## Comments on:

## "A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions" by J. P. Sherman, P. J. Sheridan, J. A. Ogren, E. A. Andrews, L. Schmeisser, A. Jefferson, and S. Sharma

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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.

## General comments:

- Structure of the paper: 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>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.

- 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.
- <u>Proxis</u>: Several proxis (pollution sources, wind sector, agriculture, PBL,...) can explain the various cycles at each station. These proxis 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.
- 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.

Specific comments:

- 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.
- 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.

- 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.
- P. 26980: please indicate the percentage of hours with RH>40% at EGB, perhaps also if an annual variability if measured for RH>40%.
- P. 26980: if possible give a reference for the hygroscopic dependence of light scattering
- 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?
- P. 26983: were negative scattering Ångström exponents never measured ?
- P. 26984: To my knowledge, the uncertainties on b and  $\beta$  are probably quite high.
- P. 26984: if hours with scattering coefficient lower than 1 Mm<sup>-1</sup> 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?
- § 3: the end of the first § and the second one contains information that should be given in the experimental section.
- P. 26986: the use of medians instead of means is appreciated because most of the used parameter are not normally distributed.
- P. 26988 Several stations/parameters present a decrease not only in fall (that is discussed), but also in spring (not discussed).
- 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 ?
  - Line 23: The cycle in single scattering albedo reflects the difference in scattering and absorption cycle.
  - Line 29: The single scattering albedo being an intensive properties and should therefore not depends on the amount of aerosol.
- 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).
- 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 ?
- 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 ?

- 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.
- 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) ?
- 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 ?
- 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 ?
- P. 27004 lines23-25: Is this really statistically significant regarding the inter-annual variability and the uncertainties ?
- P. 27005 lines18-20: where the medians done before to calculate the intensive properties or after ?
- § 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 ?
- 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 ?
- p. 27007 lines 20-25: Is it possible that these variations are just in the uncertainties ?
- 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"?
- § 3.4.4 The order of the explanation and of the item on the figure are opposite
- Table 1: are the cloud fraction and the spectrally-averaged surface albedo the same for the 4 stations ?
- A map with the stations would help the reader to understand the special changes of aerosol parameters
- Fig. 1 and 2: is there a reason to separate the annual cycles into 2 figures ?