

Authors' Response

We sincerely thank the two anonymous reviewers for their excellent suggestions, particularly those related to improvements in paper organization, suggestions for the use of proxies to help explain temporal and regional aerosol variability, and uncertainty analysis. 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. The revisions were too extensive to submit a marked-up copy, as discussed with Copernicus Editorial Support. We include a very detailed outline of the revised manuscript in response to reviewer #1 comment 1 below. 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. We are confident that the improved science and readability of the revised document justifies the extent of the changes made.

We respond to reviewer comments in the space below. We number the individual comments for easy referencing. The **responses to reviewer #1 are given first and are in black font.** The **responses to reviewer #2 are in red font.** The color-coding is necessary, given the large number of comments from both reviewers. We also enumerate the individual comments from each reviewer for easy referencing. 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. 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.

Reply to Reviewer #1 (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.)

Anonymous Reviewer 1:

This paper presents a 4 years climatology of extensive and intensive aerosol parameters as well as a direct radiative forcing efficiency estimation at four stations of the USA. For two of the four sites, a trend analysis is done by comparing the annual, weekly and diurnal cycles with the ones of the 1997-2000 period published in 2002 by Delene and Ogren. The systematic relationships among extensive and intensive aerosol properties are analyzed in order to constrain models parametrization of aerosol optical properties and to make assumption about aerosol sources and processes. The methodology of the climatology analysis is similar to the one used by Delene and Ogren (2002) and the one of relationships among aerosol optical properties to the Andrew et al. paper (2011). Comparison between the sites are also presented.

- 1. The result section is organized as follow: 3.1 seasonal cycle, 3.2 weekly cycle, 3.3 diurnal cycles and 3.4 systematic relationships among aerosol optical properties. In each of these section, the cycle is described for each station, comparing also PM10 and PM1 results, then the spatial variability (difference between stations) is described and the long term temporal variability (1997-2000 period towards 2010-2013 one) is presented. The weekly and diurnal cycle as well as the systematic**

relationships are also presented per season. This structure induces a lot of repetitions of the same/similar information and does not allow the reader to get the main results concerning each station and the spatial variability between the stations. It is also quite difficult to check if all the various information, for example concerning one station, are coherent through the paper. I think that a complete description of each station, followed by a comparison between the stations would greatly improve the paper.

Authors' Response:

We appreciate the reviewer's suggestion. We agree that the manuscript contained a lot of repetition. To this end, we have made major revisions to the paper organization, with a primary goal of minimizing repetition and providing the most coherent picture of each site and the variability between sites. We compared several different paper structures, including that suggested above by reviewer #1. Another goal was to implement as many of the very good suggestions from both reviewers as feasible into this structure. Based on these comparisons, we have implemented the paper organization summarized below. The changes to the manuscript were very extensive and required moving, consolidating, adding, re-phrasing, and deleting material. We have also considerably expanded the *Supplemental Materials* submitted along with the paper so as to include a detailed uncertainty analysis and other items suggested by the reviewers, while still keeping the manuscript as short as possible.

Changes in manuscript:

The revised manuscript is structured as follows:

Section 1-Introduction

- Most of this section is similar to the first manuscript draft. We did modify the stated objectives and the differences between our paper and D&O2002 to be consistent with changes made to the document (described in our responses to reviewers)

Section 2-Methodology

2.1 Air sampling infrastructure at the sites

- nearly identical to original manuscript

2.2 Measurements and instruments

- nearly identical to original manuscript

2.3 Data processing, quality assurance, and calculated AOPs

- Section added to consolidate material spaced over several sections and to add quality assurance information suggested by reviewer #2 comment 2. This section also includes justification for including SGP absorption data for the 2010-2013 years (but not for the trend analysis), in response to reviewer #1 comment 22.

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*, including propagation of uncertainties.

2.5 Data analysis methods

- Section added to consolidate data analysis methods

2.5.1 Temporal cycle analysis

- We briefly outline the basic techniques for studying seasonal, monthly, weekly, and diurnal variability. We also define the criteria for statistical significance of differences in AOPs

2.5.2 Meteorological analysis

- Paragraph outlining the meteorological products used to interpret temporal variability, including wind sector, temperature, and published PBL heights for each region.

2.5.3 Significance and trend analysis

- Paragraph outlining the method used to quantify regional AOP variability (Sect. 4.2) and the method for trend analysis of AOPs at BND and SGP (Sect. 4.3)

2.5.4 Systematic relationships

- Paragraph outlining the technique for systematic relationships reported in Sect. 4.4

Section 3-Site descriptions

- This section provides the context for results presented in Section 4. It was expanded in response to reviewer #1 comment 1. The descriptions for each site includes: (1) site location; (2) known regional pollution sources; (3) reported aerosol chemistry for the sites; (4) seasonality of temperature and rainfall (i.e. max/min months); and (5) results from PBL climatologies near the sites (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 either PM10 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 and to check for consistency with reported aerosol chemistry for the sites and meteorology at the 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. We include a short paragraph comparing our systematic relationships with those presented for mountain sites by Andrews et al. (2011), in response to reviewer #1 comment 30.
- 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

- This section is modified to reflect new results (based on the use of proxies suggested by Reviewer 1). We also have gone to great lengths to verify that all conclusions are supported by results, in response to reviewer #2 comment 3.

The Supplemental Materials document is structured as follows:

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 containing the percentage of hours for each month at each site for which $PM1 \sigma_{sp} \leq 1 Mm^{-1}$. Intensive AOPs are not calculated for these hours, so as to avoid noise resulting from ratios of two small numbers. Delene and Ogren (2002) and Anderson et al. (2011) have also employed this approach.

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

- The EGB aerosol sampling system does not include active control of RH. We include brief discussion to show it is unlikely that the moderately-elevated summer RH values at EGB alter the reported σ_{sp} cycles at EGB. The argument is based on scattering hygroscopic growth factors measured at APP and frequency distribution of nephelometer RH at EGB.
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Section S4-Annual cycles of PM10 aerosol optical properties

- We include boxplots of monthly-binned $PM1 \sigma_{sp}$ and σ_{ap} for all over the entire 2010-2013 period and over individual years. The boxplots complement the mean values presented in the paper by showing the σ_{sp} and σ_{ap} distributions. They also illustrate that the monthly-geometric mean values presented in the paper are by and large unbiased by individual years.

Section S5-Temperature dependence of scattering coefficient and single-scattering albedo

- We include boxplots of temperature-binned σ_{sp} and ω_0 and correlation of warm-season σ_{sp} with temperature as supporting evidence for claims in the paper regarding aerosol types and possible processes

Section S6-Annual cycles of PM10 aerosol optical properties at APP, BND, and SGP

- We include this for completeness, since we now discuss primarily $PM1$ AOPs in the paper (reviewer #1 comment 7)

Section S7-Weekly and diurnal cycles of $PM1$ scattering and absorption fractions 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 S8-Pollution-rose diagrams for R_{sp} , ω_0 , and b at APP, BND, EGB, and SGP

- Pollution-rose diagrams showing the dependence of wind direction on σ_{sp} and σ_{ap} for individual seasons are included in the paper. The dependence of R_{sp} , ω_0 , and b on wind sector is also used to help interpret the annual AOP cycles at the sites. Section S7-Table of annually-averaged $PM10$ and $PM1$ AOPs

Section S9-Table of annually-averaged AOPs

- We include a table contains annually-averaged values of $PM10$ and $PM1$ geometric mean */ geometric standard deviations (scattering and absorption coefficients) and arithmetic mean \pm standard deviations (intensive AOPs) for all sites.

Section S10- Annual cycles of temperature and relative humidity at APP, BND, EGB, and SGP

- Boxplots of monthly-binned temperature and relative humidity over the entire 2010-2013 period.

2. **Trend analysis:** Two stations (SGP and BND) have long-term measurement allowing trend analysis to be performed. As trend analysis, the authors choose to compare the medians of the aerosol parameters for the 1996(7)-2000 period to the ones of the 2010-2013 period. I do not consider this method as valuable for trend analysis for the following reasons: 1) special cases and extremes such as very hot or cold seasons cannot be statistically screened by a four years median. As an example, these was probably quite unusual meteorological situation in one of the 1997-2000 February and December months at SGP (see scattering annual cycle, Fig. 3). 2) the comparison of the 2 extremes in time (beginning and end of the measuring period) does not allow to make any assessment about what happen in between. This method considers as prerequisite that changes in the 2000-2010 period are continuous, what is not proved. Measurements at SGP and BND were continuously performed from 1997 to nowadays, so that various statistically relevant techniques could be used to estimate long-term trends.

Authors' Response:

We agree with the reviewer's suggestions and have made several changes to address this concern.

Changes in manuscript:

We removed all sections containing inter-period comparisons for BND and SGP. The sections removed from the first manuscript are *Sect. 3.1.3-Influence of time period studied on annual cycles of aerosol properties at SGP and BND* (p, 26993-26996), *Sect 3.2.2- Influence of time period studied on weekly cycles of aerosol properties at SGP and BND* (p, 26999-27000), and *Sect 3.3.2- Influence of time period studied on diurnal cycles of AOPs at SGP and BND* (p, 27004-27005). We replaced these sections with a single section (Sect. 4.3) that examines long-term trends in the monthly-averaged aerosol optical properties at SGP from 1997-2013 and at BND from 1996-2013. We apply the Mann-Kendell test for significance of trends and the Sens-Slope test for trend magnitudes (% change per decade). We report the trend test results in Table 6, along with a ~2-page summary of results and comparison with results reported by Collaud-Coen et al., 2013. This modification also satisfies Reviewer 1's suggestion (comment 5) that we place greater emphasis on results from the 2010-2013 period of this study and that we shorten the paper.

3. **Uncertainties analysis:** No estimation of the uncertainties of the aerosol optical parameters are given in the paper. Such estimates are necessary to estimate if the described differences, for example between seasons for a site or between sites, are statistically relevant. For example, Anderson and Ogren (1998) gave a complete uncertainty analysis of the scattering and backscattering coefficients. Applying error propagation methods, these uncertainties lead to very large ones for b involving to consider with caution the trend analysis or the seasonal differences for this parameter. If a complete uncertainties analysis could not be performed, percentiles should at least be given and discussed.

Authors' response:

We now include an uncertainty analysis (outlined below). The uncertainties in b are actually not large, when co-variances between aerosol optical properties are considered. A detailed uncertainty analysis is now included in *Section S1 of the Supplementary Materials*. The total measurement uncertainty $\Delta b = 0.0036$ (0.0032) for PM₁₀ (PM₁) size cut when aerosol optical property (AOP) values approximating annual-average AOP values at the four sites are used to calculate Δb . The uncertainty Δb is reduced due to the high correlation between scattering and hemispheric backscatter coefficients ($r \sim 0.99$ at APP), which necessitates inclusion of the co-variance term when calculating Δb . Some of the sources of nephelometer uncertainty do not need to be considered when making comparisons made at different sites/times using identical instruments and protocols (see Anderson and Ogren, 1998 and Anderson, et al. 1999).

The total uncertainty is thus reduced further to 0.0015 (0.0016) when comparing differences made by instruments using the same instruments and protocols

Changes in manuscript:

We have added a short section (*Section 2.4-Measurement uncertainties*). This section contains a table of uncertainties of measured and calculated PM10 and PM1 aerosol optical properties (Table 3), based on 1-hour averaging and using approximate annual-mean values at the sites $\sigma_{sp}=30\text{Mm}^{-1}$, $\sigma_{sp}=3.0\text{Mm}^{-1}$, $b=0.14$, $\omega_0=0.91$, $\alpha_{sp}=2.0$, and $\alpha_{ap}=1.0$. (Table 3). We refer the reader to *Sect. 1 of the Supplemental Materials*, which contains a detailed discussion and uncertainty calculations based on standard error propagation techniques (including values for all individual sources of uncertainty). The measurement uncertainties are reported as 95% confidence intervals. Some sources contributing to the measurement uncertainties are likely of very similar magnitudes for all sites (based on identical instrumentation, methods, and similar values of scattering Angstrom exponent used for scattering truncation correction, etc) so the reported uncertainties are likely over-estimated when evaluating differences between sites and between months at a given site. We follow an approach similar to Anderson and Ogren (1998) and Anderson et al. (1999) and calculate ‘measurement precision uncertainties’. The measurement precision uncertainties only include the contributions to the total measurement uncertainty which are not expected to be the same for all sites/seasons (Anderson et al., 1999). Detailed calculations and justifications are provided in Sect. 1 of Supplemental Materials and briefly outlined in Sect. 2.4. When considering differences in aerosol optical properties (AOPs) from different sites and times, one must also account for uncertainty due to atmospheric variability. To quantify atmospheric variability, we now report mean AOPs (in place of medians) and plot 95% confidence intervals (Cis) of the mean AOP values as error bars. Scattering and absorption coefficients are better-approximated by log-normal distribution so we report these properties as geometric means and 95% Cis of the geometric mean values. We report differences in the mean AOPs as **significant** if the differences are larger than both (a) twice the measurement precision uncertainties (Table 3); and (b) two times the 95% CI of the mean AOP values.

- 4. Proxies: Several proxies (pollution sources, wind sector, agriculture, PBL,...) can explain the various cycles at each station. These proxies have however to be taken into account in a coherent way though the paper. For example, the PBL height is presented as a main parameter to explain the diurnal cycles, but is not at all taken into account concerning the annual cycles. It is however well-known that PBL height have annual cycle with usually minima in winter and maxima in summer. Seidel et al. (2012) published a PBL height climatology for the whole USA, including seasonal and diurnal variations.**

Authors’ response:

This is a great suggestion and served as the primary motivation for many of the manuscript changes! We now include proxies for each site: (1) pollution rose diagrams showing the dependence of AOPs on wind-sector; (2) known regional pollution sources; (3) published aerosol chemistry at each site; and (4) reported PBL heights for the sites and/or regions. We do not believe that the results from the Seidel paper can be applied at the four sites but we have located results from other studies located at the sites or in the region.

Changes in manuscript

We now include pollution-rose diagrams for each season at each site. The pollution-rose diagrams show the dependence of scattering and absorption coefficient on wind direction for each season and site. (Fig(s).5-8). 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 also include pollution-rose diagrams for the following aerosol properties for each site/season in Fig(s) S16-S22 of Supplemental Materials: (1) PM1 scattering fraction; (2) single-scattering albedo; (3) hemispheric backscatter fraction. We discuss temporal and regional variability in the context of pollution-rose diagrams, published aerosol chemistry at the sites, reported PBL heights for the regions, and known regional pollution sources.

Tabulated PBL heights from studies based on measurements near the four sites are given in Sect(s) 3.1-3.4. The PBL heights from APP are unpublished monthly median morning and afternoon profiles for each month of the year, based on 18 months of PBL heights derived by a co-located micro-pulsed lidar. We report median morning/afternoon PBL heights for each season at APP (Sect. 3.1). Published values of PBL heights based on studies near the other sites are reported in Sect(s) 3.2-3.4. We cite reported median morning and afternoon mixing layer heights for each season at SGP (Delle Monache, et al., 2004); mean mixing layer heights for each season at Joliet, IL, located ~130km NNW of BND (Holzworth, 1964); and seasonal dependence of of mixing layer heights > 1km reported for each season near Buffalo, located ~170km SE of EGB (Holzworth,1964). The reported results near BND and EGB are not segregated by time of day so we can use PBL heights as a proxy for seasonal variability at these sites but not as a proxy for diurnal variability. However, the seasonality of the reported boundary layer heights clearly indicates the large seasonality of convection at BND and EGB.

5. **Relevance and length of the paper:** The climatology of the SGP and BND for most of the parameters were already published by Delene and Ogren (2002) and Andrew et al. (2011) also presented systematic relationship among aerosol optical parameters for a lot of stations. This paper presents a climatology of 2 other stations (APP and EGB) with a new parameter (Absorption Ångström exponent) are extent the systematic relationships published by Delene and Ogren (2002) to more optical properties. Considering that a lot of information are described several times in the paper (for example the long-term trends are presented in relation with the annual, weekly and diurnal cycles with not much new information in some cases), the paper could be centered on the new results and be shortened.

Authors' response:

We thank the reviewer for these helpful suggestions and have completely re-structured the paper to better organize the material. The changes emphasize new results and eliminate much redundancy.

Changes in manuscript:

See the detailed outline of our revised manuscript in response to Reviewer #1 comment 1. The structure of the revised manuscript directly addresses all of the major suggestions made by both reviewers regarding paper structure

6. **Some figures are given as supplement material. They are however largely discussed in the paper. For example, only the weekly cycles of the absorption exponent are presented in the paper (Fig. 6), but the 2 first § of 3.2.1 (corresponding to about one page) describe the weekly cycles of all other parameters from which only one (scattering coefficient) is given as supplement.**

Authors' response:

We now include a figure (Fig. 3) showing the weekly and diurnal σ_{sp} cycles. The weekly and diurnal weekly σ_{sp} cycles are shown in Fig. 4. Most intensive AOPs exhibit minimal variability on weekly and diurnal time scales, except for modest variability in ω_0 and DRFE for some seasons. The other intensive AOPs demonstrate negligible and/or statistically-insignificant variability on weekly and diurnal timescales.

Changes in manuscript:

We show the weekly and diurnal σ_{sp} and σ_{ap} cycles for all sites/seasons in Figs. 3-4 and discuss these cycles in Sect. 4.1. We report features of the σ_{ap} cycles common to all or most sites in Sect. 4.1.1. We report features of the annual, weekly and daily σ_{ap} cycles that are unique to individual sites in Sects. 4.1.2-4.1.5, in the context of regional aerosol sources, σ_{ap} pollution-rose diagrams, and reported PBL heights at/near the site. We also include the weekly and diurnal cycles of the other AOPs in the Supplemental Materials (Fig(s). S9-S15). We briefly discuss any significant variability in these AOPs.

- 7. There is a lot of information on each figure, the figures are quite small and the axis and legends are really difficult to read (I have to enhance the figure by 300%). For example, PM10 and PM1 results are systematically plotted, even if results are similar. One could be used throughout the paper and a short section could discuss the difference between PM10 and PM1.**

Authors' response:

We agree with the reviewer's suggestions and have implemented the below-mentioned changes.

Changes in manuscript:

We increased the font size in the figures and re-scaled several of the plots to make the variability much easier to see. We now report PM1 AOPs throughout the paper (unless otherwise stated) and have relegated results for all APP, BND, and SGP PM10 aerosol optical properties (AOPs) except PM10 scattering Angstrom exponent (which is more relevant than PM1 scattering Angstrom exponent) 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). Removing the PM10 traces from the figures reduces the information in each figure, suggested above by reviewer #1. The plots are now readable when viewing at 100%.

- 8. For the annual cycle, the results for the whole year should not be linked (with line) to the monthly results to avoid confusion. Similarly the results for the whole week should be separated from the daily results.**

Authors' response:

Good catch! We fixed the problem.

Changes in manuscript:

We separated the line connecting results for entire year, month, and day from the corresponding 'ALL' data points for all traces in the plots.

- 9. P. 26980: please indicate the percentage of hours with RH>40% at EGB, perhaps also if an annual variability if measured for RH>40%.**

Authors' response:

Unlike APP, BND, and SGP, the sampled air RH is not actively-controlled at EGB. Naturally this will lead to questions as to whether the scattering and back-scattering measured at EGB is also representative of 'dried aerosols'. The temperature inside the nephelometer is several degrees higher than the ambient air temperature for all seasons so the instrument RH is much less than the sampled RH. However, small enhancements in scattering coefficient above 'dried aerosol levels' are still likely during summer at EGB. We make the argument that the lack of RH control at EGB results in negligible effect during non-summer

months and a small effect on geometric mean scattering coefficients during summer, but not near enough to explain the seasonal scattering variability demonstrated at EGB.

To estimate the magnitude of the scattering enhancement, we applied scattering hygroscopic growth gamma fit parameters (Quinn et al., 2005) based on humidified light scattering and hemispheric backscattering measurements at APP and SGP (not included here) to the hourly-averaged light scattering and hemispheric backscattering values for hours when the nephelometer internal RH exceeded 40%. For gamma values encompassing the 5th through 95th percentiles (i.e., basically the entire range of possible growth factors), the correction of light scattering and hemispheric backscattering from modestly-elevated RH values (RH~45-50%) to values at RH=40% was ~3-4% and the uncertainty in applying static correction factors for sites with no humidified scattering measurements (BND, EGB) was similarly small. Based on these relatively small adjustments, hours with elevated nephelometer RH were retained and no RH corrections were applied to the scattering measurements for these hours. Application of typical summer hygroscopic growth factors measured at APP ($f(\text{RH}) \sim 1.5-1.6$) can be applied to show that scattering enhancements of ~50-60% are possible for RH=85% and that the enhancements are ~20% or less for RH=70%. Even during July and August, the internal nephelometer RH at EGB only exceeds RH>50% for 60% of the hours and RH>70% for 8-18% of the hours. Scattering enhancements of 20-50% during 8-18% of the hours will produce a small enhancement in geometric mean scattering coefficient during these months but this enhancement is nowhere close to the seasonal differences in scattering observed at EGB (Fig.2) and reported in this paper.

Changes in manuscript:

We have added a 1-page section to the Supplemental Materials (Section S3). Table S7 shows the % of hours that the EGB nephelometer internal RH exceeds 40%,50%,60%, and 70% for each month of the year. Arguments along the lines of that provided above are used to reason that the results presented in this study are likely unaltered by moderately-elevated nephelometer RH during summer at EGB.

10. P. 26980: if possible give a reference for the hygroscopic dependence of light scattering

Author's response:

Done

Changes in manuscript:

We now include the following reference for the hygroscopic dependence of light scattering:

Sheridan, P. J., Delene, D. J., and Ogren, J. A.: Four years of continuous surface aerosol measurements from the Department of Energy's Atmospheric Radiation Measurement Program Southern Great Plains Cloud and Radiation Testbed site, *J. Geophys. Res.*, 106, D18, 20735-20747, doi:10.1029/2001JD000785, 2001.

11. P. 26982: If I understand it well, the Nephelometer were heated to ensure RH<40%, but not the PSAP? This means that different inlets were used for both instruments?

Authors' response:

The inlet immediately before the switched impactor box is actively heated to an $\text{RH} \leq 40\%$. The flow splits off to the nephelometer and PSAP just after the impactor box. The sample lines leading from impactor box to the nephelometer and PSAP are short (less than 1 meter) and the lines are insulated. The nephelometer internal temperature is higher than the temperature of the impactor box so $\text{RH} \leq 40\%$ is satisfied for the

nephelometer. Relative humidity is measured at the nephelometer inlet to verify this and is calculated internal to the nephelometer (based on internal temperature measurement and assumption of constant dewpoint). A small heater block is placed inside the PSAP (see discussion in Sect. 2.1) so the $RH \leq 40\%$ condition is likely satisfied in the PSAP as well but no RH measurements are made there. The flow schematic is shown in Sheridan, et al., 2001 and other similar papers.

Changes in manuscript:

We now include a reference to the Sheridan, et al., 2001 paper at the end of the following sentence in Sect. 2.1

“To reduce the confounding effects of relative humidity (RH) on the aerosol measurements, the sample air is gently heated at all sites except EGB to achieve sample line and instrument $RH \leq 40\%$.

(Sheridan, et al., 2001).”

12. P. 26983: were negative scattering Ångström exponents never measured?

Authors' response:

Scattering Ångström exponent (SAE) can in principle be negative for the most dust-influenced sites but none of the four sites in this paper fits that description. Negative SAE are rarely measured at the four sites. This is seen from the plot of PM1 scattering fraction versus SAE (Fig. 9g in revised manuscript). Data points are only plotted for a given SAE bin if the number of data points in that bin are at least 0.1% of all data points. There are no data points for $SAE < 0.6$ at APP and BND. There are not data negative SAE data points for SGP but one slightly positive data points near zero. The number of negative SAE occurrences at SGP is thus less than 0.1%.

Changes in manuscript:

None.

13. P. 26984: To my knowledge, the uncertainties on b and β are probably quite high.

Authors' responses:

The uncertainties in b are actually not large for the sites and time period of this study, when co-variances between aerosol optical properties are considered (See detailed analysis in Section S1 of the Supplementary Materials) and the uncertainties in β are nearly the same as those of b . The total measurement uncertainty $\Delta b = 0.0036$ (0.0032) for PM10 (PM1) size cut when near-annually-averaged aerosol optical property values at the four sites are used to calculate Δb . The uncertainty Δb is reduced due to the high correlation between scattering and hemispheric backscatter coefficients ($r \sim 0.99$ at APP), which necessitates inclusion of the co-variance term when calculating Δb (See Sect. 1 of Supplemental Materials). Some of the sources of nephelometer uncertainty do not need to be considered when making comparisons made at different sites/times using identical instruments and protocols (see Anderson and Ogren, 1998 and Anderson, et al. 1999). The total uncertainty is reduced further to 0.0015 (0.0016) when comparing differences made by instruments using the same instruments and protocols

Changes in document:

We added a detailed uncertainty analysis (Sect. S1 of Supplemental Materials) and added a short section to the paper (Sect. 2.4) summarizing the results relevant to comparisons of AOPs measured at different sites/times

14. P. 26984: if hours with scattering coefficient lower than 1 Mm⁻¹ are discarded, the “cleanest” atmospheres are not taken into account. Would it change the results presented in this study? Is there an annual cycle of the percent of discarded hours?

Authors’ response:

We use the scattering and absorption coefficients from all hours in calculating the statistics for these variables, so as not to bias the results toward ‘less clean air’ conditions. If anything, the mean intensive properties would be made more unclear by inclusion of such hours, as most of the intensive properties would be comprised of ratios of two small numbers (Table 2). Other studies such as Delene and Ogren (2002) and Andrews (2011) have also neglected to consider aerosol intensive properties for such hours, for identical reasons.

Changes in document:

We have added a table in the Supplemental Materials (Table S6) which shows the annual cycle of hours with PM1 scattering coefficient less than 1 Mm⁻¹ at each site. We also include the following sentences in Section 2.5 –Data analysis methods:

“We follow a similar approach to that taken by D&O2002 and Andrews et al. (2011) and only use hours for which σ_{sp} at 550 nm is at least 1.0 Mm⁻¹ for the PM1 size cut to calculate the intensive AOP statistics, so as to reduce noise resulting from taking ratios of two small quantities (Table 2). Filtering the intensive AOPs for low- σ_{sp} hours discards 1.4% of the hours at APP, 0.1% at BND, 1.8% at EGB, and 0.5% at SGP. These percentages are uniform across seasons, except for slightly higher percentages during fall at EGB and SGP (Table S6). We use all hours in calculating σ_{ap} , σ_{sp} and σ_{bsp} statistics, to avoid a bias toward ‘less clean’ conditions. Lack of PM10 measurements and use of a single-wavelength PSAP preclude calculation of R_{sp} , R_{ap} , and α_{ap} at EGB.”

15. § 3: the end of the first § and the second one contains information that should be given in the experimental section.

Authors’ response:

Good suggestion! We assume that the reviewer is referring to the material on lines 2-20 of p. 26986.

Changes in manuscript:

We created a new section (Section 2.5 Data analysis methods) to consolidate all of the data analysis material, such as that suggested by the reviewer.

16. P. 26986: the use of medians instead of means is appreciated because most of the used parameter are not normally distributed.

Authors’ response:

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. 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.

Changes in manuscript:

We copied part of our response to reviewer #1 comment 3 here. *“When considering differences in aerosol optical properties (AOPs) from different sites and times, one must also account for uncertainty due to atmospheric variability. To quantify atmospheric variability, we now report mean aerosol optical properties (in place of medians) and plot 95% confidence intervals (Cis) of the mean aerosol optical property values as error bars. Scattering and absorption coefficients are better-approximated by log-normal distribution so we report these properties as geometric means and 95% Cis of the geometric mean values. Differences in the mean AOPs are significant at the 95% confidence interval if the uncertainty ranges do not overlap. The uncertainties in each case are the larger of (a) measurement precision uncertainties (Table 3); and (b) two times the 95% CI of the mean AOP values”*

17. P. 26988 Several stations/parameters present a decrease not only in fall (that is discussed), but also in spring (not discussed).

Authors’ response:

Our intention was to stress the steeper changes in several AOPs (scattering and absorption coefficients and hemispheric backscatter fraction) in going from summer to fall, relative to those from summer-to-spring. We agree that we did not make this point effectively in places.

Changes in manuscript:

We now discuss both the significant summer-fall and summer-spring differences common to all sites in the first paragraph of *Sect 4.1.1. Temporal variability common to all sites.*

18. P. 26990: line 16-18: Is there other kind of large aerosol than dust in winter? Is there more dust in winter or the ratio between dust and other aerosol is greater in winter?

Authors’ response:

The secondary winter σ_{sp} peaks at BND, SGP, and possibly EGB result from the addition of large, highly-reflecting PM1 particles during these months, as seen from the pollution-rose plots, the winter increase in ω_0 , decrease in b , and relatively small change in R_{sp} . We located published aerosol chemistry for all sites that indicate large and highly-variable ammonium nitrate concentrations during winter months at BND (Buscu-Guven et al., 2007), SGP (Parworth et al., 2015), and EGB (Rupakheti, et al., 2005). The temperature-dependence of σ_{sp} during cold-season months (Fig. S5 in Supplemental Materials) is consistent with the temperatures favorable for gas-to-particle partitioning reported for SGP by Parworth et al. (2015) and for EGB by Rupakheti, et al., 2005. The upper Midwestern U.S (where BND is situated) is also home to very high winter ammonium nitrate concentrations (Hand et al., 2015). We then examined the dependence of winter σ_{sp} and (for BND and SGP). Significant winter increases in σ_{sp} and ω_0 are observed for northerly wind sectors (Fig. 6a and Fig. 8a), corresponding to colder air masses passing over regions with high levels of ammonium nitrate precursors. We hypothesize that the secondary winter σ_{sp} peaks at BND and SGP are influenced by temperature-dependent gas-to-particle partitioning of ammonia and nitric acid to form ammonium nitrate. This hypothesis is consistent with reported winter aerosol chemistry at BND and SGP and with pollution-rose plots and temperature-dependence of σ_{sp} . The influence of ammonium nitrate on the small winter σ_{sp} is inconclusive. Wind sector analysis and published chemistry support the hypothesis but the dependence of σ_{sp} on temperature during winter at EGB does not support this.

Changes in manuscript:

We include the discussion outlined above in our discussion of annual σ_{sp} cycles at the sites (Sect. 4.1).

19. P.26990 Line 23: The cycle in single scattering albedo reflects the difference in scattering and absorption cycle.

Authors' response:

We agree that this point was poorly-phrased and have attempted to improve its clarity, as shown below.

Changes in manuscript:

We replaced the passage on P.26990 of the original manuscript with the following passage, now located in *Sect. 4.1.1 Temporal variability common to all sites*:

“July-August σ_{sp} maxima are observed at all sites (Fig. 2a), with steeper σ_{sp} decreases from summer to fall than from summer to spring. Summer/spring and summer/fall σ_{sp} differences at EGB and APP are approximately twice the magnitude of those observed at BND and SGP. Scattering coefficient reaches a minima during October at all sites except APP, where it is lowest in December. Absorption coefficient is highest in summer and lowest in winter at all sites (Fig. 2b), although the differences between the summer maxima and the surrounding months are only significant at BND and EGB. Summer-to-autumn σ_{sp} decreases are larger than those of σ_{ap} , leading to minimum ω_0 in October at all sites (Fig. 2e). Hemispheric backscatter fraction is highest in October at all sites (Fig. 2d). The confluence of early-autumn decreases in σ_{sp} and ω_0 and increases in b is indicative of less production and/or more efficient removal of large, highly-scattering particles during early autumn, relative to summer. This effect is most noticeable at EGB and APP (Fig. 2) and also is seen in the summer-spring differences at APP. October ω_0 minima contribute to DRFE maxima (least negative) at EGB and BND but no significant autumn DRFE changes are seen at APP and SGP (Fig. 2f). Photochemistry likely influences the summer σ_{sp} maxima and σ_{sp} that are larger in spring than in fall at all sites. The temperature-dependence of σ_{sp} (Fig. S5) and differences in monthly-median temperatures (Fig. S23) combine to yield differences in σ_{sp} that are of similar magnitude to the large observed summer-spring (July-April) σ_{sp} differences at EGB and APP and to the observed summer-autumn (July-October) σ_{sp} differences at EGB, BND, and SGP (Fig. 2a). The summer-autumn σ_{sp} difference based on temperature considerations is less at APP than the observed σ_{sp} difference, leading us to hypothesize an additional contributor to the autumn σ_{sp} decrease. Cloud and fog scavenging of large, highly-reflecting particles would be consistent with cooler September temperatures, higher RH (Fig. S23), and cloud cover in September at APP. The inverse relationship between σ_{sp} and b seen in the annual cycles at all sites (Figs. 2a and 2d) is indicative of the influence of particle growth (and possibly cloud or fog scavenging) on σ_{sp} . Wet deposition likely impacts σ_{sp} most in summer and least in spring and fall, given the seasonality of precipitation at the sites. Secondary σ_{sp} maxima are observed during winter at all sites except APP (Fig. 2a). When combined with winter σ_{ap} minima, the result is a winter ω_0 maxima at these sites (Fig. 2e).

”

20. P.26990 Line 29: The single scattering albedo being an intensive properties and should therefore not depends on the amount of aerosol.

Authors' response:

The reviewer is correct and the wording used was poor. We were referring to the results of the systematic relationship between ω_0 and σ_{sp} (Fig.10 of original manuscript), which reveals that aerosols at the four sites reported in this study are more absorbing (lower ω_0) under low loading conditions such as fall at EGB.

Changes in manuscript:

We removed this sentence.

21. P. 26994 lines 5-8: Due to the inter-annual variability and to the fact that the authors do not explicitly use the 2000-2010 measurement for the trend analysis, it is not possible to conclude that the reduction (...) may have occurred during the current period (even if “may” is used).

Authors’ response:

We agree with the reviewer that the information presented in the original manuscript does not allow for us to rule out the possibility that the reduction may have occurred during the current period.

Changes in manuscript:

We have implemented the suggestion made in reviewer #1 comment 2. We replaced all of the inter-period comparison of the 2010-2013 period vs Delene and Ogren, 2002 with a more appropriate trend analysis. The trend analysis uses the entire 1996-2013 period at BND and 1997-2013 period at SGP. Results of the trend analysis are reported in Table 4 and discussed in *Sect 4.3 Long-term aerosol optical property trends at BND and SGP*.

22. P. 26994: The SGP absorption trend were not analyzed in Collaud Coen et al. (2013) because “Unfortunately the 14 yr absorption record at SGP was influenced by high frequency humidity changes due to air conditioning cycling and those data are therefore not included in this study.” Were the SGP absorption now corrected to be used for the trend analysis?

Authors’ response:

Most occurrences of high noise in the PSAP were edited from the data at SGP. Upon further analysis there was a particular time period during the 2010-2012 summers when the dew points were extremely high and the noise was particularly troublesome. On average about 15% of the summertime data was removed from the PSAP data stream during this time. Considerable effort was made to reduce the PSAP RH since this time. Comparison of the PSAP 2010-2012 summer data with 2013-2014 shows no noticeable difference. For this reason we decided to include the PSAP data in the discussion and plots of the short term seasonal, weekly and diurnal trends, but remove it from the 1997-2013 long term trend analysis as even small changes could impact the long-term trend.

Changes in manuscript:

For reasons discussed above, we still report absorption data from SGP for the 2010-2013 period (Sect(s) 4.1, 4.2, and 4.4). We do not include SGP absorption as part of the trend analysis (Sect. 4.3), as a large fraction of the 1997-2010 period was before we were able to mitigate the problem.

We also add the following paragraph to *Sect. 2.3-Data processing, quality assurance, and calculated AOPs*:

“Unlike the other sites, RH can have a pronounced effect on the absorption measurements at SGP, particularly for the summer months of 2010-2012. Excessively high temperatures during the summer months of June-August and the early part of September of these years resulted in high daytime dew points that often were as high as 20 to 22 °C during the late afternoon. The high sample humidity coincided with unusually high noise in the PSAP. The hours with noisy σ_{ap} data were removed. On average this resulted in a 15% loss or 3.6 hours per day in the data. Since this time, effort was made to lower the sample RH through insulation of the optics block, use of a Nafion drier on the instrument inlet and rerouting the trailer ventilation. In a previous paper on long-term aerosol trends by Collaud Coen et al. (2013) the absorption data at SGP were excluded for this reason. After further analysis we decided to include the SGP absorption data in this paper for the short term seasonal, weekly and diurnal trend analysis but remove them for the long term trend analysis. Despite the data loss, the SGP summertime absorption coefficients

don't show a remarkable difference compared to the other sites nor does the 2010-2012 time period vary significantly from 2013, when the noise was not as apparent."

23. Often features are described but not correlated with a phenomenon or tentatively explained. For example : p. 26998 line 6. Why is the week minima on Sunday and Monday not seen at BND in winter and fall and at EGB in spring? Line 16: why the peak day varies with season? line 18: why the absorption peaks on Tuesday in autumn at BND ?

Author responses:

The use of proxies to help explain AOP variability has formed the basis for many of the major changes to this manuscript, as discussed in our response to reviewer #1 comment 4

Changes in manuscript:

We have completely revised the manuscript and modified one of the paper objectives. See response to reviewer #1 comment 4. One of the paper objectives (Objective 1, stated in the Introduction) is to " provide an explanation of temporal and regional AOP variability that is consistent with regional aerosol sources and transport and with reported aerosol chemistry at the sites"

24. P. 27001: is it possible to show the influence of Barrie on APP measurement in a Figure or table? This is not major point, but I take this occasion to say that some dependences could be directly presented in figure to help the reader to understand the influence of the proxis, and some of the figures presenting cycles could be omitted.

Authors' responses:

We assume that the reviewer was referring to the influence of Barrie on measurements made at EGB, since Barrie is located near EGB. We have added several proxies to help interpret the reported variability in AOPs at the sites. The pollution-rose diagrams for σ_{ap} and σ_{sp} measured at EGB (Fig. 7) show that wind sectors arriving at the EGB station from Barrie (NE of EGB) are generally clean and infrequent during all seasons.

Changes in manuscript:

We have added pollution-rose plots for each site and season. The plots show the dependence of σ_{ap} , σ_{sp} , and some intensive AOPs (ω_0 , b , and R_{sp}) on wind direction. We include the σ_{sp} and σ_{ap} pollution-rose plots for each site and season in Fig(s). 5-8. The other pollution-rose plots are included in the Supplemental Materials (Fig(s). S16-S22).

25. P. 27002 lines 19-23: would it be possible to show the dependence between the absorption and PBL height by plotting the diurnal Max/min (or max-min) as a function of a parameter describing the convection (irradiance or T) ?

Authors' response:

We thank the reviewer for her/his suggestion. As a proxy for the effect of convection on the diurnal cycles of measured near-surface AOPs, we calculated the difference between daily maximum and daily minimum σ_{ap} for each day of the study period. We performed similar calculations for the difference between daily maximum and minimum σ_{sp} . We then plotted each of these differences versus daily maximum surface temperature for each season at each site. We used the correlation coefficients between daily max minus daily min absorption versus temperature to estimate the effect of convection on the diurnal σ_{ap} and σ_{sp} cycles. Unfortunately, the correlations for individual seasons at the sites did not demonstrate any noticeable relationship with the observed diurnal variability of σ_{ap} and σ_{sp} . We do not feel comfortable

including the correlations in the manuscript without a better understanding of other factors which could be affecting the relationships.

Changes in manuscript:

We use PBL heights reported from studies based at or near the four sites and their seasonal dependence to help explain the AOP seasonal and diurnal cycles. We realize that the reported values are seasonal means (or medians) and are in most cases from time periods different than the study. However, we believe that the reported values provide a suitable proxy for interpreting the seasonal and diurnal σ_{ap} and σ_{sp} cycles.

26. P. 27003 line 4: do you think that the ground use (cropland or forest) could modify the morning PBL height by a factor of 2h?

Authors' response:

Our statement in the original manuscript was "One other interesting feature in the σ_{ap} cycles is that the time of morning peak at APP and EGB lags the time of the peak at BND and SGP by roughly two hours for all seasons. Possible explanations include (1) differences in morning boundary layer heights over cropland and forested areas; and (2) differences in traffic sources (as discussed in the previous paragraphs)." This is another case where the available information does not help us to draw conclusions, even when we include proxies such as pollution-rose diagrams and reported PBL heights for the sites. We can only hypothesize (or more like speculate in this case) as to the possible source for this phenomena

Changes in manuscript:

We remove mention of the differences in morning boundary layer heights over cropland and forested areas as a possible explanation. We cannot test this hypothesis.

27. P. 27004 lines5-8: please give at least a tentative explanation to explain the absorption Ångström exponent cycles. Could the observed cycles be in the uncertainty of the absorption Ångström exponent?

Authors' response:

The precision measurement uncertainty for α_{ap} is close to 0.1, at 95% confidence (Table 3 of revised manuscript). This means that differences $\Delta \alpha_{ap} \geq 0.2$ among sites or for different months at a given site are significant. We report temporal and regional differences α_{ap} as significant if they are larger than ~ 0.2 and insignificant if they are not. As an example, α_{ap} is not statistically-different from 1 (theoretical value for black carbon) unless α_{ap} is greater than ~ 1.2 or less than ~ 0.8 . The diurnal and weekly variability in AAE cycles is not statistically-significant at the 95% confidence interval in most cases. The winter weekly variability in AAE at APP and summer diurnal variability in AAE are both marginally-significant at this confidence level. (Fig. S15). See our response to Reviewer #1 comment 3 for how we determine the 95% CI of mean aerosol optical properties. The seasonal AAE cycles are significant for APP, BND, and SGP (Fig.2), especially at APP.

Changes in manuscript:

We discuss temporal variability (Sect. 4.1) in α_{ap} and systematic relationships involving α_{ap} (Sect. 4.4.2) in the context of the uncertainty of α_{ap} . We also use this uncertainty to determine under what conditions α_{ap} is statistically-different than 1.

28. P. 27004 lines23-25: Is this really statistically significant regarding the inter-annual variability and the uncertainties?

Authors' response:

We now include a trend analysis for SGP (1997-2013) and BND (1996-2013), in place of the inter-period comparisons of seasonal, weekly, and diurnal variability at SGP. The trend analysis is suggested by Reviewer 1 (comment 2). For reasons discussed in our response to Reviewer #1 comment 22, we do not include SGP absorption measurements in the trend analysis.

Changes in manuscript:

See our response to reviewer #1 comment 2 for details of the trend analysis that we now include in place of the inter-period comparisons of AOP seasonal, weekly, and diurnal variability at BND and SGP

29. - P. 27005 lines18-20: where the medians done before to calculate the intensive properties or after?

Authors' response:

The σ_{sp} values for each hour were placed in the appropriate scattering bin of size 10Mm^{-1} and hourly-averaged intensive properties were also placed in the appropriate σ_{sp} bins. The mean intensive properties were then calculated for each scattering bin.

30. § 3.4: do you see some systematic difference between your analysis on continental sites and the results of Andrew et al. (2011) on FT sites?

Authors' response:

See the changes to the manuscript described below.

Changes in manuscript:

We now include the following paragraph at the end of Sect. 4.4.1-Annual systematic relationships among AOPs:

“AOPs at the rural continental sites reported here have similar covariances (Fig. 9) as those at a majority of mountain sites reported on by Andrews et al. (2011). Andrews et al. (2011) also reported relationships amongst AOPs based on long-term aircraft measurements made over BND and SGP, although their free tropospheric AOP relationships for BND and SGP only extended up to $\sigma_{sp} \sim 25 \text{Mm}^{-1}$. Most of the free troposphere AOP relationships reported for SGP (Andrews et al., 2011) are similar to the corresponding near-surface AOP relationships (Fig. 9) but there are some noticeable differences for BND. Andrews et al. (2011) reported the following AOP relationships as σ_{sp} increased from 0-25 Mm^{-1} at BND: (1) b increased slightly (0.12 to 0.13); (2) ω_0 remained nearly constant (less than 0.01 increase); and (3) α_{sp} increased by a larger amount (~ 0.12 to 0.17) than in our study (Fig. 9e). The differences between these relationships and those in Figs. 9a, 9b, and 9e could be due to smaller particles that undergo less atmospheric processing (particle growth, cloud scavenging, and deposition) in the free troposphere above BND, relative to particles near the surface.”

31. - P. 27006 and figure 9: for most of the station and season, the single scattering albedo versus scattering coefficient slope is larger for low aerosol concentrations (low scattering coefficient) and smaller for high concentrations. Do you have an explanation for this feature?

Authors' response:

We think that the answer is simply that the ω_0 values are bounded by physical constraints to an upper limit of unity and values are bounded to a lower limit of zero.

Changes in manuscript:

We do not speculate as to the change in the slope, for the reason outlined above.

32. p. 27007 lines 20-25: Is it possible that these variations are just in the uncertainties?

Authors' response:

Results of the uncertainty analysis that we have added to the paper (Sect. 2.4-Measurement uncertainties) and described in detail (Sect. S1 of Supplemental Materials) confirm that these variations in DRFE are not statistically-significant at the 95% confidence interval.

Changes in manuscript:

The beginning of first paragraph in *Section 4.4.1-Annual systematic relationships among AOPs* now reads as follows, where we underline the relevant statement to this response:

“Single scattering albedo increases and b decreases with increasing σ_{sp} at all sites (Figs. 9a and 9b). Hemispheric backscatter fraction demonstrates an inverse relationship with ω_o over the entire ω_o -range at EGB and for $\omega_o > 0.85$ at the other sites (Fig.9c), a condition representative of all months (Fig(s). 2d and 2e). The co-variability of ω_o and b leads to a DRFE dependence on σ_{sp} that is statistically-insignificant for all sites, with the exception of the lowest σ_{sp} conditions at APP (Fig.9d).”

33. P. 27007 lines 27-28: What do you mean by “ the b vs scattering coefficient relationship was slightly more important than the single scattering albedo vs. scattering coefficient relationship” ?

Authors' response:

We meant that the influence of b on DRFE for APP for increasing σ_{sp} appears to be slightly larger than the influence of ω_o on DRFE. DRFE becomes slightly less negative with increasing σ_{sp} and is influenced by b and ω_o (Table 2). Single-scattering albedo increases with increasing σ_{sp} . which tends to drive DRFE toward more negative values. Hemispheric backscatter fraction increases with increasing σ_{sp} . which tends to drive DRFE toward less negative values. A shift in DRFE toward slightly less negative values with increasing σ_{sp} would then imply that the decrease in b exerts a slightly larger influence on DRFE than does the increase in ω_o . We agree that the wording was poor.

Changes in manuscript:

The systematic relationship between DRFE and σ_{sp} is not discussed in the revised manuscript, since the observed differences in DRFE are not significant at 95% confidence (see response to Reviewer 1 comment 32).

34. - § 3.4.4 The order of the explanation and of the item on the figure are opposite

Authors' response:

We agree with the reviewer

Changes in manuscript:

We switched the ordering of the individual plots in the systematic relationship figures so that the plots are now in the order discussed in the manuscript

35. Table 1: are the cloud fraction and the spectrally-averaged surface albedo the same for the 4 stations?

Authors' response:

The cloud fraction and the spectrally-averaged surface albedo not the same for the 4 stations. By assuming no geographical variation of non-aerosol variables such as cloud fraction and spectrally-averaged surface albedo, the intrinsic radiative forcing efficiency of the aerosols at the 4 stations can be compared. Similar comparisons have been carried out by Delene and Ogren (2002) and Andrews (2011). The neglect of geographic variation in non-aerosol properties in the equation for DRFE results in only an estimated value and we have added clarification to this extent in Sect. 2.3. We also note that we interchange the order of the first two tables in the new manuscript, based on earlier mention of the instruments (Table 2 of original manuscript) in the revised manuscript.

Changes in manuscript:

We clarify the description of DRFE in Section 2.3-Data processing and quality assurance so that the final paragraph in Section 2.3 reads as follows:

“Haywood and Shine (1995) present simple equations for calculating top-of-atmosphere (TOA) aerosol direct radiative forcing (DRF) and direct radiative forcing efficiency (DRFE, Table 2) for an optically-thin, partially-absorbing atmosphere. DRFE represents the DRF per unit aerosol optical depth (τ) and is to first-order independent of τ . If globally-averaged values for all non-aerosol parameters are used (Table 2), the simple equation for DRFE provides a means of comparing the intrinsic forcing efficiency of the aerosols measured at different sites and times, through DRFE dependence on ω_0 and on up-scatter fraction β . The DRFE values themselves are only approximations when globally-averaged values are used. The up-scatter fraction represents the fraction of incoming solar radiation that is scattered by atmospheric aerosols back to space. Up-scatter fraction been related to b by the approximation of Wiscombe and Grams (1976). A second-order curve fit of the points in their Fig. 3 as reported in Sheridan and Ogren (1999) provides the parameterization shown in Table 2.”

36. A map with the stations would help the reader to understand the spatial changes of aerosol parameters

Authors' response:

We agree with the reviewer's suggestion.

Changes in manuscript:

We have added a map with the stations (Fig.1)

37. Fig. 1 and 2: is there a reason to separate the annual cycles into 2 figures?

Authors' response:

The figures were separated solely for readability, as the individual plots were too difficult to read plots of all 9 AOPs are on same figure (Each plot is too small).

Changes in manuscript:

We increased the font size in the figures and re-scaled several of the plots to make the variability much easier to see. We also removed the PM10 traces from most plots and only display PM1 (in response to Reviewer #1 comment 7). We are now able to make the plots smaller (so more plots per figure). Even now we are only able to include 8 plots on a figure while still making the plots readable. We report 9 AOPs (scattering and absorption coefficients and 7 intensive parameters). As a result, we display the seasonal variability of PM1 absorption fraction in Fig. S8 of Supplemental Materials and we display seasonal variability for the other 8 AOPs in Fig.2 of the paper. The PM1 scattering and absorption fractions display similar behavior so we only include PM1 scattering fraction in Fig. 1. One of our goals is to keep the size of the paper manageable, which is also implied by Reviewer #1, comment 5.

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.)

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 key 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.

Authors' response:

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 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 key 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 section titled *Section 2.3-Data processing, quality assurance, and calculated AOPs*. See the detailed outline of revised paper, located as a response to reviewer #1 comment 1.

Changes in manuscript:

We include the following paragraphs at the beginning of *Section 2.3-Data processing, quality assurance, and calculated AOPs*:

“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 available 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 0.7 and invalidated when the filter transmission drops below 0.5 because high filter loading 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 (all day) and Monday (early morning) σ_{ap} hours over the period of this study. Quality-assured σ_{ap} data at SGP are 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. PSAP filters are changed on weekends at the other sites.

Unlike the other sites, RH can have a pronounced effect on the absorption measurements at SGP, particularly for the summer months of 2010-2012. Excessively high temperatures during the summer months of June-August and the early part of September of these years resulted in high daytime dew points that often were as high as 20 to 22 °C during the late afternoon. The high sample humidity coincided with unusually high noise in the PSAP. The hours with noisy σ_{ap} data were removed. On average this resulted in a 15% loss or 3.6 hours per day in the data. Since this time, effort was made to lower the sample RH through insulation of the optics block, use of a Nafion drier on the instrument inlet and rerouting the trailer ventilation. In a previous paper on long-term aerosol trends by Collaud Coen et al. (2013) the

absorption data at SGP were excluded for this reason. After further analysis we decided to include the SGP absorption data in this paper for the short term seasonal, weekly and diurnal trend analysis but remove them for the long term trend analysis. Despite the data loss, the SGP summertime absorption coefficients don't show a remarkable difference compared to the other sites nor does the 2010-2012 time period vary significantly from 2013, when the noise was not as apparent.

The four NOAA-ESRL network sites discussed in this paper are located such that there are no major local aerosol sources in the predominant upwind directions, although there are some aerosol sources that are typically downwind but that may occasionally be 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 persists 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. We also incorporate proxies (See our response to reviewer #1 comment 4) such as pollution-rose diagrams, known regional pollution sources, published aerosol chemistry at the sites, published PBL heights in the regions, and temperature-dependence of light-scattering. This additional information has strengthened the "science of our results" so that we are now better-able to explain variability in a manner that is consistent with the proxies. Nevertheless, we have improved the wording of our interpretations and to only make conclusion based on results.

- 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 (Figs. 2a-b), especially at APP and EGB. See the detailed outline of revised paper for information regarding changes in paper organization. The revised outline is located in our response to reviewer #1 comment 1.

Changes in manuscript:

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

“The annual AOP cycles are larger than the weekly and diurnal AOP cycles at all sites. Nearly all annual AOP cycles are significant, with cycle amplitudes larger than the 95% confidence intervals of both the monthly-mean AOPs (Fig. 2) and the measurement uncertainties (Table 3). July-August σ_{sp} maxima are observed at all sites (Fig. 2a), with steeper σ_{sp} decreases from summer to fall than from summer to spring. Summer/spring and summer/fall σ_{sp} differences at EGB and APP are approximately twice the magnitude of those observed at BND and SGP. Scattering coefficient reaches a minima during October at all sites except APP, where it is lowest in December. Absorption coefficient is highest in summer and lowest in winter at all sites (Fig. 2b), although the differences between the summer maxima and the surrounding months are only significant at BND and EGB. Summer-to-autumn σ_{sp} decreases are larger than those of σ_{ap} , leading to minimum ω_0 in October at all sites (Fig. 2e). Hemispheric backscatter fraction is highest in October at all sites (Fig. 2d). The confluence of early-autumn decreases in σ_{sp} and ω_0 and increases in b is indicative of less production and/or more efficient removal of large, highly-scattering particles during early autumn, relative to summer. This effect is most noticeable at EGB and APP (Fig. 2) and also is seen in the summer-spring differences at APP. October ω_0 minima contribute to DRFE maxima (least negative) at EGB and BND but no significant autumn DRFE changes are seen at APP and SGP (Fig. 2f). Photochemistry likely influences the summer σ_{sp} maxima and σ_{sp} that are larger in spring than in fall at all sites. The temperature-dependence of σ_{sp} (Fig. S5) and differences in monthly-median temperatures (Fig. S23) combine to yield differences in σ_{sp} that are of similar magnitude to the large observed summer-spring (July-April) σ_{sp} differences at EGB and APP and to the observed summer-autumn (July-October) σ_{sp} differences at EGB, BND, and SGP (Fig. 2a). The summer-autumn σ_{sp} difference based on temperature considerations is less at APP than the observed σ_{sp} difference, leading us to hypothesize an additional contributor to the autumn σ_{sp} decrease. Cloud and fog scavenging of large, highly-reflecting particles would be consistent with cooler September temperatures, higher RH (Fig. S23), and cloud cover in September at APP. The inverse relationship between σ_{sp} and b seen in the annual cycles at all sites (Figs. 2a and 2d) is indicative of the influence of particle growth (and possibly cloud or fog scavenging) on σ_{sp} . Wet deposition likely impacts σ_{sp} most in summer and least in spring and fall, given the seasonality of precipitation at the sites. Secondary σ_{sp} maxima are observed during winter at all sites except APP (Fig. 2a). When combined with winter σ_{ap} minima, the result is a winter ω_0 maxima at these sites (Fig. 2e).”

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 ($R_{sp} > 0.8$ 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 (Fig. S8 and Table S8).

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

Authors' response:

The relationships involving absorption Angstrom exponent exhibited seasonal changes.

Changes in manuscript:

We qualify this in the abstract of 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:

"The objectives are to:

1. *provide an explanation of temporal and regional AOP variability that is consistent with meteorology, regional aerosol sources, and reported aerosol chemistry at the sites;*
2. *identify possible AOP trends at the long-term sites (SGP and BND);*
3. *determine whether systematic relationships exist for key aerosol properties relevant to radiative forcing calculations. "*

- 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 Watauga 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, quality assurance, and calculated AOPs*).

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, quality assurance, and calculated AOPs*. 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, quality assurance, and calculated AOPs*. 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. We use geometric means and 95% confidence intervals of the geometric means to report σ_{sp} and σ_{ap} . 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 geometric means for reporting σ_{sp} and σ_{ap} and arithmetic means for reporting intensive AOPs. We also report 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 US 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 annual σ_{ap} cycle at APP (Fig. 2b) is out of phase with the annual cycle of EC concentrations reported for rural eastern US IMPROVE sites (Hand et al., 2012). Hand et al. (2012) cited sources such as residential heating for the fall-winter EC concentration maxima. Absorption coefficient at APP exhibits a summer maximum and a winter minimum, though the summer σ_{ap} maximum is not significantly different from early fall and spring σ_{ap} (to 95% confidence). Absorption Ångström exponents of ~ 1.3 -1.4 during colder months (Fig. 2h) suggest a mix of black and brown carbon. They are also consistent with a biomass-burning OA factor in the winter aerosol mass spectra measured at APP (Fig. S2 of Link et al., 2015) and may result from winter residential wood-burning (U.S. Census Bureau, 2010; Zhang et al., 2010). However, the diurnal σ_{ap} cycles (Fig. 4b) suggest an influence from local traffic during all seasons and α_{ap} values of 1 or less for non-winter months suggest that EC is the major contributor to σ_{ap} during these months.”

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:

We agree with the reviewer

Changes in manuscript:

We removed the statement.

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 R_{sp} 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. We agree that the wording could be improved upon and have modified the discussion so that it reads as outlined below.

Changes to manuscript:

The discussion of diurnal and weekly σ_{ap} cycles at BND (Sect. 4.1.3) reads as follows:

“The weekly and diurnal σ_{ap} cycles during summer and autumn (Figs. 4c and 4d) are consistent with a large influence from regional diesel emissions during these seasons and possibly during other seasons, although the weekly and diurnal σ_{ap} cycles are only significant in summer and autumn. Maximum σ_{ap} extends from sunset to sunrise for all seasons (Fig. 4d), with a broad minimum extending from just after sunrise to just before sunset. Large seasonality of PBL heights is obvious in the diurnal σ_{ap} cycles (Fig. 4d), consistent with large (factor of 3) summer-to-winter PBL height differences reported for the region by Holzworth (1964). The absence of early morning and late afternoon local commuter peaks at BND is not surprising, since emissions from interstate highway traffic and agricultural activity represent the largest local sources of absorbing aerosols. Long-distance trucking comprises a large portion of interstate highway traffic in the region and both this and farming activities typically persist throughout the day. The diurnal σ_{ap} cycles for individual days of the week show the same broad features as the corresponding weekly-integrated diurnal σ_{ap} cycles (Fig. 4d) for all seasons, with the exception of differences between post-dusk and pre-dawn σ_{ap} for individual days of the week. During summer, post-dusk σ_{ap} is slightly larger than pre-dawn σ_{ap} for each day during Monday-Friday, leading to a gradual build-up of absorbing aerosols in the PBL. Post-dusk σ_{ap} is less than pre-dawn σ_{ap} on Saturday and Sunday. The resulting weekly σ_{ap} cycle (Fig. 4c) and the σ_{ap} cycles for individual days suggest a nearly-constant source of absorbing aerosols from sunrise to sunset, with largest emissions from Monday-Friday. Both interstate truck traffic and farming activities are consistent with the observed diurnal and weekly patterns during summer but truck traffic is likely the source more capable of contributing to the large summer diurnal and weekly σ_{ap} cycle amplitudes (60% and 40%, respectively), given the higher summer PBL heights in the region. The fall weekly σ_{ap} cycle (Fig. 4c) also exhibits a build-up of absorbing aerosols from Monday-Tuesday, followed by lower aerosol loading during the remainder of the week. This cycle is not consistent with known weekly cycles in truck traffic or agricultural practices near the site. Scattering coefficient exhibits a similar weekly cycle as σ_{ap} during autumn (Fig. 3c) and the weekly σ_{ap} and σ_{sp} cycle amplitudes are similar (~25%) during fall. Similarities in the autumn weekly σ_{ap} and σ_{sp} cycles could simply be the result of a smaller compensating effect on σ_{sp} from daytime secondary aerosol production during autumn (i.e. less photochemistry) or it could be due to sources of scattering and absorbing aerosols that are more similar in autumn than in summer. Diesel emissions from agricultural activity would seem more capable of contributing to the weekly σ_{ap} cycle during autumn, when PBL heights are lower. Biomass burning is a less likely source, even though Buzcu-Guven et al. (2007) reported a significant biomass-burning influence on OM mass (38%) at BND. Absorption Ångström exponent demonstrates minimal day of week variability during autumn (Fig.S15) and α_{ap} values of 1.1-1.2 are not statistically-different from the theoretical value of 1 for BC (Bergstrom et al., 2002). ”

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.

23.

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) to 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 depths: DJF(480m, 480m, 4080m); MAM(980m, 950m,1040m); JJA(1090m,1380m,1310m); SON(860m, 790m, 600m)."

In *Sect. 3.3 EGB*

"Holzworth (1964) used daily soundings at Buffalo, NY (located ~170 km SE of EGB) to calculate monthly mean maximum mixing layer depths: DJF (510, 480, 530m); MAM (780, 810, 1070 m); JJA(1180, 1440, 1360 m); SON (1190, 530, 700 m). "

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).”

24. 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:

Upon further inspection, we agree with the reviewer that there is minimal direct local traffic influence in fall. In response to reviewer #2 comment 2, we include a new section (Sect.2.3- Data processing, auality assurance, and calculated AOPs) in the revised manuscript. The section provides information on the quality assurance conducted, including how local sources are removed. Please also see our response to reviewer #2 comment 2, where we discuss this.

Changes in manuscript:

The paragraph discussing the fall diurnal σ_{ap} cycle at EGB (Sect. 4.1.4) now reads as follows:

“The autumn diurnal σ_{ap} cycle (Fig. 4e) appears to be less influenced by less convection and more influenced by frequent transport from the south (Fig. 7b), along with less regional traffic than during summer. Monthly-averaged σ_{ap} during September-October (Fig. 2b) remains near summer levels (except for August) but σ_{ap} is much lower for all wind sectors except the urban-influenced southerly wind sectors, for which σ_{ap} was similar in value to summer (Fig. 7b). The diurnal σ_{ap} and σ_{sp} cycles exhibit very little structure during fall so lower fall PBL heights may be partially offset by lower production of scattering and absorbing aerosol and/or more efficient removal mechanisms. Some additional source may be responsible for the early-week increase in σ_{ap} , similar to that observed during autumn at BND (Fig. 4c). The source of absorbing aerosol persists throughout the day and into the evening (not shown) and may be local agricultural activities.”

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. S/SE wind directions correspond to air masses arriving at EGB from the heavily-populated southern Ontario region (Fig.7), including Toronto (~70km south of EGB). Aerosol chemistry studies at EGB (Chan et al., 2010; Liggio, et al., 2010) report higher levels of pollution for air masses from the south, including higher EC levels and EC/CO ratios.

Changes in manuscript:

We now include pollution-rose diagrams for each season at each site. The pollution-rose diagrams show the dependence of scattering and absorption coefficients on wind sector for each season and site. (Figs.5-8). We use the pollution-rose diagrams along with published aerosol chemistry at the sites and known pollution sources to help explain aerosol temporal variability at the four sites (Sect(s) 4.1.2-4.1.5). In addition to σ_{ap} and σ_{sp} , we include pollution-rose diagrams for the following aerosol properties for each site/season in Fig(s) S16-S22 of Supplemental Materials: (1) PM1 scattering fraction; (2) single-scattering albedo; (3) hemispheric backscatter fraction. The pollution-rose diagrams for σ_{ap} and σ_{sp} measured at EGB are provided Fig.7 of revised manuscript.

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 (Fig. 9g) are also consistent with SAE being an indicator of the relative contribution of fine-mode aerosol to scattering.