Response to Referee Comments (RC2) for acp-2019-131 "Quantifying aerosol size distributions and their temporal variability in the Southern Great Plains, USA"

Referee Comments received on 13 July 2019

We would like to thank the reviewer for their time and comments. We have responded to their comments below.

The authors describe a quality controlled four-year dataset of aerosol size distribution with diameter ranging from 7 nm to 14 microns. The dataset was developed by combining measurements from SMPS, APS, and CPC at the DOE SGP site. Statistics of aerosol number, surface, and volume concentrations are presented for different seasons. The authors also carried out power spectral analysis of the temporal variation of aerosol size distribution and show a diurnal cycle in the concentration of small particles ranging from 7 to 30 nm for all four seasons. The diurnal variation is attributed to new particle formation.

The dataset will be useful for validating models, and future studies of aerosol processes. The key results presented largely confirm findings of earlier studies. The topic is well suited for Atmospheric Chemistry and Physics, and overall the manuscript is well written. Following are my comments and suggestions.

1) One focus of the manuscript is the quality-controlled aerosol size distribution dataset. Were particle losses through the inlet and inside APS (especially for coarse mode particles) taken into consideration?

Particle losses for the SMPS size distribution were estimated and accounted for in the SMPS data. Particle losses for the APS size distribution were not taken into consideration, but based on the authors' experiences, it is estimated that particle losses in the APS were likely small for most of the APS size distribution. We have added the following statement in the Appendix where we explain the quality control methodology.

"Here, it is important to note that estimated corrections were made to the SMPS size distributions to account for potential particle losses due to diffusion in the inlet and system tubing. Corrections were not made to the APS size distribution data for possible particle losses within the inlet and system tubing, but it is expected that these losses are likely small for most of the APS size distribution. For example, experiments have shown approximately unit transmission efficiencies for particles with diameters up to 4 μ m for the SGP inlet system. For larger sizes where low particle counts make it difficult to characterize transmission efficiencies experimentally, modeled transmission efficiencies predict significantly increasing biases for particles with diameters greater than ~10 μ m (Bullard et al., 2017)."

We have also added a clarifying statement in the text (Page 5, Line 4), where we mention a decrease in the transmission efficiency in the APS for the largest particles, and now explicitly state that this was not corrected for in this dataset.

Reference: Bullard, R. L., Kuang, C., Uin, J., Smith, S. and Springston, S. R.: Aerosol Inlet Characterization Experiment Report. U.S. Department of Energy ARM Climate Research Facility. DOE/SC-ARM-TR-191, doi:10.2172/1355300, 2017.

2) Equation 1- I don't think this is how lognormal aerosol size distribution is defined Is N(In(Dp) cumulative size distribution? If so, the limits of integration are incorrect.

We have updated this equation to accurately reflect the aerosol number size distribution, as opposed to the aerosol number concentration within a specific size bin, which was present in the original version.

3) Page 5, Line 26: Reference Wang et al., 2009 is missing.

We have included this reference to the reference list.

4) Equation 3: please check the numerator on the righthand side.

We thank the reviewer for catching the additional power of two that showed up in the numerator of this equation. We have fixed the numerator of Equation 3.

5) Page 9, line 12: The peak of small particle concentration occurs around UTC 22-24 (CST 16:00-18:00) during winter. I am wondering if boundary layer deepens until CST16:00-18:00 during winter time.

Unfortunately, boundary layer height data at SGP for our 2009-2013 period are estimated from 4x daily (at approximately 5:30, 11:30, 17:30, and 23:30 UTC) radiosondes that cannot resolve exactly when the boundary layer height reaches its maximum depth, which is typically around 20:00-21:00 UTC in the winter time but can be later (Liu and Liang, 2010). However, we are not stating that the timing of peak boundary layer depth needs to occur simultaneously with the peak concentrations of N_{7-30nm} at the surface, as it can take up to several hours to mix aerosol particles at the top of the boundary layer down to the surface, depending on their altitude and the vertical mixing time scales (e.g., Chen et al. 2018). Also, this vertical mixing process would typically be slower in wintertime boundary layers that are more stable than in the other seasons, which may also partly explain the several-hour shift in peak N_{7-30nm} concentrations in the winter time. We have added some additional discussion about this vertical mixing process as well as an additional figure (see below) and a related discussion regarding the seasonal evolution of the SGP boundary layer for the 5-year focus period of this study. With these changes, we believe that we have provided additional evidence to support our statements regarding the importance of the boundary layer development for each season's N_{7-30nm} diurnal cycle.



Figure 9: Diurnal cycle of boundary layer heights at SGP for each season, as estimated from radiosonde data. The circles represent the median boundary layer height for the top 25% of the weekly data in terms of power associated with the diurnal cycle in N7-30nm (High Power). Similarly, the diamonds represent the median boundary layer height for the bottom 25% of the weekly data (Low Power). The horizontal lines above and below the circles and diamonds represent the 25th and 75th percentiles (interquartile ranges) for this data. The numbers in parentheses represent the number of weekly time periods used in this analysis. The abscissa offset for each radiosonde launch time is for viewing purposes and does not reflect any shift in timing for each of the 4 radiosonde launch times for the different seasons.

6) Page 10, line 26: "The peak concentrations of the 12-hour cycle for all seasons occurred between 04 and 12 UTC (23 and 07 CDT) and between 16 and 24 UTC (11 and 19 CDT) for both N_T and N7-30nm." There is no second peak for N_T or N7-30nm between 4 and 12 UTC, at least for MAM and DJF (Figures 5 and 7).

We are unclear what the reviewer is referring to in terms of their comment that "there is no second peak for NT or N7-30nm between 4 and 12 UTC," as there is no figure that corresponds to this statement in the manuscript. In Figures 5 and 7, there is a second significant peak in the power spectrum for the 12-hour cycle, particularly for ALL, MAM, and DJF. We believe the reviewer may be referring to Figures 6 and 8 when making the above statement. However, Figures 6 and 8 are only representative of the 24-hour cycle and does not include any information about the 12-hour cycle.

Below are the same figures as Figures 6 and 8a, but for the 12-hour cycle, which pictorially demonstrates the statement highlighted by the referee above. However, since this is only a

minor point in the manuscript, we believe that including these figures in the manuscript is not necessary. We did add the phrase "(not shown)" in the manuscript after the statement to make it clearer that this statement cannot be found in any of the figures shown in the current manuscript.



Figure (Above): Normalized frequency of the daily time of peak concentrations associated with the 12-hour cycle in N_T . This figure only includes weekly data chunks that had normalized power associated with the 12-hour cycle greater than that of the corresponding seasonal estimate of the red noise spectrum power. The numbers in parentheses represent the number of weekly data chunks that met this criterion.



Figure (Above): Normalized frequency of the daily time of peak concentrations associated with the 12-hour cycle in N_{7-30nm} . This figure only includes weekly data chunks that had normalized power associated with the 12-hour cycle greater than that of the corresponding seasonal estimate of the red noise spectrum power. The numbers in parentheses represent the number of weekly data chunks that met this criterion.