Interactive comment on "Characterisation of aerosol size properties from measurements of spectral optical depth: a global validation of the GRASP-AOD code using long-term AERONET data" by Benjamin Torres and David Fuertes

Anonymous Referee #3

Received and published: 16 December 2020

General comments:

The aim of this paper is to show that the GRASP-AOD code has the potential to be used for large scale datasets either for aerosol climate studies or for near real time modeler needs. The validation based on 2.8 million GRASP-AOD retrievals using AERONET AOD observations from 30 sites during 20 years makes the work robust enough to reach appropriate conclusions. The paper is to long taking into account the methodology used, the results and the prior knowledge published about this type of AOD inversion codes. I suggest making a synthesis relying on the bibliography already published, including the new considerations used that can improve this type of AOD inversion codes (comparative and differences with other papers already published). The paper is well written and into the scope of AMT. I recommend the publication of this paper, but there are some issues should be addressed prior to publication. The Editor will judge.

We would like to thank the anonymous referee 3 for reviewing the manuscripts and the positive comments.

The AOD inversion codes have used in different papers from many years. These type of inversion codes are based on the aerosol scattering equation that express the dependence of the spectral variation of AOD on the aerosol size distribution, and also depend of the Q_{ext} parameters (particle extinction efficiency factors), which in turn depend on the wavelength, the refractive index and particle radius. As example, King et al. (1978) already pointed out that the definition of the particle radius interval on which the inversion method can be correctly used, and the assumption of realistic refractive index values are the most crucial points in any rigorous application of inversion methods applied to spectral series of the AOD. On the other hand, the independent information content on the optical characteristics of columnar aerosols is contained primarily in the particle radius interval from 0.1 to 2 microns, approximately, for AOD measurements covering spectral range 340-1020 nm. On the other hand, the iterative procedures modified the radius interval within the prescribed ranges, and the best results were obtained for reduced radius range. In this sense, with this type of codes the results are limited to the accumulation mode. On the other hand, some AOD inversion algorithms use a single refractive index, while the true is dependent on wavelength. The assumption of an a priori defined refractive index in the AOD inversion procedures may lead to very different derive size distributions, but other authors (e.g., Yamamoto and Tanaka, 1969; King et al., 1978; González and Ogren, 1996) show that the shape of the retrieved aerosol size distribution is not substantially altered as a result of using such assumptions. In this sense, this paper should take into account previous work and show the improvements that can be made. Taking into account previous results, obviously these type of inversion algorithms would not work well for coarse particle modes just considering only the AOD spectral values. Spectral aureole data (sky radiances) are required to achieve good results in coarse mode.

We recognize the knowledge of the referee regarding aerosol property retrievals. We have added some of the comments from this paragraph along the document (especially in the introduction and new section 4.3). We reckon that this update has enriched the article. However, we would like to add some points in the discussion:

- The article of King et al. 1978 uses the interval 0.1-4.0 μm when inverting AOD measurements from 0.440-1.030 μm. Moreover, the authors add the following comment while setting their election: "Although this matter (referring to the election of the interval) has been considered by Yamamoto and Tanaka (1969) for both Junge -and Woodcock-type aerosol size distributions, it is very important to realize that there is no absolute rule which determines the radii limits having the most significant contribution to the attenuation measurements… Since the size distribution function is not known in advance, it is apparent that occasional trial and error is required in order to determine the radius range over which the inversion can be performed".
- In the work by Gonzalez and Ogren (1996), the interval is limited between 0.1-2.0 μm, maybe since the spectral range considered is slightly smaller: 0.35-0.88 μm. Note here that the claimed low sensitivity to radii variation (or we shall say the ratio between the radius and the wavelength known as size parameter) does not mean that the contribution of coarse particles to estimate the total extinction can be neglected to characterize the aerosol optical depth. Actually, the fact of reducing the radius interval to 0.1-2.0 μm at González and Ogren (1996) originated an irreal-excess of particles at smaller radii that tried to optically compensate that large particles were dismissed (see tests done with synthetic measurements, examples in fig.3 or fig.4). This "fake" effect adds more uncertainties in their size distribution characterization (moments, effective radius, etc.). We certainly admit the low sensitivity to retrieve coarse mode size parameters, but the effect of ignoring its contribution in the retrieval would create errors in an overall characterization of size properties.

- We agree that a basic analysis about the variation of Q_{ext} functions would show that the coarse mode radii are very close to the geometrical-optic region (accounting the spectral range used in the study), and therefore, the Q_{ext} values slightly vary from the asymptotic value of 2. The sensitivity of the AOD measurements to those radii is very small. This fact is not hidden along the work and it affects the characterization of the coarse mode as shown in the paper and as largely commented in Torres et al. 2017. But this does not mean that the optical extinction due to these particles is zero (see for instance the Modified Kernel Functions for Optical Thickness represented as function of the radius in figure 5.1b of King and Dubovik 2013). The fact of neglecting its contribution creates undesirable effects as the ones found in the work by González and Ogren (1996).
- The inversion strategy proposed here, which the solution is predefined by two log-normal functions, presents some advantages with respect to previous strategies (which resided in the multiplication of a rapid varying function - typically Junge - and another of slower variation - which is the one retrieved at each predefined interval -). These advantages are presented along the manuscript and they cannot be just summarized by a compilation of previous results. The most important are recapped here:
    1. It allows to separate the optical contribution of the modes. As the SDA, GRASP-AOD code separates fine mode optical depth (highly dependent on the wavelength) from the coarse mode contribution (almost spectrally independent in the range 340-1020 nm). We have largely proven the robustness of this retrieval through comparisons with AERONET retrievals.
    2. It allows to accurately characterize the fine mode radius under certain conditions ($\tau(440)>0.2$ and $\alpha>1.2$). The RMSE compared to AERONET retrieval (=0.023μm) is quite good considering the information contained. This detailed characterization represents an important novelty compared to the forementioned codes, or some others used for only τ measurements, such as the ones inspired by LET techniques.
    3. The coarse mode contribution is represented by only three parameters (two in fact since the standard deviation is quite constrained) and well characterized in terms of mode optical depth. We are aware that different pairs of R_{Vc} and C_{Vc} produce similar spectral coarse AOD values (larger concentrations compensates an increase of the mode radius), but coarse mode contribution is well accounted by GRASP-AOD. Note that most of the values of R_{Vc} that are retrieved in the paper (AERONET

retrievals) are under the limits established by King et al. 1978 (<4.0 μm), the values of the volume distribution beyond this interval are forced/fixed by the log-normal function.

- The effect on the refractive index (due to anomalous diffraction theory of Van de Hulst (Van de Hulst, 1957) as primarily discussed in Yamamoto and Tanaka (1969)) would be commented later. At this point, we would like only to recall that it was already presented in Torres et al. 2017 with some ideas proposed by M. King who was one of the referees of that study.

Lines 85-95. To motive the importance of this work, the authors comments that many AERONET sites are plagued by several months of partial cloudiness (no sky radiance measurements) . . . but later they use climatological values for refractive index and information about radius modes. How it is possible for this type of AERONET stations, and how representative are these values? also for future applications to night measurements. The columnar aerosol properties change from day to night, depend on sources, the air masses transport, the planetary boundary layer high ... Also, a study of the GRASP-AOD sensitivity to the refractive index is needed.

The representativity of the chosen climatological values (based on retrievals with clear sky conditions) would depend on the site. The averaged value strategy presented here should be considered as a first reasonable approach. Though the dependence on refractive index (mostly real as indicated by King et al. 1978 and Torres et al. 2017) of GRASP-AOD application was deeply discussed at Torres et al. 2017, we have added a discussion point (new section 4.2) where we have treated Mongu site (with climatological values around 1.51-0.021i) with a generic refractive index of 1.45-0.005i.

- Line 185. The GRASP-AOD code assumes the refractive index as known. Which one has been chosen for each AERONET station and aerosol type? Can be Included in Table-1? On the other hand, the aerosol type selected for each station (Table 1) can be the more frequent (climatology), but not all ways are the same. As example, the Saharan dust outbreaks. How these facts affect the inversion products?

We have used moving monthly means (2 adjacent months) for all sites using Version3 AERONET aerosol retrieval. We cannot include a table containing all the values since one site would contain already 8x12 values. We present here as an example (Table A), the values found/used for GSFC.

The issue commented in the last lines was more or less discussed at pag. 24 lines 450-455 (at the end of Section 3.2.1 - discussion of R_{Vf} characterization - which has been kept). We have suggested that future reprocessings may use more developed climatologies (e.g. considering different values for different Ångström exponents) which may improve some of the results obtained in this study.

	Real Refractive index				*Imaginary Refractive index*			
	440	*670*	*870*	*1020*	*440*	*670*	*870*	*1020*
January	1.443	1.437	1.435	1.429	0.004	0.004	0.005	0.005
February	1.462	1.447	1.444	1.434	0.005	0.005	0.005	0.005
March	1.473	1.463	1.461	1.454	0.005	0.005	0.006	0.006
April	1.467	1.455	1.453	1.447	0.005	0.005	0.006	0.006
May	1.468	1.456	1.453	1.449	0.005	0.005	0.006	0.006
June	1.458	1.441	1.438	1.433	0.005	0.005	0.006	0.006
July	1.456	1.440	1.436	1.431	0.005	0.006	0.006	0.006
August	1.449	1.435	1.431	1.425	0.005	0.005	0.006	0.006
September	1.441	1.427	1.424	1.420	0.004	0.005	0.005	0.005
October	1.432	1.424	1.422	1.419	0.004	0.004	0.004	0.004
November	1.426	1.420	1.419	1.418	0.004	0.004	0.004	0.004
December	1.425	1.428	1.427	1.425	0.003	0.004	0.004	0.004

Table A Example of the refractive index values used to run GRASP-AOD. - GSFC site -

Lines 190-195. If the refractive indices are assumed, what happens, as example, with stations where there are many clouds and cannot be computed with the sky radiance data? There are no data? Do you use the climatological value? How much data have you used to obtain this climatological value, and how is it distributed throughout the year? In order to these results will be realistic, an extensive database should be available and the appropriate refractive index value used for each atmospheric condition. The purpose of this work is to show that the GRASP-AOD application has the potential to be used for large scale datasets.

If not data at all is available, standard refractive indices should be considered. From the moment, that there will be some full AERONET inversions the existing archive of the hypothetical new site could be reprocessed. Further new reprocessing could be done as the climatological database is updated. To run a site with 20 years of data as the examples presented here takes around 6 hours with current processors (no much time needed).

As commented before, we have added a new section 4.2 discussing what happen if climatological refractive indices are not available by reprocessing one site with standard refractive indices.

Lines 480-525. Obviously, the algorithm does not work well for coarse particle mode just taking into account only the AOD spectral parameters and a climatological value of the refractive index. But we already knew these results from the papers published related with these type of inversion codes. The sky radiance data is needed to achieve good results in coarse mode. I think this section should be shortened or removed from the paper. Also, the last sentence of the abstract is a well-known result and it is not new.

This point was partially discussed before. Nevertheless, we understand the comment of the referee and we have actually considered to erase the subsection. After a discussion with the editor, we have decided to keep it mainly for two reasons: a) Even though the results are similar to previous analysis, the strategy proposed by GRASP-AOD presents itself some novelties that are worth to comment. b) In future works, we will explore in detail the GRASP-AUR application which has the same strategy as GRASP-AOD to represent the size distribution. Certainly, the results obtained by GRASP-AOD in the coarse mode will be taken as a reference in the new characterizations as partly done in new section 4.3 (old 4.2)

Lines 200-225. The criteria are based mostly on analyst's experience. The authors show "Due to the low sensitivity of GRASP-AOD to the shape of the modes. . . we have used strong a priori constraints on the actual values for the standard deviation of both modes. . . in practice, their values are very similar to the given initial guess values". On the other hand, in Line 340 the authors show: "The larger uncertainties observed for Solar Village compared to GSFC can be extrapolated to all sites with coarse mode predominance with respect to the sites with fine mode predominance", and the following lines. Taking into account the papers published so far, it is clear that this methodology can only be applied to places where the fine mode predominates. In my opinion, this work should be drastically reduced, showing only those aspects that can improve the results of the works already published. On the other hand, the usefulness of using climatological values in the a priori assumptions should be better discussed.

The low sensitivity to standard deviation (even to fine mode) was discussed in Torres et al. 2017. We believe that the strategy of bimodal lognormal functions for only AOD measurements is a novelty of the work by Torres et al. 2017. In this sense, the low sensitivity to the standard deviation cannot be summarized from previous works.

Regarding the comments in Line 340 it refers explicitly to the separation of optical depth fine/coarse mode ($\tau_f(500)$). Different works about SDA algorithm (O'Neill et al. 2003 or Eck et al. 2010) obtained similar results as acknowledged along the article.