Review of 'Using Spectral Methods to Obtain Particle Size Information from Optical Data: Applications to Measurements from CARES 2010', by Atkinson et al., 2017.

General comments

The manuscript presented by Atkinson et al. describes the retrieval of particle size related information from multi-wavelength aerosol extinction, scattering and absorption filed measurements using spectral deconvolution method that is typically used in remote sensing applications. The authors aim to compare the retrieved values with values that are calculated directly from size distribution measurements in order to validate the retrieval approach and to discuss its limitations. This work contains substantial contribution to further verification of remote sensing measurements using in-situ instruments. I recommend publication after the following comments have been addressed. Most importantly, as the main goal of this work is to evaluate the spectral deconvolution algorithm by comparison to size distribution measurements an additional effort should be made by the authors to describe and present the error propagation or uncertainty calculation inherent to each calculation from the uncertainties in each measured parameter.

Specific and technical comments

1) Line 232: "…averaged AERONET-SDA differences of 10% +- 30% for large FMF values > 0.5". It is not clear if the authors mean a difference of -20% to +40% or from 0% to +40%?
2) Line 254: since measurement of aerosols light extinction are by definition only apply to the forward direction it is unclear what the authors mean by truncation errors in CRD?
3) Line 313: data in table 1 regarding the PSU-CRD does not correspond to the text.
4) Line 323: a slope of 0.87 in the correlation between two CRD instruments at the same wavelength is significant. What is the uncertainty on this value? How was this 13% error mitigated in the data analysis? Was any correction applied? And how sure are the authors that the same "error" would apply to the 1064nm or the 405 nm CRD's? The authors are sure that with this 13% difference between the instruments "the two instruments were measuring the same aerosol with comparable measurement quality". I do not agree with this statement.
5) Line 343-350: SMPS scans typically take several minutes. A car passing by or a wind gust will cause significant changes to the aerosols population in time scales of seconds. This can be verified by looking at total aerosols concentration data taken with a CPC with a 1 sec resolution. To overcome mid-scan dramatic changes some dead volume is typically applied to allow for mixing of the aerosols and to smooth rapid changes. What measures were taken to insure that each individual SMPS scan is not interrupted by such events?

6) All figures should have some indication of the uncertainty of the presented values in order for the reader to appreciate the variation in the data within/between data series and temporal variation such as the diurnal cycle.
7) Lines 398-399: the authors claim that the difference between FMF_{CRD} and FMF_{sum} is due to significant contribution by large particle. Wouldn't it be possible to fine some support for this claim in the SMPS data?
8) Lines 419-423: I am afraid I don't understand how the differences between the two sites (and not the difference between CRD and SUM in site T0) "highlights the fact that there is not a precise definition of "fine" and "coarse" in terms of a specific size cut in the optical method." Additionally it is not clear what do the aouthors mean by the shape of the size distribution. Is it the width and/or amplitude ?
9) In figure 4 errors are needed to establish if the temporal variability is real or within uncertainty. This is important for conclusions presented in lines 461-463 and 477-478.
10) Figure 5: why is the discrepancy mostly clear in the first half of the day then the second half of the day in site T0?