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.)

- We thank anonymous reviewer #1 for her/his excellent suggestions, particularly those related to improvements in paper organization 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. We include a 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. In our responses to individual reviewer comments, all references to figure, table, and manuscript section numbers are numbers of the materials in the revised manuscript, unless otherwise-stated. As an example, a reference to *Table 1* means that the discussed material will be located in Table 1 of the revised manuscript, not Table 1 of the original manuscript.
- We number the individual comments below for easy referencing in this document and for cross-referencing between this document and *Response to Reviewer #2*. The individual reviewer comments are given in bold type-face. We follow each enumerated reviewer comment with authors' response and any changes in the manuscript. Our responses are in plain text.

Anonymous Reviewer 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.**

<u>Authors' Response:</u>

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 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

Section 2-Methodology

2.1 Air sampling infrastructure at the sites

2.2 Measurements and instruments

2.3 Data processing and quality assurance

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

2.4 Measurement uncertainties

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

2.5 Data analysis methods

- Section added to consolidate data analysis material.

Section 3-Site descriptions

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

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

3.2 Bondville, Illinois, USA (BND)

3.3 Egbert, Ontario, Canada (EGB)

3.4 Lamont, Oklahoma, USA (SGP)

Section 4-Results and discussion

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

4.1 Temporal variability of aerosol optical properties

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

4.1.1 Temporal variability common to all sites

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

4.1.2 Temporal variability at APP

4.1.3 Temporal variability at BND

4.1.4 Temporal variability at EGB

4.1.5 Temporal variability at SGP

4.2 Regional variability of aerosol optical properties

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

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

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

4.4 Systematic relationships among aerosol optical properties

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

4.4.1 Annual systematic relationships among aerosol optical properties

4.4.2 Seasonal relationships involving absorption Angstrom exponent

Section 5-Summary and conclusions

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

Section S1-Measurement uncertainties

- Detailed uncertainty analysis, including contributions of individual sources, propagation of uncertainties (including covariance between AOPs), and calculation of total and measurement precision

uncertainties, following an approach similar to Anderson and Ogren (1998) and Anderson et al. (1999). The results of the uncertainty analysis relevant to the comparisons of mean AOPs in the paper are summarized in Sect. 2.4 of the paper.

S1.1 Uncertainties in total and hemispheric backscatter coefficients

S1.2 Uncertainties in absorption coefficient

S1.3 Uncertainties in calculated aerosol optical properties

S1.4 Comparing measurements made at different locations and times

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

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

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

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

Section S4-Annual cycles of PM10 aerosol optical properties

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

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

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

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

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

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

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

2. **<u>Trend analysis:</u> 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 Δb=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 ~ 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 σ_{sp}=30Mm^{-1}, σ_{sp}=3.0Mm^{-1}, b=0.14, ω_0 =0.91, α_{sp}=2.0, and α_{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 quantity 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. **<u>Proxies:</u> 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.**

<u>Authors' response:</u>

We now include proxies for each site (pollution rose diagrams and reported PBL heights for studies conducted in the four regions. We do not believe that the results from the Seidel paper can be applied at the four sites. Seidel presents results of comparisons of PBL heights using seven techniques applied to radiosonde climatologies from several countries, including the U.S. However, the results shown are statistical calculations over the global dataset, along with seasonal variations at a few specific locations (Prague, Majuro, Marshall Islands, Oakland..) and latitudinally-averaged values integrated over all seasons.

<u>Changes in manuscript</u>

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

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, et al., 1964); and seasonal dependence of of mixing layer heights > 1km reported for each season near Buffalo, located ~170km SEof EGB (i Portelli et al., 1981). 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:
The weekly cycles of AOPs other than absorption coefficient are statistically-insignificant and/or minimal in most cases. The scattering coefficient demonstrates modest weekly and diurnal cycle amplitudes (typically ~10%). Single-scattering albedo and DRFE also demonstrate modest diurnal variability in summer and fall, as the result of larger absorption coefficient variability during those seasons (mainly summer). The other intensive AOPs demonstrate negligible and/or statistically-insignificant variability on weekly and diurnal timescales.
Changes in manuscript:
We have significantly shortened the discussion of weekly and diurnal cycles of AOPs, with the exception of absorption coefficient σ_{ap} (which demonstrates significant weekly and diurnal variability in summer and fall). We show the weekly and diurnal σ_{ap} cycles for all sites/seasons in Fig. 2 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). S2-S9). We briefly discuss any significant variability for these variables (i.e. scattering, summer and

fall ω_0 and DRFE) in the final paragraph of Sect. 4.1.1 and any significant site-specific variability in Sect(s) 4.1.2-4.1.5.

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.

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 5^{th} through 95^{th} 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((RH) ~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 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 nclude 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 RH≤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 RH≤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≤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≤40%.

(Sheridan, et al., 2001)."

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

Authors' response:

Scattering Ångström exponent 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. 8 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 Δb=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~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 the same approach taken by D&02002 and Andrews et al.(201l) and only use hours for which σ_{sp} at 550nm 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 (see Table 2). This filtering discards 1.4% of the hours at APP, 0.1% at BND, 1.8% at EGB, and 0.5% at SGP. The above-stated percentages are uniform across seasons, except for slightly higher percentage during fall at EGB and SGP (Table S6). We use all hours in calculating σ_{sp} and σ_{bsp} statistics to avoid a bias toward 'less clean' air conditions. "

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 quantity 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:

Dust is the only known super-micron aerosol measured at the rural, continental BND site. PM1 scattering fraction R_{sp} is slightly lower during cold-season months (Fig.2). When combined with elevated scattering in winter, relative to fall and spring) this implies more dust, likely wind-blown. The co-incident increase in PM1 scattering also implies more of some source of large, sub-micron particles, although we are not aware of any such source. It is very difficult to do more than conjecture the source for the higher scattering in winter at BND, based on the available measurements and proixies

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

Authors' response:

This is correct. In the case of APP, though, the seasonal cycle in scattering is much larger than that of absorption so that the seasonal cycle in SSA is very similar to the seasonal cycle in SSA. This can be seen qualitatively by examining the annual cycles for scattering coefficient, absorption coefficient, and SSA at APP (Fig.1). We do agree that this point was poorly-phrased and have attempted to improve its clarity.

Changes in manuscript:

We replaced the passage on P.26990 of the original manuscript with the following passage, now located in *Sect. 4.1.2 Temporal variability at APP*:

"*The relatively small annual winter-to-summer increase in σ_{ap} (~50%) compared to that of σ_{sp} (factor of 3 winter-to-summer increase) suggests that different sources and/or processes influence the annual PM1 σ_{sp} and σ_{ap} cycles at APP. The annual cycles in b, ω_0, and α_{ap} at APP clearly follow that of σ_{sp}. The summer σ_{sp} peak coincides with a distinct minima in b* (30-40% lower than all other seasons) and maxima in ω_0 (~0.07 higher than during winter).**"**

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. We delay discussion of the tendency toward smaller, darker aerosols under low loading conditions until Sect. 4.4 Systematic relationships among aerosol optical properties.

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 5and 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.

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:

We have included proxies such as pollution rose diagrams and reported climatologies of PBL heights to help explain some of the site-specific phenomena in Sect. 4.1. See our response to reviewer #1 comment 4. These proxies can help to explain some features of variability at the individual sites. Unfortunately, there are more cases than not where the proxies cannot account for these features and such features cannot be explained, given the available set of measurements and proxies. The examples listed above by the reviewer represent some of these cases. In these cases, all that we can do is hypothesize as to possible sources for the phenomena, based on knowledge of the sites.

Changes in manuscript:

We have included proxies such as pollution rose diagrams and reported climatologies of PBL heights to help explain some of the site-specific phenomena in Sect. 4.1. In other cases, we hypothesize as to significant sources of variability, based on the best available information. In a few cases, we needed to omit the discussion in order to keep the paper to a manageable length (See reviewer #1 Comment 5).

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_{ap} measured at EGB (Fig. 6, Fig. S18) 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_{ap} pollution-rose plots for each site and season in Fig(s). 4-7. The other pollution-rose plots are included in the Supplemental Materials (Fig(s). S10-S25). In most (but not all) cases, the scattering and pollution-rose plots are similar. We chose to include the σ_{ap} pollution-rose plots in the paper because σ_{ap} demonstrated the largest weekly and diurnal cycles. We reduced the number of figures in the paper by re-structuring the paper as described in our response to reviewer #1 comment 1.

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 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 border-line significant at this confidence level. (Fig. S9). 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 provide a tentative explanation of the AAE seasonal variability in *Sect. 4.1 Temporal variability common to all sites*. Specifically, we state that

"Absorption Angstrom exponent is lowest during summer months and highest during winter months at APP, BND and SGP. The summer-to-winter difference in α_{ap} is clearly larger at APP (~0.9) than at BND and SGP (~0.5). In the colder months, the combined values of α_{sp} and α_{ap} suggest a mix of absorbing aerosol such as black carbon (BC), along with brown carbon and/or dust (e.g., Cazorla et al., 2013) at SGP, BND, and APP. Monthly-mean α_{ap} is less than 1 during summer months at each site, with the lowest values at APP. Gyawali et al. (2009) performed simulations using Mie theory to show that α_{ap} values much less than 1 are possible (their Fig(s). 8 and 9) when absorbing particles are coated. Clarke et al (2007) also reported a large number of α_{ap}(470/660nm) values clustered between 0.7-1.1 for pollution plumes during extensive flights over North America as part the of the INTEX/ICARTT experiment in summer 2004, although they did not hypothesize as to the source of the low α_{ap} values."

We further elaborate on this in *Sect. 4.4.2 Seasonal relationships involving absorption Ångström exponent*

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:

The systematic variability plots for these four low altitude rural continental sites can be compared with those for the free troposphere measurementes reported in Andrews et al. (2011). In general, the AOPS at rural continental sites reported on here have similar covariances as was observed for the mountain sites (e.g., SSA decreasing for lower loading and SAE increasing for higher loading). None of the rural continental sites demonstrate systematic variability indicative of strong dust (e.g. Mount Waliguan and Izana) or biomass burning influences (e.g., Mount Lulin). Additionally, two of the data sets reported on in Andrews et al. (2011) were obtained from long term aircraft measurements over BND and SGP. The systematic variability of the surface AOPs at BND and SGP is quite similar to what is observed in the free troposphere over these sites. The one exception is the relationship between b and loading at BND: b decreases with loading at the surface and increases with loading in the free troposphere.

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:

Regions of the ω_0 vs.σsp curves with positive slopes indicate different sources of scattering and absorbing aerosols while the flat portions of the curve indicate that the sources of scattering and absorbing aerosols are the same. Stated differently, a flat portion of the ω_0 vs.σsp curve indicates that scattering and absorption increase or decrease together in a way such that the relative fractions of scattering and absorption to total aerosol light extinction (i.e. ω_0) remains constant. This is typically the result of adding more of the same type of aerosol, as compared to processes such as particle growth and/or deposition, which preferentially add or remove one type of aerosol and thereby modify the ω_0 of

the mixed aerosol. Based on this argument, we hypothesize that the difference in slopes (smaller slope for larger scattering) is due to a varying mix of aerosols that tends toward a common source of scattering and absorption at higher scattering values. with particle growth often leading to the larger relative contributions by scattering, as discussed in Sect. 4.1. The smaller slopes at higher scattering values seem to implicate primary sources for the highest scattering events, while the larger slopes at the lower scattering values could implicate particle growth and/or preferential removal of primarily scattering aerosols at low loading levels.

Changes in manuscript:

The first paragraph in *Section 4.4.1-Annual systematic relationships among AOPs* now reads as follows:

"Single scattering albedo increases and b decreases with increasing σ_{sp} at all sites (Fig(s). 8). 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, a condition representative of all months (Fig. 2). The co-variability of ω_o and b leads to a DRFE dependence on σ_{sp} that is statistically-insignificant for all sites (differences less than twice the DRFE measurement uncertainty). Greater influences by smaller, darker particles under low loading conditions and by larger, brighter particles under high loading conditions have been reported for SGP and BND by D&O2002. The tendency toward lower ω_o and higher b for low-loading conditions is consistent with preferential removal of large, scattering particles by cloud scavenging and/or deposition. It can also be the result of new particle formation with growth by condensation and/or coagulation to optically-active sizes (Andrews et al., 2011). Regions of the ω_0 vs. α_{sp} curves with positive slopes indicate different sources of scattering and absorbing aerosols while the flat portions of the curves correspond to the same sources of scattering and absorbing aerosols. Based on this argument, we hypothesize that the change in slopes with increasing σ_{sp} is due to a varying mix of aerosols that tends toward a common source of scattering and absorption for higher σ_{sp} values (σ_{sp} larger than ~50Mm^{-1}). "

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 (Fig(s). 8). 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, a condition representative of all months (Fig. 2). T<u>he co-variability of ω_o and b leads to a DRFE dependence on σ_{sp} that is statistically-insignificant for all sites (differences less than twice the DRFE measurement uncertainty)</u>."

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. S2 of Supplemental Materials and we display seasonal variability for the other 8 AOPs in Fig.1. 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.