

Reviewer #1

We highly appreciate the valuable comments on our manuscript and the great editing. Your comments were quite helpful and we have incorporated them when possible into the revised paper. We hope that you will be satisfied with our responses and the corresponding revisions for the original manuscript. Please find below the comments/suggestions (bold blue color) and our responses (red color, with manuscript changes indicated in red italic).

Treatment of truncation error of TSI 3563 nephelometer in the retrieval process is not clearly described. The authors only mentioned that the optical closure was done according to Liu and Daum, 2000 and Mack et al. 2010. In Mack et al. 2010, the measured optical properties by TSI 3563 nephelometer were corrected according the empirical parameterization by Andsen and Ogren (1998), assuming only submicron particles were present.

The conventional truncation error correction (Anderson and Ogren, 1998) has been developed for typical bimodal size distributions with sub- μm and super- μm particles. Since this correction works reasonably well for common atmospheric situations (e.g., Anderson et al., 1999; Titos et al., 2014), it has been implemented into the standard data processing and quality control procedure supported by the ARM Program. We have modified the manuscript to reflect this.

(pages 18-19, lines 489-503): “The inclusion of a non-Lambertian light source and incomplete angular integration are well-known nonidealities of the integrating nephelometer TSI 3563. To reduce systematic errors associated with these nonidealities, several conventional corrections have been suggested for both slightly absorbing (e.g., Anderson and Ogren; 1998) and highly absorbing (e.g., Bond et al., 2009) particles. In particular, Anderson and Ogren (1998) have demonstrated through detailed Mie calculations and typical bimodal size distributions that systematic truncation errors are quite small (<10%) and substantial (up to 50%) for sub- μm and super- μm particles, respectively. Also, Anderson and Ogren (1998) have suggested a conventional truncation error correction, which is a common premise in many studies based on integrated nephelometry (e.g., Anderson et al., 1999; Titos et al., 2014). For our study, we use publically available and corrected data for TSI 3563 integrating nephelometer obtained from the ARM Archive. The corrections that have been applied include the conventional truncation error correction according to Anderson and Ogren (1998), and form the basis of the standard initial data processing and quality control procedure. The latter is implemented in the ARM data ingest protocol for Aerosol Observing System (AOS) with the TSI 3563 integrating nephelometer.”

The uncertainties of size distribution, density, and refractive index etc. have been introduced by this correction to the observed optical properties. I would suggest the authors to correct the angular non-idealism according to the measured truncation error of TSI 3563 nephelometer (Anderson et al. 1996) in the MIE calculation in order to simulate the nephelometer output optical properties, instead of correcting the measured optical properties directly.

We agree with the reviewer that Mie calculations can be successfully applied for simulating nephelometer measurements with well-known nonidealities (please see the discussion in the previous paragraphs). In this application, however, estimates of the total and backscattering coefficients, which are needed later in the analysis, require us to use the standard data processing procedures. In addition, following the reviewer’s suggestion would require us to use uncorrected but quality controlled data from the nephelometer. Given that such data are not generally available, we have elected to use only the standard corrected nephelometer data so that results are more widely applicable within the research community.

Comparison with the alignment method (1) On P. 4954 L. 10, the authors mentioned that the size range of TSI SMPS is 0.01-0.48 μm . What type model of SMPS was used in this study? What is the full size range of this SMPS?

The SMPS used in this study is a TSI Model 3936 with a long-column DMA and a 3772 CPC; the size range of 10 - 478 nm was chosen before the deployment as a tradeoff between range and resolution (time and size). Please note that we had no control over the SMPS data collection protocols, sensor configuration, QC, and data processing.

(2) If the full size range of SMPS is greater than 480 nm and overlaps with APS size range (0.52-19.8 μm), how does the APS size distribution match the SMPS size distribution in the overlapping geometric size range when applied with the retrieved time dependent densities from this study?

Based on this comment, details on merging the SMPS and APS size distributions have been added to the text. We appreciate the fact that the reviewer's comment indicated more explanation was necessary.

(pages 15-16, lines 392-418): "Before considering examples of the merged SMPS-APS distributions (Figure 8), two points should be made. First, the upper size limit of the SMPS used in this study is 0.48 μm (electrical mobility size), while the lower size limit of APS is 0.52 μm (aerodynamic size) (section 2). Second, the APS data near the lower limit are typically characterized by high uncertainties, and thus these data are avoided during the size distribution merging (e.g., Khlystov et al., 2004; Figure 1). Similar to previous studies, we found that the APS data considered here frequently have unreliable counting for the first three bins (size range 0.52-0.58 μm). Therefore, a sufficient overlap (e.g., geometric size range about 0.38-0.48 μm) between the measured SMPS and reliable APS size distributions does not exist. As a result, a direct application of the conventional alignment method for a given SMPS-APS dataset is not possible.

To merge the SMPS and APS distributions, we apply a simple approach (section 2) using the general framework that forms the basis of the conventional alignment method. We start with the replacement of the highly uncertain APS data for the three first bins by those obtained from a linear logarithmic extrapolation (log-log scale). This extrapolation involves reliable APS data from nearby bins (size range 0.58-0.67 μm) and provides "corrected" data for the first three bins (size range 0.52-0.58 μm) only. Then, the "corrected" APS size distribution is shifted horizontally along the abscissa D_p according to an assumed value of the effective density -- a procedure similar to the alignment method (Figure 1a). If during such horizontal shifting a fraction of APS spectra overlaps with fixed SMPS distribution, this "overlapping" APS fraction is removed from further consideration. Thus, the merged SMPS-APS distribution (for a given effective density) includes the fixed SMPS spectra and "non-overlapping" fraction of APS data. The merging criterion is a value of the effective density that provides closure for two optical properties ($\sigma_{s,obs}$ and β_{obs}). We emphasize that our approach and the conventional alignment method have very similar shifting procedure of APS spectra, but distinct criteria for obtaining the combined SMPS-APS size distribution: the merging criterion based on optical closure (our approach) and alignment criterion based on size distribution fitting in the overlap size range (alignment method)."

(3) In Sect. 4.3, the authors indicated that by visual inspection, there are no major alignment problems in the overlap region. Would it be possible to statistically compare the retrieved densities and the densities calculated by the alignment method as in Hand and Kreidenweis (2002) and Khlystov et al. (2004)?

This is great suggestion. However, there is no overlap size range for the SMPS and APS data considered here. Therefore, the conventional alignment method, which requires a sufficiently large overlapping size range, is not directly applicable to these SMPS-APS data. We are hopeful that future applications of the retrieval described in this paper, with the appropriate size configurations, will allow the suggested statistical comparison.

RH adjustment and uncertainties. The authors estimated the uncertainties of the method with an ideal case. How about the uncertainties introduced by the RH adjustment into the real case? Since the major chemical composition were measured during the campaign, would it be possible to justify the empirical parameterizations used in the RH adjustment?

We agree with the reviewer that the RH corrections are based on parameterizations with well-known limitations. We also agree with reviewer that including the chemical composition data for improved RH corrections would be beneficial. However, currently available chemical composition measurements (ACSM and SP2 data) are insufficient for evaluating the aerosol hygroscopic parameters. For example, these two instruments (ACSM and SP2) are not sensitive to marine salt, which plays significant role in coastal environment (e.g., Titos et al., 2014). Since direct measurements of the hygroscopic growth parameter or/and scattering enhancement are not available, we are basically forced to rely on appropriate parameterizations for the RH corrections.