### Item-by-item response to Reviewer #2

The authors gratefully acknowledge the anonymous reviewer for carefully reading the manuscript and providing constructive comments. This document contains the authors' responses to comments from reviewer #2. Each comment is discussed separately with the following typesetting

### \*Reviewer's comment

Author's response

\*The manuscript of Pérez-Ramírez et al. is devoted to the retrieval of the aerosol volume content and effective radius using the data on spectral aerosol optical depth, obtained with the use of sun and star photometers. Authors applied a modification of the least square method, which they called Liner Estimation (LE), method to the solution of the Fredholm's equation. Inversion of only the direct solar radiation measurementsallows assessment of the aerosol parameters with significantly better temporal resolution compared to standard AERONET procedure. In order to minimize biases between standard AERONET algorithm retrievals and LE, correction functions are proposed using the multi-year database of observations. The approach developed was applied to AERONET and starphotometry measurements at the city of Granada (Spain) and day-to-night time-evolution of columnar aerosol microphysical properties have been analyzed. The paper can be published in AMT once the general and specific comments have been addressed

### \* General comments

\*1. Please specify, what are the advantages of your method over a number of known.

The advantage of the method proposed is that it requires only AODs as inputs while the operational AERONET algorithm needs both sun and sky radiance values. That makes larger number of retrievals possible during the day.

However, current AERONET algorithm is more consistent as sun and sky radiance measurements allows the separation between absorption and scattering and thus allows the retrieval of particle refractive index and phase function. That is not possible with methodology proposed in this manuscript. Therefore, our approach cannot replace the operational AERONET algorithm. Indeed, we present the retrieval of Linear Estimation as a complementary tool to achieve higher temporal resolution retrievals.

Also, the approach presented allows the retrievals of aerosol microphysics for many instruments that only make direct sun irradiance measurements (e.g. MICROTOPS and PFR sun radiometers). This is of particular interest for nighttime measurements when sky radiances are not possible due to weakness of the sources, either the moon or stars.

Please note also that the inversion of direct sun measurements can be performed using other algorithms, for example inversion with regularization. In terms of accuracy the LE and regularization approaches are similar; however LE is faster and more stable to errors in the input data.

\*2. Throughout the paper the quantities V and  $r_{eff}$  are used, not specifying the size interval to which they refer. But inversion is carried at different values of  $r_{min}$  and  $r_{max}$  (p. 2339). This obviously has an effect on the values of the parameters retrieved.

The referee is correct mentioning that the retrievals depend on  $r_{min}$  and  $r_{max}$ . The inversion can be run in the interval 0.05-10  $\mu$ m. But constraining the inversion to the appropriate range  $r_{min}$ ,  $r_{max}$  reduces the discrepancy and provides more accurate retrievals. This point is clarified in section 2 of the manuscript, but we will pay attention in the revised manuscript to make this point more clear.

\*3. Authors have limited analysis to only two microstructure parameters – volume content and effective radius. They are calculated from the retrieved size distributions. But no comparison of the LE size distributions with initial models and AERONET retrievals has been performed.

The Linear Estimation approach does not provide reliable particle size distribution as an output, so only bulk particle properties were considered.

\*4. Spectral range of sun and star photometers in the paper is rather narrow – 380 - 1020 nm. The ratio of the maximal wavelength to minimal one is less than 3. How can you retrieve aerosol size distributions in the radius interval from 0.05 to 10 micrometers with ratio of maximal to minimal radius equal to 200? Do authors really declare the possibility to retrieve volume of content of the coarse aerosol fraction from extinction measurements in the spectral region 380 - 1020 nm? If so, it should be proved.

We agree with the referee that the range interval is quite narrow and limits the contribution of very small and very big particles. However we would like to mention that there is no direct relationship between ratios of maximal/minimal values for wavelengths and radii. In our approach we estimate particle volume and surface from a linear combination of the measurement kernels, so the main contribution to the particle bulk properties is provided by particles in that size range, where these kernels are most efficient. But despite these ambiguities, simulation (and comparison with AERONET) demonstrates that 40% uncertainty of particle size and volume estimation is achievable.

\*5. The statement that retrievals of columnar volume aerosol content by inverting AODs do not depend on particle refractive index is wrong. Scattering efficiency factors for particles of fine fraction in visible region is approximately proportional to n-1. So, retrieval with overestimated refractive index leads to underestimation of the volume content and effective radius, and vice versa.

Reviewer is right as scattering efficiency depends on refractive index. What we wanted to mention in that sentence was this dependence is weaker than for particle backscattering, which is used in the retrievals by multi-wavelength lidar measurements. As mentioned before, the retrievals of refractive index by our approach possesses quite large uncertainties while effective radius and particle volume content are obtained with uncertainty below 40%.

However we insist on the point that we consider a search space for refractive index. Simulations were performed for the search space while fixing the refractive index at specific values, both Scenario I and II used in Section 3. The results of these simulations showed the patterns mentioned by the referee. But the most important result was that all these differences were within the 40% uncertainties claimed. This uncertainty threshold is supported by the comparisons done with correlative retrievals by the operational AERONET algorithm. We will introduce corresponding revisions to the manuscript to clarify this point.

\*6. Authors analyzed parameters of correction as function of the relative input of the fine fraction into AOD at 500 nm. But this parameter is not measured directly instead of Angstrom exponent. Possibly it might be better to connect correction immediately with Angstrom exponent.

We agree that the fine mode fraction is not measured but is computed for the same set of spectral optical depths used for the retrieval. As the Angstrom exponent depends on the range of wavelengths used for its computation, we preferred to use the fine mode fraction to compute the correction functions. Nevertheless, the relation presented in Fig. 1 allows a direct conversion of the correction functions using the Angstrom exponent computed between 440 and 870 nm. This will be clarified in the revised manuscript.

#### Specific comments

## \*P. 2335, line 16 - What do you mean under "measurement kernels"? Are they the measured parameters simply?

By "measurement kernels" we refer to the kernel functions related to the extinction at 380, 440, 500, 670, 870 and 1020 nm that are the wavelengths used for AODs. The expression "measurement kernels" might induce confusions and will be removed in the revised manuscript. The matrix 'K' of equations 2 and 3 will be referred just as the kernel functions.

# \*P. 2337, line 9 -What values of real and imaginary parts of refractive index have you used in inversion? How changes in the refractive index affected the retrieval of fine and coarse fraction?

The retrieval of fine and coarse mode fractions are done through the Spectral Deconvolution Algorithm (O'Neill et al., 2003) which makes use of the spectral curvature of the Angstrom Exponent. Thus, there are no dependencies with refractive index. For more information, please consult the bibliography (O'Neill et al., 2001, 2003).

\*P. 2340, lines 17-19 - Why you do not utilize AOD at 340 nm? -What do you mean under effective radius? Does its value depend on the size interval used in inversion?

In the ultraviolet region the spectral dependence of absorption can be quite important, and as the filter at 340 is placed at this region, its inclusion can induce noise in the inversion and the dependence of the retrieval with refractive index would be larger. Inversions at this wavelength are not made by the operational AERONET algorithm either. Therefore, we think that the range 380-1020 nm is appropriate.

## \*P. 2341. -What values of the refractive index were used in numerical simulations?

We thank the referee for pointing that out. We forgot to mention in the original manuscript that both for Scenario I and II, the size distribution used as inputs have refractive index of 1.40 + 0.001i.

Later, from the synthetic data, the inversion by LE is run using the ranges mentioned in the manuscript, varying the real part of the refractive index from 1.35 to 1.65 with a stepsize of 0.025, while the imaginary from 0 to 0.015 with step size of 0.005. These points will be clarified in the revised manuscript.

### \*P. 2341, line 4 - Correct Nf/Nc on Nc/Nf.

Corrected.

## \*P. 2342, line 26 -How can you compare effective radius given by AERONET (radius range 0.05 -15micrometers) with yours retrievals?

The objective of such comparisons is to evaluate the effective radius retrieved by LE versus other more robust retrievals that make use of sun and sky radiances. That is why we believe that the comparisons with operational AERONET are appropriate.

\*P. 2346, lines 3 – 6 - The retrievals of columnar volume aerosol content by inverting AODs have more limitations as they DO DEPEND on particle refractive index. -Lack of information about coarse particle contribution is caused by another reason.

We agree with the referee comment. That sentence will be replaced in the revised manuscript by something like "... We note that the retrievals of columnar volume aerosol content by inverting AODs possess larger uncertainties due to the more limited information ....."

### **BIBLIOGRAPHY**

O'Neill, N.T., Dubovik, O., and Eck, T.F.: Modified Ångström exponent for the characterization of submicrometer aerosols, Applied Optics, 40, 2368-2375, 2001a.

O'Neill, N.T., Eck, T.F., Holben, B., Smirnov, A., and Dubovik, O.: Bimodal size distribution influences on the variation of Angstrom derivatives in spectral and optical depth space, Journal of Geophysical Research, 106, 9787-9806, 2001b.

O'Neill, N.T., Eck, T.F., Smirnov, A., Holben, B., and Thulasiraman, S.: Spectral discrimination of coarse and fine mode optical depth, Journal of Geophysical Research, 108, 4559, 2003.

Veselovskii, I., Kolgotin, A., Griaznov, V., Müller, D., Wandinger, U., and Whiteman, D.N.: Inversion with regularization for the retrieval of tropospheric aerosol parameters from multiwavelength lidar sounding, Applied Optics, 41, 3685-3699, 2002.

Veselovskii, I., Dubovik, O., Kolgotin, A., Korenskiy, M., Whiteman, D.N., Allakhverdiev, K., and Huseyinoglu, F.: Linear estimation of particle bulk parameters from multi-wavelength lidar measurements, Atmospheric Measurement Techniques, *5*, 1135-1145, 2012.