We are very thankful to both the referees and the editor for their comments and suggestions. We acknowledge both referees' recommendation to have this manuscript resubmitted at a later date with more mature findings. However, in the light of the editor's further guidance on this issue, we have revised the manuscript in several places as outlined below. We sincerely hope that these revisions not only satisfy the editor's (and the journal's) expectations of a campaign overview paper, but also address at least some of the concerns raised by the referees.

1. The following text was added at the end of the first paragraph in the Introduction section:

"This paper provides an overview of the CARES project, and presents: 1) the scientific background and motivation for the field campaign, 2) the operational and logistical information pertinent to the execution of the campaign, 3) an overview of key observations and initial findings from the aircraft and ground-based sampling platforms, and 4) a roadmap of planned data analyses and focused modeling efforts that will facilitate the integration of new knowledge into regional and global climate-chemistry models."

2. The following short paragraph was added at the end of the Introduction section:

"The goal of this overview paper is to provide a coherent description of the project objectives, a campaign summary, and a context for mature scientific results that will be reported in future publications. We begin in section 2 with a brief review of previous research related to the CARES science questions. In section 3, we describe the campaign venue, sampling platforms (ground sites, aircraft), and the associated instruments and measurements. In section 4, we present an overview of the key observations from the various airborne and ground-based instruments. In section 5, we conclude with a summary of main initial findings and a roadmap for future work."

3. Some non critical text and references in section 2 (Brief Review of Previous Research Related to CARES Objectives) were deleted without impacting the main function of this section.

4. The following paragraph was added in section 4.3.5 (Offline Analyses of Particle Samples) to highlight new CARES results from a paper that is presently in press in Journal of Geophysical Research.

"CCSEM/EDX and STXM/NEXAFS analyses of TRAC-collected particles onboard the G-1 on June 15 (a strong SW flow event) showed the presence of sea salt particles over Sacramento. Forward trajectories simulated by WRF (Fast et al., 2012) coupled with Lagrangian particle dispersion model (Doran et al., 2008) analysis confirmed that the sea salt particles were transported from the Pacific Ocean/Bay Area. Interestingly, these particles were found to be internally mixed with organics, which were likely SOA species formed in these particles during the transit from the Bay Area. Surprisingly, these particles had experienced substantial chloride depletion that could not be explained by the known reactivity of sea salt with nitric and sulfuric acids. This study, recently published by Laskin et al. (2012), is the first field evidence that SOA, consisting of weak organic acids, may effectively react with sea salt particles and displace HCl gas, leaving behind particles depleted in chloride and enriched in the corresponding organic salts."

5. Previous section 5 (Outline of envisioned analyses) was rewritten to include a summary of main initial results from the campaign. The new section 5, labeled "Summary and Future Directions," is reproduced verbatim below:

"5. Summary and Future Directions

The CARES field campaign was designed to examine the interaction between anthropogenic and biogenic emissions in SOA formation, black carbon ageing, and their effects on the associated optical and CCN activation properties. The campaign was carried out from June 2nd through 28th, 2010, in

Central Valley, California, centered on the Sacramento urban area. Two heavily-instrumented ground sites – one within the Sacramento urban area (site T0) and another in Cool, CA, a small town about 40 km to the northeast in the foothills area (site T1) – were set up to characterize the evolution of meteorological variables, trace gases, aerosol precursors, aerosol size, composition, and optical and CCN activation properties in freshly polluted and aged urban air. On selected days, the DOE G-1 aircraft was deployed to make similar measurements upwind and across the evolving Sacramento plume in the morning and again in the afternoon. The NASA B-200 aircraft, carrying remote sensing instruments (HSRL and RSP), was also deployed to characterize the vertical and horizontal distribution of aerosols and aerosol optical properties within and around the plume. The CARES campaign overlapped temporally with the CalNex campaign in the Central Valley and Southern California regions in May and June 2010. As part of CalNex, the NOAA Twin Otter aircraft, carrying a combination of downward-looking ozone/aerosol and Doppler wind lidars and a multi-axis DOAS system, moved its operation from Southern California to Sacramento to collaborate with CARES from June 14th to 28th. The main initial findings from the CARES campaign are summarized below:

- On approximately 20 days out of a total of 27, the Sacramento urban plume transport was largely controlled by southwesterly winds that drew the polluted air to the northeast over the forested areas in the Sierra Nevada foothills where it mixed with biogenic emissions by late afternoon or early evening. On the remaining ~7 days (June 10-13, 16, and 20-21), the southwesterly wind pattern was interrupted by northwesterly flows, which transported the Sacramento plume to the southeast into the San Joaquin Valley, where there was relatively much less mixing with biogenic emissions.
- The period from June 22nd through 28th also experienced a steady buildup of aged aerosols due to recirculation of air in the region, coupled with warmer temperatures toward the end of June. These conditions resulted in the highest pollution days at the end of the campaign, from June 25th through 28th. Observations across the relatively cleaner and more polluted periods as well as across the SW and NW flow regimes thus provide an exceptional opportunity to examine aerosol formation and evolution processes in the same region under a range of environmental conditions.
- The urban site T0 experienced significantly higher mixing ratios of the primary emission species such as CO, NO_y, and anthropogenic VOCs compared to the rural foothills site T1, and the diurnal behaviors of these species were also similar to each other, as expected. In contrast, the diurnal behavior of biogenic isoprene mixing ratios at both the sites followed that of the surface temperatures. The peak mixing ratios ranged between 2 and 12 ppbv around 14:00 PDT while the minimum mixing ratios were nearly zero from midnight until dawn. Since the T1 site was located amidst biogenic emissions, isoprene mixing ratios there were generally about 0.5 to 3 ppbv higher than at T0. The diurnal behavior of photochemically produced O_3 at the T0 and T1 sites were quite similar despite the marked differences in the precursor trace gas composition and concentrations between the two sites. The highs ranged between 60 and 80 ppbv, except for a peak of nearly 120 ppbv on June 28th. The daily O_3 peaks at T0 typically occurred around 15:00 PDT while it was often delayed by ~3 h at T1 on days when the urban plume was transported to the site during the SW flow periods.
- Sub-micron non-refractory aerosol composition observed at both the ground sites and aboard the G-1 aircraft was dominated by organics, followed by sulfate, followed by nitrate and ammonium, while chloride was negligibly small. OA concentrations at the ground sites ranged between <0.5 and 10 $\mu g\ m^{-3}$ STP and displayed a diurnal cycle that was similar to that of O_3 at both sites, which is consistent with photochemical production of SOA from anthropogenic and biogenic VOCs. OA concentrations typically peaked at the T0 site around 15:00 PDT while it peaked around 18:00 PDT

or later at the T1 site as the urban plume was transported to the foothills area during the SW flow periods. Enhanced concentrations of aged organic aerosols, likely recirculated from the foothills area the previous day, were often observed in the residual layer during the morning flights over the Sacramento urban area, which is consistent with preliminary WRF simulation results presented in Fast et al. (2012). The period from June 22nd to 28th experienced a steady buildup of OA, with concentrations reaching more than 25 μg m^{-3} STP aloft in the urban plume as it was transported east to the foothills area on the afternoon of the 28th.

- On selected days during both SW and NW flow periods, the daytime evolution of key trace gases, aerosol composition, mixing state, size distribution, and optical properties in the Sacramento urban plume was observed by the G-1 as it sampled upwind of, across, and downwind of the drifting plume in the morning and afternoon. These semi-Lagrangian in situ observations were complemented by NASA B-200 observations of vertical profiles of aerosol optical properties, which provided a more complete picture of the 3-dimensional structure of the evolving urban plume and the surrounding air. In the latter half of the campaign, this picture was further enhanced by NOAA Twin Otter observations of vertical profiles of O_3, NO_2, HCHO, CHOCHO, and wind speed in Sacramento and surrounding areas.

- Single particle mass spectrometers (SP-MS) deployed on the G-1 (A-ATOFMS), at T0 (SPLAT II), and at T1 (PALMS) also showed that the vast majority of aerosol particles characterized during the study were composed of oxygenated organics mixed with various amounts of sulfates: from sulfate-dominated particles to those containing mostly organic species. In addition, fresh and processed soot particles, biomass burning aerosol, amines, sea salt (both fresh and processed), and a small number of mineral dust and other inorganic particles were observed. The A-ATOFMS provided semi-Lagrangian aerial snapshots of particle mixing states in the evolving urban plume. In contrast, the SPLAT II, which was operated almost continuously for the entire campaign period, provided a detailed view of the evolution of relative fractions of different primary and secondary particle types in a single day, albeit at a fixed urban site. Size- and composition-selected SOA-dominated particles were also analyzed using SPLAT II to study evaporation kinetics at room temperature (Vaden et al., 2011a). This study is the first to present field evidence that evaporation of these ambient organic particles was extremely slow and size-independent, suggesting that the particles were in a quasi-solid state.

- SP2 instruments (outfitted with a scattering channel) deployed on the G-1 and at both the ground sites provided data on BC number and mass concentrations for particles between 60 and 600 nm BC mass equivalent diameters as well as coating state data for particles between ~175 and 350 nm. Preliminary analysis of G-1 SP2 data for June 28th showed an increase in the coating mass to BC mass ratio in the urban plume between the morning and afternoon flights. Particles were also collected using TRAC and DRUM samplers on all three platforms for offline analyses to further probe the chemical composition, mixing state, and morphology.

- SO_2 emitted from oil refineries in the Bay Area appears to have been routinely transported to the Sacramento area during the SW flow periods. These SO_2 plumes were associated with increased number concentrations of ultrafine and Aitken mode particles, which were likely nucleated via H_2SO_4 formation from SO_2 photooxidation, followed by growth to the observed sizes during transit. The Aitken mode was typically observed at both the ground sites in the morning around 09:30 PDT, followed by continued growth to accumulation mode sizes until mid-afternoon, likely due to condensation of photochemically formed SOA species. In contrast, SO_2 mixing ratios were negligibly small and the Aitken mode aerosol number concentrations were also significantly lower in the sampling domain during the NW flow periods.

- Coarse mode aerosols, mostly consisting of sea salt, were found to be transported from the Pacific Ocean/Bay Area to the Sacramento area on several occasions during the SW flow periods. Electron microscopy and X-ray spectro-microscopy analysis of TRAC collected particles onboard the G-1 over Sacramento on June 15 (a strong SW flow event) revealed that the sea salt particles were internally mixed with organics, which are likely SOA species formed in these particles during transit from the Bay Area. Surprisingly, these particles had experienced substantial chloride depletion that could not be explained by the known reactivity of sea salt with nitric and sulfuric acids (Laskin et al., 2012). This study is the first to present field evidence that SOA, consisting of weak organic acids, may effectively react with sea salt particles and displace HCl gas, leaving behind particles depleted in chloride and enriched in the corresponding organic salts.
- In situ aerosol optical properties were measured at near-UV and visible spectral regions onboard the G-1 and at T0 and T1 ground sites with several instruments, including nephelometer, PSAP, and PAS. A CRDS instrument was also deployed at the ground sites. When the coarse mode particles were present, the scattering coefficients (λ = 550 nm) reached as high as 40 to 75 Mm^{-1} and 20 to 50 Mm^{-1} at the T0 and T1 sites, respectively, and were about 2 to 4 times higher than the scattering coefficients of submicron particles. In contrast, the scattering coefficients ranged between 4 and 20 Mm^{-1} when the coarse mode particles were largely absent during first week of June and later again during the NW flow periods. MFRSR observations at each ground site provided the diurnal, day-to-day, and site-by-site variations in column aerosol microphysical and optical properties, such as aerosol optical depth, single scattering albedo, and asymmetry parameter. Consistent with the in situ optical properties measurements, MFRSR AOD values were observed to range from about 0.025 (representing very clean air) at the start of the campaign to about 0.12 towards the end of the campaign. Additionally, coarse aerosol mode size distributions derived from MFRSR data were also found to be consistent with the in situ size distribution measurements at both ground sites.
- Observations of CCN concentrations are also available at multiple supersaturations (0.07 to 0.5 %) at both ground sites. The T1 site also included measurement of size-resolved CCN concentrations and variable relative humidity nephelometry (commonly referred to as *f*(RH) measurement).

The CARES measurements have been processed and uploaded into the final ARM data archive (http://campaign.arm.gov/cares). These measurements comprise a rich data set for: 1) investigating SOA formation from anthropogenic and biogenic precursors and the potential interactions between them; 2) characterizing the time scales of BC ageing and evolution of its mixing state; and 3) quantifying the roles of BC mixing state, organics, and coarse mode aerosols on the observed optical and CCN activation properties. As mentioned throughout this paper, several detailed studies using various CARES data are planned or presently underway and will be reported via subsequent publications in this special issue and elsewhere as appropriate. Here we briefly summarize some of the key science questions that can be investigated using CARES data.

Secondary Organic Aerosols

a) Can we reliably infer IVOC (and SVOC) concentrations in the urban air, how they varied diurnally and scaled with VOC and CO concentrations, and how did these aerosol precursor gases correlate with organic aerosol number and mass concentrations in freshly polluted urban air?

b) What were the chemical composition, volatility spectrum, and hygroscopicity of OA, and how did they evolve as a function of atmospheric processing time and photochemical age?

c) Is there evidence for enhanced SOA formation in the urban plume when it mixed with biogenic emissions (e.g., during southwesterly flow conditions) compared to instances when the urban plume did not mix with biogenic emissions (e.g., during northwesterly flow conditions)? Can the observed SOA in the aged urban plume be separated into anthropogenic and biogenic fractions using carbon isotope analyses and other methods based on PTRMS observations and positive matrix factorization of AMS mass spectra?

d) How did the size distribution of aerosols evolve with SOA formation in the urban plume? Does the SOA condensation kinetics appear to be driven by Raoult's Law type gas absorption thermodynamics or is it similar to that of a condensing non-volatile species?

e) What was the role of organic species in the observed growth of ultrafine particles to CCN and optically active sizes?

Aerosol Mixing State

a) What was the distribution of BC mass fraction (BC mixing state) as a function of particle size in fresh and aged urban plumes? How rapidly did POA, BC, SOA, and inorganics become internally mixed?

b) What were the relative roles of condensation and coagulation processes in shaping the aerosol composition and size distribution?

c) What were the contributions and mixing states of other primary emissions such as biomass burning aerosol, mineral dust and sea salt, and how did these aerosols evolve?

d) What were the effects of aerosol mixing state on the ensemble aerosol optical properties, hygroscopicity, and CCN activity?

Aerosol Optical Properties

a) What was the role of changes in BC mixing state and morphology on enhanced light absorption?

b) Was there increased (by OA over BC) near-UV absorption? Did OA absorption extend into the visible part of the spectrum? If so, how did it relate to OA composition?

c) What were the absolute and relative contributions of sub-micron and super-micron aerosols to the total aerosol direct radiative forcing?

d) Which compounds or particle types have the strongest radiative impacts, and can these be related to specific emission sources or atmospheric formation processes?

The resulting detailed picture for the evolution of different types of carbonaceous aerosols and their optical and CCN activation properties will then help improve the key aerosol process and property modules that are used in regional and global climate models. Specific modeling studies that are planned by CARES participants include:

- Local closures for optical and CCN activation properties.
- Constrained Lagrangian modeling of SOA formation and interactions between anthropogenic and biogenic emissions.
- Constrained Lagrangian modeling of black carbon mixing state evolution.
- Regional simulations of SOA formation that include long-range transport of trace gases and aerosols
- Assessments of new treatments of SOA and aerosol mixing state on aerosol optical and hygroscopic properties and their impact on radiative forcing over California and surrounding regions.

In closing, it is reiterated that the purpose of this early overview paper is to summarize the scientific objectives, the platforms and instrumentation, the sampling strategies, and the key observations collected during the campaign, and develop an initial list of specific science questions that could be investigated with the CARES data set. It is hoped that this paper will facilitate further analyses of this remarkably rich data set as well as stimulate ideas for novel, collaborative studies."

Additionally, we have addressed all the specific comments of Referee #1 as follows:

Comment: Page 1303 line 23. The Zhang et al. 2007 reference refers solely to AMS data, which does not include refractory components of submicron particles. One should be careful on making statements on the organic mass fraction based on these types of data. One could say, carbonaceous compounds may constitute up to 90 % of the dry nonrefractory submicron particle mass

Response: Good point. We have added "non-refractory" in the revised manuscript.

Comment: Page 1319, line 10. Add references?

Response: Two references were added: "Trace gas measurements included CO (Kleinman et al., 2007), NO, NO_2, NO_y, O_3, and SO_2 (Springston et al., 2005)."

Kleinman, L. I., Daum, P. H., Lee, Y.-N., Senum, G., Springston, S. R., Wang, J., Berkowitz, C., Hubbe, J., Zaveri, R. A., Brechtel, F. J., Jayne, J. T., and Onasch, T. B.: Aircraft observations of aerosol composition in New England and Mid-Atlantic States during the summer 2002 NEAQS Field Campaign, J. Geophys. Res., 112, D09310, doi:10.1029/2006JD007786, 2007.

Springston, S. R., Kleinman, L. I., Nunnermacker, L. J., Brechtel, F., Lee, Y. -N., and Wang, J.: Chemical evolution of an isolated power plant plume during the TexAQs 2000 study. Atmos. Environ., 39, 3431-3443, 2005.

Comment: Page 1320, line 3, change wit to with.

Response: Typo has been fixed.

Comment: Comparisons are made between the aircraft and ground sites for both gas and particle components. Were any measurements made prior/during/after the mission to assess the agreement (precision) of these paired instruments. No discussion is given assessing the quantitative accuracy of the AMS data. Why not compare to predicted mass from the measured size distributions, as often done by other investigators to get a rough idea on the data quality.

Response: Side-by-side tests were made before the campaign for some instruments (e.g., PAS), but it was not feasible to do conduct this exercise (before, during, or after the campaign) for most instruments because of logistical challenges.

We have added a brief discussion on the accuracy of AMS data in the revised manuscript as follows:

"Comparisons of the estimated and measured aerosol volumes for all three platforms are shown in Figure S1 (in Supplementary Material). The estimated volumes were calculated from the AMS species and BC masses using density of 1.75 g cm^{-3} for sulfate, nitrate, and ammonium, 1.53 g cm^{-3} for chloride, and 1.8 g cm^{-3} for BC. While some day-to-day variations in the agreement between the estimated and measured volumes were observed, especially for the G-1 on June 27 and 28, the overall agreements were reasonably good with regression fit slopes of 0.91 for T0, 1.0 for T1 and 1.32 for G-1. Further analysis is needed to determine the source of discrepancy in the G-1 data for June 27 and 28."

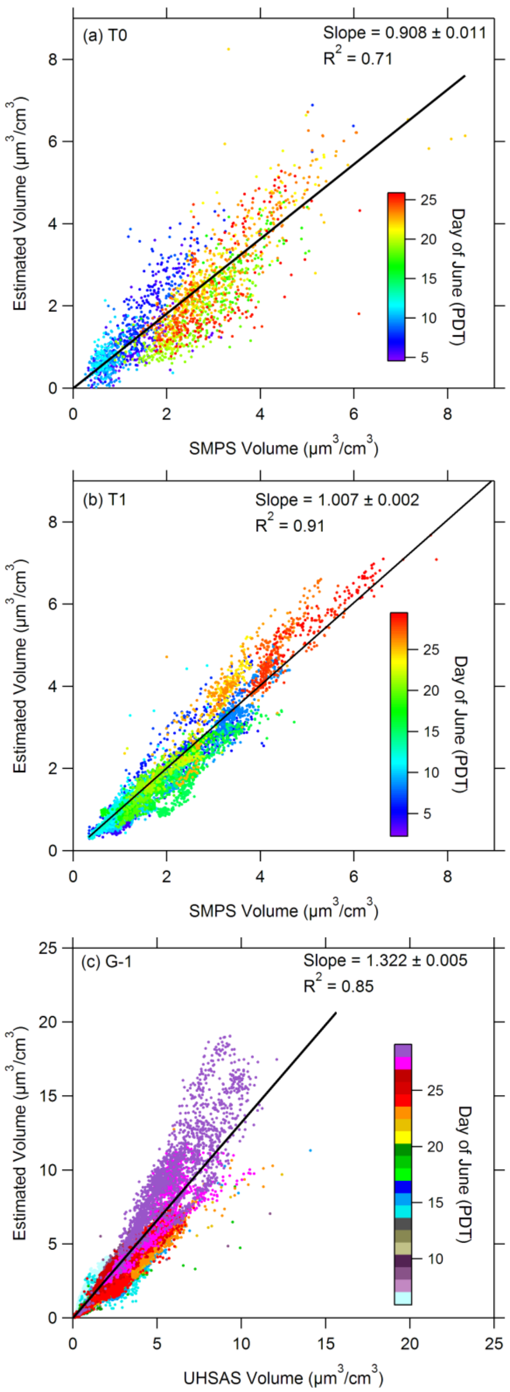

Fig. S1. Comparison of estimated and measured aerosol volumes for (a) T0, (b) T1, and (c) G1 (UHSAS volume is restricted to below 0.5 µm). Some day-to-day variations in the agreement between estimated and measured volumes are observed for all three platforms, especially for the G-1 data for June 27 and 28, but the overall agreements are reasonably good.

Comment: Figure 11. It makes sense that the contour scales for the APS are much lower than the SMPS due higher number concentrations associated with small particle sizes. However, can the authors comment on the comparison between these two instruments in their size overlap region?

Response: We have added a brief discussion as follows:

"Comparison of the number size distributions measured by the SMPS and APS in the overlap region is illustrated in Figure S2. Aerodynamic diameter from APS was converted to geometric diameter by assuming a density of 2.25 g cm^{-3} for coarse mode particles, which were found to be sea salt aerosols as discussed below. Number concentrations from the two instruments appear to agree very well around 0.56 μm geometric diameter."

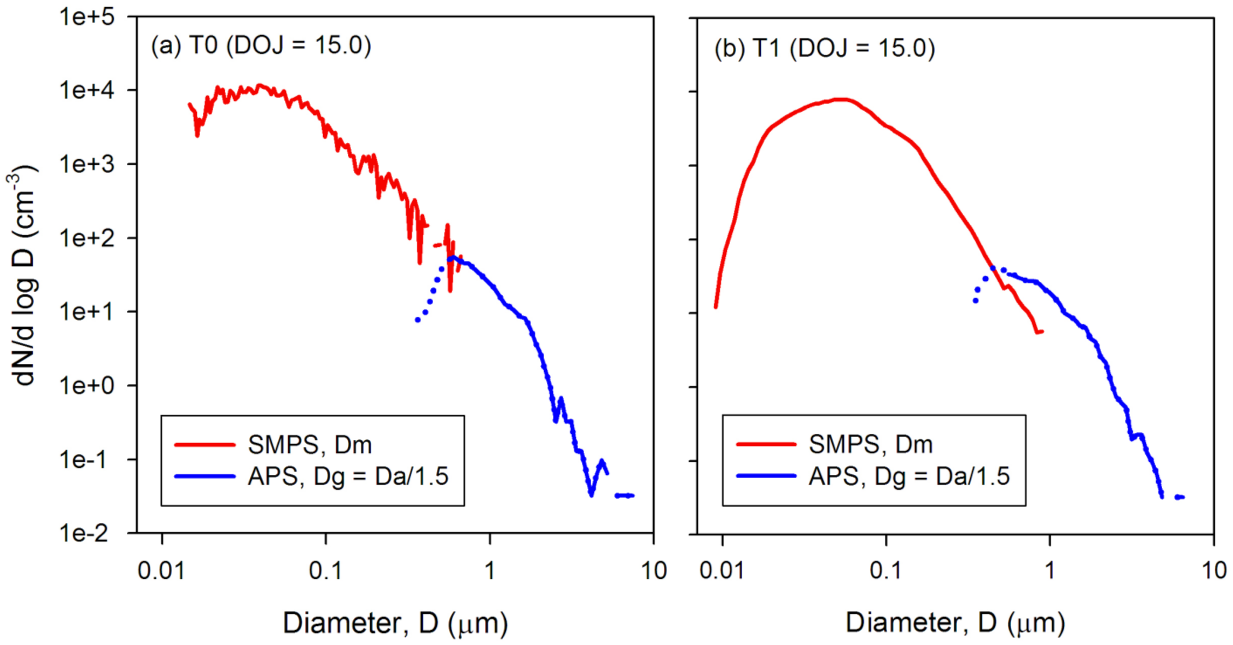

Fig. S2. Examples of comparison of the number size distributions measured by the SMPS and APS in the overlap region: (a) T0 and (b) T1, both on midnight of June 15. SMPS measures mobility diameter (D_m), which is equal to geometric diameter (D_g), assuming the particles are spherical. Density of coarse mode particles is assumed to be that of sea salt (ρ = 2.25 g cm^{-3}), and the APS aerodynamic diameter (D_a) is divided by 1.5 (i.e., square root of 2.25) to convert it to geometric diameter (D_g), assuming the particles are spherical. The dotted portion of the APS size distribution represents first 7 bins (D_a = 0.523 to 0.777 μm) where the APS appears to underestimate the number concentration compared to SMPS.

Comment: Page 1331, line 28, it is stated the APS measures up to 20 μm. Is it possible to sample particles that size. Maybe a few words are need on how inlet and sampling losses are handled and what, in practice, is the real upper size limit. Along these lines, is it even reasonable to plot up to 10um in Fig 11, and is the drop-off in the size APS size distributions at about 5um due to no particles in the atmosphere or just sampling limitations? Sampling efficiencies of large particles are again an issue in the optical section (4.4.1). It is unclear how much of the results, such as most surface area associated with particles smaller than 5um diameter, is due to sampling.

Response: The APS (TSI, Shoreview, MN) is indeed designed to sample up to 20 μm aerodynamic diameter. It was placed directly below the inlet (i.e., at the bottom of the vertical column) where it drew air at a flow rate of 5 L min^{-1} of the 120 total L min^{-1}. This placement prevented any bends in the tubing and thus minimized any inertial impaction losses of coarse particles. The “rain hat” on the top of the inlet stack is estimated to allow particles of at least 30,000 nm, although we did not actually characterize the inlet system on site for particle losses. We have added text to this effect in the manuscript. We believe that our APS data are robust, and they indicate that 95% of the surface area was below 5 μm. The 10 μm channel scattering data at both the ground sites are also consistent with the corresponding APS data, and a more comprehensive analysis of all the optical data in combination with aerosol size and composition data is presently underway and will be reported in a separate paper.