Reply to Reviewer #2 (comments for Atmos. Chem. Phys. Discuss., 14, 26971–27038, 2014, A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions, by J. P. Sherman, et al.)

- We thank anonymous reviewer #2 for her/his excellent suggestions. We are particularly grateful for suggestions regarding paper organization, grammar, and for catching inconsistencies and other errors. We've gone to great lengths to implement a very large majority of the suggestions made by both reviewers. These efforts include a complete re-organization of the paper and major additions to the *Supplemental Materials* to be submitted with the revised paper. We include a detailed outline of the revised manuscript in our response to reviewer 1 (reviewer #1 comment 1). We also include the outline at the end of this document for easy access. The outline reflects several major changes in paper organization and content suggested by the reviewers. Brief mention of the rationale for new sections are also included in the outline. All added, modified, and deleted sections are based on recommendations of the reviewers. In our responses to individual reviewer comments, all references to figure, table, and manuscript section numbers are numbers of the materials in the revised manuscript, unless otherwise-stated. As an example, a reference to *Table 1* means that the discussed material will be located in Table 1 of the revised manuscript, not Table 1 of the original manuscript.
- We weighed the advantage of incorporating both reviewers' comments into a single document (easy of cross-referencing our implementation of reviewer suggestions) against the disadvantage (excessively long document which is extremely difficult to navigate). We decided to keep the responses separate. We number the individual comments below for easy referencing in this document and for cross-referencing between this document and *Response to Reviewer #1*.
- The individual reviewer comments are given in bold type-face. We follow each enumerated reviewer comment with authors' response and any changes in the manuscript. Our responses are in plain text.

Anonymous Reviewer 2:

Important results are presented; however, a major revision to paper is necessary. Significant issues with grammar and organization need to be fixed. Grammar issues are pointed out in notes and areas with yellow highlighting. The authors switch back and forth from past to present tense. Paper lacks a few keep components. More information needs to be presented on the quality assurance conducted; specifically, on how local sources were removed. The paper lacks discussion on the availability of the data set and software used. Ideally, Digital Objective Identifiers (DOI) would be given for both the data set and software. At a minimum, a description of where the data set and software is available should be given and a description of both. Without this information, the project is not repeatable. Also, it would be nice if the software was available in an open repository so that it could be reviewed and used by others. There are several places where conclusions are made that are not supported by analysis presented.

1. **Significant issues with grammar and organization need to be fixed. Grammar issues are pointed out in notes and areas with yellow highlighting. The authors switch back and forth from past to present tense.**

   <u>Authors' response:</u>

We thank the reviewer for her/his thoroughness and agree with the suggestions. We discuss paper re-organization in response to other reviewer #2 comments. We address the grammar issues here, along with suggestions regarding font size and plot/table readability.

Changes in manuscript:

We have corrected grammar issues throughout the manuscript and improved the readability of plots and tables.

(a) We apply consistent usage of present tense throughout the manuscript when discussing the current work and only use past tense when discussing previously published work
(b) We define all acronyms (ex: NOAA-ESRL) during first usage
(c) We modified sentences containing absolute phrases
(d) We broke up long sentences into shorter sentences to improve readability, suggested by the reviewer
(e) We qualified aerosol scattering coefficient in several places by adding the word 'coefficient' or using the symbol σ_{sp}
(f) We implemented multiple suggestions made by the reviewer to re-word sentences
(g) We increased the font sizes in the figures and tables and re-scaled the plot axes so that the plots can be easily viewed without magnification. We all only include one size cut (PM1) on the plots so as to reduce the number of traces and enhance readability, as suggested in reviewer #1 comment 7.
(h) We include a broken line on all plot traces in the temporal variability plots, followed by an 'ALL' data point. The 'ALL' data point gives the mean value over the entire period. It provides the reader with a way of comparing the magnitude of the temporal variability of each aerosol parameter with the mean value over the entire dataset. We also included the 'ALL' data point on plots included in the initial manuscript but interpretation was difficult, based on lack of a clear break in the line to separate the trace from the 'ALL' data point.

2. **Paper lacks a few keep components. More information needs to be presented on the quality assurance conducted; specifically, on how local sources were removed. The paper lacks discussion on the availability of the data set and software used. Ideally, Digital Objective Identifiers (DOI) would be given for both the data set and software. At a minimum, a description of where the data set and software is available should be given and a description of both. Without this information, the project is not repeatable. Also, it would be nice if the software was available in an open repository so that it could be reviewed and used by others.**

Authors' response:

We address both of the above-stated suggestions in the new *Data processing and quality assurance* section. See the detailed outline of revised paper, located at the end of this document and as a response to reviewer #1 comment 1.

Changes in manuscript:

We include the following two paragraphs at the beginning of Section 2.3.

"Software developed at NOAA is used to log the data at the sites, automatically transmit the data to NOAA, and ingest the data into the NOAA database. The database is accessible to the individual site mentors via virtual machine software. The virtual machine software includes a graphical user interface for reviewing and editing data as well as tools for extracting the data in a variety of formats and for desired averaging times. The data acquisition, processing and virtual machine software, along with documentation, are open-source and freely available from NOAA (http://www.esrl.noaa.gov/gmd/aero/sw.html). Quality-assured data products for each site in the NOAA/ESRL network are uploaded to the World Data Centre for Aerosols and made available at

http://ebas.nilu.no/Default.aspx. The data products include hourly-averaged aerosol number concentrations (not presented in this paper), σ_{sp}, σ_{bsp}, and σ_{ap} for the PM10 and PM1 size cuts.

Data quality assurance review for each site is typically performed by the site mentor on a weekly basis. Data during periods of instrument or sampling problems and during times of instrument maintenance are invalidated. Absorption data are flagged for periods when the PSAP or CLAP filter transmission drops to less than 0.70 and invalidated when the filter transmission drops below 0.50. High filter loading greatly increases the σ_{ap} measurement uncertainty (Bond et al., 1999). The lack of PSAP filter changes on weekends at SGP leads to an under-representation of quality-assured Sunday-Monday σ_{ap} hours over the period of this study Quality-assured σ_{ap} data at SGP is only available for 38% of Sunday-Monday hours during 2010-2013, versus 70-80% of the hours for the rest of the week. Weekend days with low σ_{ap} are thus well-represented at SGP while weekend days with high σ_{ap} (leading to over-loaded PSAP filters) are under-represented. The four NOAA-ESRL network sites discussed in this paper are located such that there are no major aerosol sources in the predominant upwind directions. There are some aerosol sources that are typically downwind but that can be occasionally sampled. Occasional spikes in aerosol number concentrations, σ_{sp}, σ_{bsp}, and/or σ_{ap} are flagged as local contamination by the site mentor. These spikes are usually 15-20 minutes or less in duration and often coincide with vehicular traffic near the sites or times of peak morning commuter traffic. Broader aerosol peaks are typically retained, as they are characteristic of the sampling environment of the station. One example of a broader aerosol peak not marked as contaminated is elevated σ_{ap} which often persist for hours during mornings with surface inversions or during periods with humid, stagnant air masses. "

In response to the reviewer's comment about DOI numbers for the data - the NOAA/ESRL division responsible for many of the measurements presented here has been working on this for several years, but due to issues outside our control (the joy of working in a large government agency) have yet to be able to provide data DOIs.

3. **The reviewer mentions that "there are several places where conclusions are made that are not supported by the analysis presented". The reviewer explicitly mentions "P27014 Line 20-23: While supported by other studies; these are not conclusion that can be made based on the analysis presented in this paper. Hence, the statement does not belong in the conclusion section."**

Authors' response: We have made extensive efforts in the revised manuscript to avoid making conclusions that cannot be made based on the analysis presented in this paper or at the very least, to distinguish conjecturing from supported conclusions. More often than not, the problem resulted from poor wording. We should have qualified the statements with a phrase such as "we hypothesize…". Interpretation of the results is often very difficult, given a lack of continuous aerosol chemistry measurements and measurements of trace gases and other parameters needed to draw conclusions. This necessitates that we synergize all of the available measurements and information regarding site surroundings to make hypotheses and conjectures that are the most consistent with the data. While far from complete, the NOAA-ESRL lower tropospheric aerosol measurements, combined with column-averaged aerosol measurements made at co-located AERONET sites, give the best approximations to aerosol radiative effects in the four regions.

Changes in manuscript:

See response above

4. **Abstract Line 16: I do not understand what makes scattering coefficient "Pronounced" and for example "absorption" not "pronounced" but broad. Both scattering and absorption coefficient have summer time peaks less than 50 % of the annual medium. Scattering coefficient seems to have a second peak, whereas absorption coefficient does not. Is this second peak what makes summer peak pronounced? I would suggest description the fact that there is one peak versus two peak as the difference in scattering coefficient.**

Authors' response:

We have improved clarification of the annual cycle of aerosol light scattering coefficient. We note that secondary scattering coefficient peaks are observed at SGP, BND, and EGB (but not APP). The summer scattering coefficient peaks are clearly more narrow than the summer absorption coefficient peaks (Fig.1), especially at APP and EGB. See the detailed outline of revised paper for information regarding changes in paper organization. The revised outline is located at the end of this document for convenience.

Changes in manuscript:

The first paragraph of *Sect. 4.1.1 Temporal variability common to all sites* reads as follows:

"The annual cycles of mean AOPs are larger than the weekly and daily cycles at all sites. The seasonality of nearly all AOPs is larger than both the 95% confidence intervals of monthly-mean AOPs (Fig.2) and measurement uncertainties (Table 3) indicating that nearly all annual AOP cycles are significant. Summer σ_{sp} maxima are observed at all sites (Fig.2), with steeper decreases from summer to fall than from summer to spring. The summer σ_{sp} maxima at APP and EGB are a factor of 3-4 larger than fall and spring σ_{sp} at these sites. Summer/spring and summer/fall σ_{sp} differences at BND or SGP are about half the magnitude of those observed at APP and EGB. Summer σ_{ap} maxima are also observed at all sites, although the σ_{ap} seasonal differences at SGP are not significant except for the lower σ_{ap} measured during winter. Winter σ_{ap} minima are observed at all sites, in contrast to secondary σ_{sp} peaks present during winter at all sites except APP. The annual σ_{ap} cycle amplitude is largest at BND and EGB, where σ_{ap} is 2-2.5 times larger in summer than winter The combination of a secondary σ_{sp} peaks and σ_{ap} minima lead to winter ω_0 maxima at all sites except APP. Single-scattering albedo is lowest during fall at all sites. The summer-to-fall decrease in ω_0 is largest at EGB (~0.12) and smallest at SGP (~0.04). At all sites, the fall ω_0 minima is the result of a greater autumn decrease in σ_{sp} than σ_{ap}."

5. **P26973 Abstract line 2 Should indicate that b is for sub-micrometer particles and not total b in this context. This is important to not be confused with a shift in R which indicates more super-micrometer particles**

Authors' response:

The b values and nearly all other aerosol optical properties that we report in the revised manuscript are for PM1. We implemented the suggestion made in reviewer #1 comment 7 that we choose one size cut to report, in an effort to reduce excessive information on plots and to minimize redundancy. The exceptions are PM1 scattering fraction (R_{sp}) and PM10 scattering Angstrom exponent. The PM1 and PM10 values for b are not statistically-different in most cases. This is not surprising, given that b for visible wavelengths is more sensitive to smaller particles (Collaud-Coen 2007) and that the total aerosol at the four sites is dominated by sub-micron aerosol (Rsp > 0.8 or 0.9) .

Changes in manuscript:

We note the size cuts discussed during the first mention of size cuts in the manuscript. We now report PM10 aerosol optical properties (AOPs) in the Supplemental Materials.

6. Abstract line 5 Please state what systematic relationship had seasonal changes

Authors' response:

The relationships involving absorption Angstrom exponent exhibited seasonal changes. We qualify this in the revised manuscript.

7. P. 26976 Line 25-27: This second question cannot be answer directly from the measurements listed. Suggest deleting this question.

Authors' response:

We have re-worded some of the paper objectives, based on (i) this comment by reviewer #2; (ii) proxies suggested in reviewer #1 comment 4; and (iii) more appropriate trend analysis suggested in reviewer #1 comment 2. The elimination of second sentence of objective 2 (see below) in the initial manuscript specifically addresses the above-stated suggestion.

Changes in manuscript:

The objectives in the initial manuscript were stated as follows:

"Measured and derived AOPs are used to answer the following research questions:

1. *How do key lower tropospheric aerosol optical properties differ among the four North American continental regions and how do they vary on different timescales (seasonal, weekly, diurnal) for each region? What do the observed spatial and temporal differences imply in terms of dominant sources and processes?*
2. *How have the magnitude and variability of aerosol optical properties changed at the long-term sites (SGP and BND) since D&O2002? What does this imply in terms of possible changes in sources and processes in these regions?*
3. *Are there systematic relationships between these optical properties? How do these relationships vary with region and with season? "*

The objectives in the revised manuscript are stated as follows:

"*Measured and derived AOPs are used to answer the following research questions:*

1. *How do key lower tropospheric AOPs differ among the four North American continental regions and how do they vary on different timescales (seasonal, weekly, diurnal) for each region?*
2. *Is the temporal variability in AOPs at each site consistent with proxies related to regional aerosol sources and meteorology?*

3. *Do any statistically-significant trends exist in AOPs measured at the two long-term sites (SGP and BND)?*
4. *How do relationships between AOPs vary with region and with season? What information can these relationships provide regarding aerosol sources and processes? "*

8. P26980 Line 2: Please provide a reference or delete since I don't believe this. I believe that a very small percentage of people use wood for heating.

Authors' response:
We have provided a reference and percentages of households in Wautaga County that use wood-burning stoves. The numbers in the surrounding rural communities are difficult to obtain but the smell of wood burning is obvious during the winter, regardless of location and especially so if one visits any of the surrounding communities. While a relatively small percentage of households, these uncontrolled emissions can have a large effect.
Changes in manuscript:
We have clarified the sentence as *"Wood-burning stoves serve as the primary heating source for 6.2% of occupied housing units in Watauga County (U.S. Census Bureau, 2010) and likely a larger percentage of housing units in the surrounding rural mountain communities."*

9. **Reviewer 2:** P26983 Line 4+++: This paragraph should be moved. Suggest things be organized slightly differently. This paragraph is about data processing; likewise, paragraph below are about data processing. Hence suggest a "Data processing" sub-section.

Authors' response:
We agree with this and other comments by both reviewers suggesting re-organization of some material. The structure of the revised manuscript is provided in our response to reviewer #1 comment 1.
Changes in manuscript:
We moved the paragraphs on P.26983 of initial manuscript to a new section (*Sect. 2.3 Data processing and quality assurance)*.

10. P26984 Line 12: Why name this section Data consistency? I don't understand the term in this context. It is data quality assurance that is discussed as indicated by the first sentence. Should calling this section "Data quality assurance" or "Quality assurance".

Authors' response:
We re-organized the manuscript, based on multiple suggestions by both reviewers (including this suggestion). The revised manuscript outline is provided as response to reviewer #1 comment 1 and is also included at the end of this document.
Changes in manuscript:
We eliminated the *Sect. 2.5-Data Consistency* section in the original manuscript. We moved the material on P. 26984 Lines 13-22 and the material in the first paragraph of P. 26985 to Section 2.3- *Data processing and quality assurance.* We moved the material on P.26984 Line 21- 28 to *Section 2.5-Data analysis methods.* We also moved other similar content to these sections in order to better organize the manuscript, as suggested by both reviewers.

11. I believe more information is needed about how local sources were removed. How much data does this affect? Was the quality assurance consistent between the two time periods? If data was removed from the later period and not the first, then this could result in the observed decrease. Please provide information to illustrate that this is not the case.

Authors' response:
We implemented the above-stated suggestions, as outlined in the following sentences.
Changes in manuscript:
We implemented reviewer 1's suggestion (her/his comment 2) for a more suitable trend analysis at SGP and BND, in place of the inter-period comparisons (current period versus that reported by D&O2002). Details of the trend analysis are provided in our response to reviewer #1 comment 2. We include information regarding removal of local sources in Section 2.3- *Data processing and quality assurance.* See response to reviewer #2 comment 2.

12. **P. 26986 Line 2++: I don't believe the median is any better than the mean at reducing effects of outliers. Something like a trimean would do this. The median is better for comparisons with satellite measurements.**

Authors' response:

Multiple suggestions by reviewer #1 regarding significance of reported variability lead us to reporting aerosol optical properties as means and 95% confidence intervals of the mean values. The scattering and absorption coefficients are clearly better-approximated by a log-normal distribution so using means and standard deviations is not appropriate for these variables. Most of the intensive AOPs are suitably-approximated by a normal distribution, both for this site and for those reported by others (Collaud-Coen, et al., 2013). Means and standard deviations are then suitable statistical parameters for these variables.

Changes in manuscript:

To facilitate estimates of the significance of seasonal and regional differences in AOPs (suggested in Reviewer #1 comment 3), we switched to the use of means and 95% confidence intervals of the means.

13. **P. 26986 Line 12: Are the intensive properties calculated from the hourly averages of scattering, backscattering and absorption, or are intensive properties calculated for sub-hourly measurements and averaged? The first is what is implied; however, D&O2002 did the second.**

Authors' response:
The intensive properties are calculated from the hourly averages of scattering, backscattering and absorption. Implementing identical methods to D&O2002 is no longer necessary since we implemented Reviewer 1's suggestion for a more suitable trend analysis at SGP and BND (her/his comment 2), in place of the inter-period comparisons (current period versus that reported by D&O2002).

14. P26989 **Line 20-25: What about seasonality of AOD at the other locations? It would seem that comparing the seasonality of surface measurements with the seasonality of AOD would indicate if observed changes are due to changes in sources/sinks or changes in meteorology such as lower boundary layer heights trapping more/less aerosols.**

Authors' response: The reviewer's suggestion is a good one and we likely the topic of an upcoming paper. However, a comparison of the seasonality of surface measurements with the seasonality of AOD lies outside the scope of this already-lengthy paper. The mention of the large SE U.S AOD seasonality Goldstein (2009) was simply an example of another study in the region which yielded similar variability, even if near-surface and column-averaged results are not directly comparable.

15. **P26990 Line 20-25: Something is wrong here. Single scattering albedo is ~0.9, are we talking about backscatter or co-albedo? Please fix.**

Authors' response:

We agree with the reviewer the wording is confusing in places and we make efforts in the revised manuscript to improve clarity. We are not referring to backscatter or co-albedo. We are talking about the annual cycle of single-scattering albedo, which we defined in lines 5-7 of P.26986 as "*Temporal variability on each of the timescales was defined for the purposes of this study as the amplitude of the cycle of median values (difference between maximum and minimum values).*" The 0.13 is the difference in single-scattering albedo between the month with highest median single-scattering albedo and the month with the lowest median value.

Changes in manuscript:

We still include the following disclaimer in Sect. 2.5-Data analysis methods: "*We define the magnitude of temporal variability on each of the timescales as the amplitude of the cycle of mean values (difference between maximum and minimum values)*". In addition, we also attempt to better clarify what differences we are referring to. We use language such as "summer-to-winter differences in ω_0 at APP are ~0.06" and "The annual σ_{ap} cycle amplitude is largest at BND and EGB, where σ_{ap} is 2-2.5 times larger in summer than winter" when quantifying the variability.

16. **Line 25-29: Could you define what size range is being talked about here for "lower end"? The accumulation mode could be from 30 - 1000 nm and peak at say 100-150 nm. Are we talking about the 30-80 range or the 100-200 range? What is lower end?**

Authors' response:

This is a good point and one that we more clearly articulate, as it was poorly-worded in the initial manuscript. We are referring to the lower size range of optically-important accumulation mode particles (~100-300nm).

Changes in manuscript:

We clarify the size range as follows in *Sect. 4.1.1-Temporal variability common to all sites*:

"The stronger relationship between the annual b and σ_{sp} cycles (relative to relationships between the cycles of σ_{sp} with either α_{sp} or R_{sp}) suggests that the major seasonal changes in the aerosol size distributions at APP, BND, and SGP may lie at the smaller end of the range of optically-relevant accumulation mode particles (100-300nm), with shifts toward larger particles in summer and smaller particles in fall."

17. **P.26992 Line 25-28: Zhang et al., 2010 does not conclude this! Reviewing the Zhang et al., 2010 abstract indicates that wood burning contributes significantly to aerosols in the winter in the SE US, not that it is a "common residential heating fuel". Please correct and review all references to ensure that statement correspond to what is presented in the referenced paper.**

Authors' response:

From the bottom of 6th page of the Zhang 2010 paper: "*In winter, biomass burning was likely mainly in the form of residential wood burning (discussed in more detail below), thus spatial variability at these time periods likely reflect population densities and frequency of burning in different regions.*." Zhang then goes on to discuss this.

Changes in manuscript:

We modify the wording in *Sect. 4.1.2-Temporal Variability at APP* to make our meaning more clear:

"The influence of smaller, darker particles (larger b and smaller ω_0) and PM1 α_{ap} values of 1.3-1.4 during colder months is consistent with a mix of black and brown carbon. The hypothesis of a brown carbon influence (as opposed to dust) on winter σ_{ap} at APP is based on (1) R_{ap} values of ~0.90-0.95 (Fig. S1); (2) a biomass-burning organic aerosol factor in the winter aerosol mass spectra measured at APP (Fig. S2 of Link et al., 2015); and (3)

residential wood-burning during winter in the local community (U.S. Census Bureau, 2010) and in the SE U.S. (Zhang et al., 2010)."

18. P26997 Line 12-13: I do not understand how convection would increase regional transport. In the summer, transport would take longer time because of lower wind speeds and larger boundary layer heights.

Authors' response:

The statement on P. 26997 of the original manuscript is a hypothesis: "T*he step-like increase in σ_{sp} on Wednesday and higher values through Saturday is thus consistent with regional emissions during Monday–Friday. The fact that this occurs primarily during the warm season could be due to convectionally-driven increases in regional transport*". We are basing the hypothesis (or conjecturing) on a limited set of measurements made from a single site, along with known local or regional traffic or industrial patterns matching the observed weekly σ_{sp} or its seasonal dependence. We agree with the reviewer that summer is characterized by lower wind speeds and by larger boundary layer heights, both of which are measured at APP. APP is a high-elevation site by eastern U.S standards (~1080m asl). The regional transport is from lower elevation sites. Colder months are characterized by more stable air (inversion layers and general lack of convection). We see this at APP in the form of lower PBL heights and less frequent observation of elevated aerosol layers (measured by co-located micro-pulsed lidar). The increased stability acts as a lid in preventing the transport from these lower elevation sources up the mountain.

19. Line 24-25: Agricultural activities may not have a weekly cycle.

Authors' response:

This comment is correct. We are not stating that a weekly cycle in agricultural activities exists and in fact we are not aware of any such cycle. The sentence "*The minimum median Rsp at SGP occurred on Tuesday for all seasons, possibly due to some weekly pattern in agricultural activity near the site*" acknowledges that this could be a possibility because we do not know any other possible reason for a Tuesday minimum.

20. P27000 Line 15++: Would you really see a boundary layer effect due to the long life time of aerosols compared to that of a day. If the aerosol life time is several days, would this not make it unlikely to see a small boundary layer effect change? Likewise for local traffic.

Authors' response:

Our wording in this paragraph likely does not reflect our intent. What we meant to state is that a more shallow morning boundary layer would lead to a trapping of aerosols near the surface (leading to larger measured scattering and absorption coefficients) and that a more elevated boundary layer would lead to lower scattering and absorption coefficients. Since PBL heights typically possess a diurnal dependence similar to solar heating, a boundary layer influence on measured near-surface aerosol loading would be expected to possess a diurnal cycle that is closely related (inversely) to PBL heights.

Changes in manuscript:

We do not include the material on P. 27000 Line 15++ in our revised manuscript. Following the suggestion of reviewer #1 (comments 4, 5), we use proxies such as local or regional PBL climatologies to help explain the diurnal and seasonal cycles. We removed content such as that on P27000 in order to add the proxies, while at the same time attempting to shorten the paper (reviewer #1 comment 5).

21. **P27001 Line 25-28: Could this not be due to a different life time for absorbing aerosols compared with scattering aerosols?**

Authors' response:
Both the scattering and absorption exhibit broad afternoon minima at BND and SGP. Lines 5-8 of page 27001 describe the same diurnal cycle for scattering as that for absorption in lines 25-28. The diurnal cycle for absorption is larger than that of scattering but we do not see how the cycles presented could result from different lifetimes of scattering and absorbing aerosols. We do not dismiss the suggestion that scattering and absorbing aerosols could have different lifetimes.

22. **P27002 Line 23-25: Without a direct comparison between the measurement and the height of the boundary layer I don't see only you can conclude that there is an influence. While this could be the case, I believe you would have to do the comparison or leave it to future work.**

Authors' response:
We have implemented the suggestion in reviewer #1 comment 4 that we use proxies including PBL heights to help explain aerosol temporal variability at the four sites. We now report results from PBL height climatologies based at/near the stations (for APP and SGP) or from locations within 100-200km of the stations (for BND and EGB). We realize that the reported PBL heights cannot facilitate direct comparisons (correlation or other) with the in situ aerosol measurements. However, the reported PBL climatology results can be used along with other proxies (pollution-rose diagrams, etc) help explain the seasonal and diurnal cycles in mean aerosol optical properties (as suggested by reviewer #1).

Changes in manuscript:

We include the following in the revised manuscript (*Sect. 3-Site Descriptions*):
In *Sect 3.1 APP*

"*Planetary boundary layer (PBL) calculated from vertical aerosol backscatter profiles retrieved by a micro-pulse lidar at APP from 2/2013-8/2014 reveal a relatively weak diurnal and seasonal dependence of PBL heights. Median afternoon (morning) PBL heights are 920m (820m) in winter, 1200m (880m) in spring, 1100m (850m) in summer, and 1050m (680m) in fall.*"

In *Sect 3.2 BND*
"*Holzworth(1964) used daily soundings at Joliet, IL.(located ~130km NNE of BND) to calculate monthly mean mixing layer heights illustrating their seasonal dependence: DJF(480m, 480m, 4080m); MAM(980m, 950m,1040m); JJA(1090m,1380m,1310m); SON(860m, 790m, 600m).*"

In *Sect. 3.3 EGB*

"Mixing layer heights were reported by Portelli et al. (1981) for Buffalo, NY, located ~170km SE of EGB. The mixing layer height frequency distributions reported by Portelli et al. (1981) indicate that mixing layer heights are above 1km for <10% of the time during winter, 50% in spring, 70% in summer,and ~35-40% in fall."

In *Sect. 3.4 SGP*
"*The large annual temperature cycles at SGP result in strong seasonality in mixing layer heights and their diurnal variability. Median mixing layer heights are less than 100m during morning for all seasons and median afternoon mixing layer heights are 752m in winter, 1360m in spring, 1640m in summer, and 1390m in fall (DelleMonache et al., 2004).*"

23. **P27002 Line 26: I don't see any evidences of direct local traffic influence. Furthermore, the quality assurance section indicates that local traffic spikes are removed from the data set analyzed.**

Authors' response:

We have better-clarified our meaning of "local traffic spikes are removed from the data set analyzed". The 2-3 hour duration morning σ_{ap} peaks at APP and EGB (revised manuscript Fig.3) are characteristics of the sampling environment of the station and are not removed. One example of a broader aerosol peak not marked as local contaminated is elevated σ_{ap} which often persist for hours during mornings with surface inversions or during periods with humid, stagnant air masses. We believe that the fall EGB absorption maxima near 8am LST is likely due to traffic for the following reason: There is a minimum in absorption just two hours early (5-6am LST), followed by a rapid increase leading to a maxima at 8am, which is then followed by another decrease. This does not represent a pattern influenced by solar heating (which PBL height typically follows) or by any other known regional pollution sources or transport patterns.

Changes in manuscript:

In response to reviewer #2 comment 2, we include a new section (*Sect.2.3-Qualtiy assurance and data processing*) in the revised manuscript. The section provides information on the quality assurance conducted, including how local sources are removed. We include the following passage in Sect. 2.3:

"*Occasional spikes in aerosol number concentrations, σ_{sp}, σ_{bsp}, and/or σ_{ap} are flagged as local contamination by the site mentor. These spikes are usually 15-20 minutes or less in duration and often coincide with vehicular traffic near the sites or times of peak morning commuter traffic. Broader aerosol peaks are typically retained, as they are characteristic of the sampling environment of the station.*"

24. P27006 Line 23: I don't understand why pollution transport is the likely cause instead of other possible reasons.

Authors' response:

We understand the reviewer's questioning why pollution transport is the likely source of the high aerosol loading events at EGB, based on the materials supplied in the first manuscript draft. Pollution-rose diagrams included in the revised manuscript (see below) show a clear association between southerly wind directions and larger values of σ_{ap} and σ_{sp} measured at EGB. Southerly wind directions correspond to air masses arriving at EGB from the heavily-populated southern Ontario region, including Toronto (~70km south of EGB).

Changes in manuscript:

We now include pollution-rose diagrams for each season at each site. The pollution-rose diagrams show the dependence of absorption coefficient on wind direction for each season and site. (Fig(s).4-7). We use the pollution-rose diagrams as context for interpreting aerosol temporal variability at the four sites (Sect(s) 4.1.2-4.1.5). We include the pollution-rose diagram for absorption coefficient because absorption coefficient demonstrates the largest variability of all aerosol optical properties on weekly and diurnal time scales. We also include pollution-rose diagrams for the following aerosol properties for each site/season in Fig(s) S10-S25 of Supplemental Materials: (1) scattering coefficient; (2) PM1 scattering fraction; (3) single-scattering albedo; (4) hemispheric backscatter fraction. The pollution-rose diagrams for σ_{ap} and σ_{sp} measured at EGB are provided Fig.6 of revised manuscript and Fig.S18 of *Supplemental Materials*, respectively.

25. Line 25-26: Is this really traceable to field burning or just burning?

Authors' response:

We no longer include the seasonal dependence of systematic relationships among AOPs, with exception of those involving absorption Angstrom exponent. We modified this section so as to shorten the paper and emphasize new results, in response to reviewer #1 comment 5. Most of the systematic relationships for individual seasons are well-approximated by the annual curves so we now only show annual relationships for these aerosol properties in Sect. 4.4.1

Change in manuscript:

This statement does not appear in the revised manuscript

26. P27009 Line 8-9: Can you give percentage here?

Authors' response:

We do not understand which percentages the reviewer is referring to. We have also removed the seasonal systematic relationship curves to which the reviewer is referencing (see below).

Changes in manuscript:

In response to reviewer #1 comment 5, we have shortened the manuscript to enhance the focus on new results and to minimize redundant results. One of the changes involved eliminating the seasonal plots showing systematic relationships among most aerosol optical properties (Sect. 4.4 of revised manuscript). Only the relationships involving absorption Angstrom exponent vary with season at the sites. The relationships among other aerosol optical properties demonstrate little seasonality so only annual curves are reported for these properties. We now only include annual curves for all other aerosol optical properties. We also shortened the discussion of some of these results, which are similar to those reported for BND and SGP by Delene and Ogren (2002) and by Andrews et al.(2011).

27. Line 16: Do you not mean fine mode average particle size?

Authors' response: No. Scattering Angstrom exponent is not an indicator of fine mode average particle size for bi-modal distributions but is a better indicator of the relative amounts of coarse and fine mode aerosol volume (e.g., Shuster et al., 2006). Our systematic relationship curves of PM1 scattering fraction versus scattering Angstrom exponent at APP, BND, and SGP are also consistent with SAE being an indicator of the relative contribution of fine-mode aerosol to scattering.

Outline of revised paper and Supplemental Materials document

Changes in manuscript:

<u>The revised manuscript is structured as follows:</u>

Section 1-Introduction

Section 2-Methodology

2.1 Air sampling infrastructure at the sites

2.2 Measurements and instruments

2.3 Data processing and quality assurance

- Section added to consolidate material spaced over several sections and to add quality assurance information suggested by reviewer #2 comment 2

2.4 Measurement uncertainties

- Short section added in response to reviewer#1 comment 3 below. A much more-detailed treatment is provided in *Section S1 of Supplemental Materials*

2.5 Data analysis methods

- Section added to consolidate data analysis material.

Section 3-Site descriptions

- This section contains a more in depth description of the sites, in response to reviewer #1 comment 1. Information regarding aerosol types measured at site, known regional pollution sources, and results from PBL climatologies near the sites are given (to extent available)

3.1 Appalachian State University, Boone, North Carolina, USA (APP)

3.2 Bondville, Illinois, USA (BND)

3.3 Egbert, Ontario, Canada (EGB)

3.4 Lamont, Oklahoma, USA (SGP)

Section 4-Results and discussion

- Temporal variability in PM 1 aerosol optical properties (AOPs) are presented, with exception of PM10 scattering Angstrom exponent and the PM1 scattering and absorption fraction. We've relegated results for the other PM10 AOPs to the *Supplemental Materials*. Most of the temporal variability and systematic relationships among PM10 AOPs is similar to that of PM1 AOPs and the use of PM1 properties for APP, BND, and SGP better-facilitated comparisons with EGB (where only PM1 AOPs are measured). We implemented the suggestion made by reviewer #1 comment 7 to minimize redundancy by picking eitherPM10 and PM1 and using it throughout the paper.

4.1 Temporal variability of aerosol optical properties

- Annual, weekly, and diurnal cycles of mean AOPs are reported and discussed. We now include discussion of the three cycles in the same section to provide a more coherent picture of sources and processes influencing the variability at the four sites.

4.1.1 Temporal variability common to all sites

- We discuss variability common to all or most sites, so as to minimize repetition. Site-specific variability is discussed in sections 4.1.2-4.1.5. Proxies such as wind direction, pollution sources and PBL heights(to extent available) are discussed for each site in those sections, in response to reviewer #1 comment 4

4.1.2 Temporal variability at APP

4.1.3 Temporal variability at BND

4.1.4 Temporal variability at EGB

4.1.5 Temporal variability at SGP

4.2 Regional variability of aerosol optical properties

- Short section reporting regional differences in AOPs. The section is placed here in response to the suggestion (reviewer #1 comment 1) that we first include complete descriptions of each site, followed by a comparison between regions

4.3 Long-term aerosol optical property trends at BND and SGP

- This section replaces all of the long-term comparisons for BND and SGP that appeared in Sections 3.1-3.3 of initial manuscript with a short summary of statistically-significant long-term trends at SGP and BND. We've implemented the approach suggested by reviewer #1 comment 1 for a more statistically-relevant method for estimating trends. More detail is provided in response to reviewer #1 comment 2.

4.4 Systematic relationships among aerosol optical properties

- We modified this section so as to shorten the paper and emphasize new results, in response to reviewer #1 comment 5. Most of the systematic relationships for individual seasons are well-approximated by the annual curves so we now only show annual relationships for these aerosol properties in Sect. 4.4.1
- Relationships involving absorption Angstrom exponent (AAE) do demonstrate seasonal dependence. The relationships during summer at APP, BND, and SGP and during winter at APP are different than the annual curves. We thus report relationships for individual seasons in Sect. 4.4.2

4.4.1 Annual systematic relationships among aerosol optical properties

4.4.2 Seasonal relationships involving absorption Angstrom exponent

Section 5-Summary and conclusions

<u>The Supplemental Materials document is structured as follows:</u>

Section S1-Measurement uncertainties

- Detailed uncertainty analysis, including contributions of individual sources, propagation of uncertainties (including covariance between AOPs), and calculation of total and measurement precision uncertainties, following an approach similar to Anderson and Ogren (1998) and Anderson et al. (1999). The results of the uncertainty analysis relevant to the comparisons of mean AOPs in the paper are summarized in Sect. 2.4 of the paper.

S1.1 Uncertainties in total and hemispheric backscatter coefficients

S1.2 Uncertainties in absorption coefficient

S1.3 Uncertainties in calculated aerosol optical properties

S1.4 Comparing measurements made at different locations and times

Section S2-Statistics related to quality assurance claims made in the paper

- Table contains annually-averaged values of PM10 and PM1 geometric mean */ geometric standard deviations (scattering and absorption coefficients) and arithmetic mean± standard deviations (intensive AOPs) for all sites.

Section S3-Justification for inclusion of aerosol light scattering measurements at EGB

- The EGB aerosol sampling system does not include active control of RH so we include brief discussion to show that the moderately-elevated summer RH values at EGB do not alter the results presented in the paper. The argument is based on scattering hygroscopic growth factors measured at APP and frequency distribution of nephelometer RH at EGB.

Section S4-Annual cycles of PM10 aerosol optical properties

- We include this for completeness, since we no discuss primarily PM1 AOPs in the paper (reviewer #1 comment 7)

Section S5-Weekly and diurnal cycles of aerosol light scattering coefficient and intensive AOPs

- Other than a few exceptions, AOPs demonstrate little variability on weekly and diurnal scales. We reference this section in discussing a few cases of moderate variability and include the weekly and diurnal cycles for all AOPs for completeness.

Section S6-Pollution-rose diagrams for PM1 aerosol light scattering coefficient and intensive AOPs

- Pollution-rose diagrams showing the dependence of wind direction on aerosol light scattering coefficient, single-scattering albedo, PM1 scattering fraction, and hemispheric backscatter fraction for each season at each site (Supplemental Figs. S10-S25). Pollution-rose diagrams for absorption coefficient are included in the paper (Figs. 4-7) because absorption coefficient demonstrates the greatest diurnal and weekly variability.

Section S7-Table of annually-averaged PM10 and PM1 AOPs

- Table contains annually-averaged values of PM10 and PM1 geometric mean */ geometric standard deviations (scattering and absorption coefficients) and arithmetic mean± standard deviations (intensive AOPs) for all sites.