which compares the AOD of aircraft profiles to the AOD measured by sun photometer at two sites in the SEUS. Second, we have added a brief discussion of two recent articles relevant to the hypothesis of the layer aloft. The following sentences were added to the conclusion.

“The hypothesis is partially supported by the spatial similarity of summertime biogenic emission and summertime AOD over the SEUS. Although, Alston et al. [2012] found that the spatial similarity depended on the spatial resolution AOD of the data used in the analysis, and their analysis of AOD and surface aerosol mass over Georgia did not fully support the hypothesis. Kim et al. [2015] found that the increase of the planetary boundary layer height during the summer could bring the seasonality of the AOD and surface aerosol mass into agreement without the need for an aerosol layer aloft. “

Specific points:

1 - Section 3.1: Were there any other measurements that could be related to groundbased measurements similar to Fig. 4 in Section 3.1. This could open up an avenue of comparison with older field intensive studies in the same region, e.g., Atlanta Supersite Experiment in 1999.

Although detailed comparison of aircraft and surface measurements and historical measurements are scientifically interesting and there is potentially a lot that can be learn from them, aside from surface measurements presented in section 3.1 (included to support assumptions used to calculate AOD) they are beyond the scope of this analysis.

2 - Typo: Line 9 3143 change expecting to expected

We have corrected this typo.

3 - Section 4.3: It would have been more conceptually thorough if the authors had brought in some satellite (CALIPSO for vertical profile comparison, OMI as another indicator of the lack of biomass burning aerosols) observation comparisons in Section 4.3. Even if that work is being done by another author, a reference to those results would have been helpful to understand what dynamics the entire region was undergoing at the time of these flights. Or the authors could have compared their results

with time averaged satellite information, which could open up the avenue for discussing how similar or dissimilar summer 2013 was from other summers.

We have added an additional figure and references to some complementary work (see the response to the overview comments of the first and second reviewers).

4 - Section 4.3: There did not appear to be much summarization at the end of each respective section. Is that on purpose? The only section that does an adequate job in relating that section's work to the field at large is Section 4.3.

This comment makes largely the same point as comment 5 and is addressed in the response to that comment.

5 - Conclusions: Most of this section speaks to the hypothesis of an elevated layer of aerosols which account for changes in AOD and the author's perspective on that. What about the other results? How do these other results shown in Figs 1-12 support the author's thesis statement? There was a lot of work done to create these different analyses, yet at the end there's scant mention of them.

Many of the other results are included to support the conclusion of a modest enhancement in the transition layer. For example Fig. 4, demonstrates that aerosol is well mixed down to the surface below the minimum altitude of the aircraft and that extinction can be extrapolated to the surface in the AOD calculation. Fig. 11 shows the statistical significance of the observed enhancements. Figs. 8 and 12 are included to demonstrate that other long lived species have a profile similar to CO. Fig. 12 also shows a reduction in the precursor of particulate sulfate, SO₂, further supporting the observed enhancement of particulate sulfate in the transition layer. In the conclusion, rather than reiterating these supporting agreements we discuss the disagreement between our observations and the hypothesized layer. To explain how the absence of a large layer of enhanced aerosol in Fig. 3 motivated a more refined analysis, we have added a paragraph to the conclusion to tie together the altitude-binned aggregate in Figs 3-5 with the altitude-normalize aggregate presented in Figs. 7-10.

“Here, we have examined in situ vertical profiles of aerosol and found the dry aerosol to be well mixed in the lowest layer. Above the lowest layer the aerosol mass and extinction decreased with increasing altitude above that layer (Figs. 3-5). The hygroscopic growth of aerosol resulted in a layer of enhanced extinction near the top of the mixed layer. The aerosol water accounted for approximately a third of the AOD which would account for a portion of the summertime AOD enhancement. The hypothesized, large enhancement of secondary aerosol aloft was not apparent in these aggregate profiles. However, after normalizing the altitude to the vertical structure and using the CO concentration to quantify the vertical mixing during shallow cumulus convection, we were able to resolve a modest enhancement of aerosol in the transition layer.”

6 - Figures: Fig 13 is somewhat confusing. What does the blue portion of the figure represent? A couple of sentences added to the caption would help orient the reader.

We have added the following sentences to the caption for Fig. 13.

“The idealized blue profile of extinction at the center of the figure shows the vertical location of each contribution to AOD. The light blue area represents the extinction of dry aerosol, and the darker blue area shows the enhancement to aerosol water.”

Additional changes:

- 1) We changes the notation of the hygroscopicity parameter from κ to κ_{opt} to avoid confusion with hygroscopicity derived from more direct measurements of the diameter growth factor i. e. HTDMA.**
- 2) We have added some more background detail on biomass burning in the SEUS to section 3.2 as suggest by B. Yokelson (through personal communication).**

“Biomass burning is common in the SEUS during the fall, winter, and spring, but is less common during the summer. Zhang et al. [2010] found that in the summer of 2007 biomass burning contributed between 2 and 10% to measurements of $PM_{2.5}$.”