Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2016-334-AC5, 2017

Atmospheric Measurement Techniques Discussions
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Interactive comment on "Advanced characterization of aerosol properties from measurements of spectral optical depth using the GRASP algorithm" *by* B. Torres et al.

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Received and published: 19 June 2017

Interactive comment on "Advanced characterization of aerosol properties from measurements of spectral optical depth using the GRASP algorithm" by B. Torres et al. T. F. Eck (Referee) thomas.f.eck@nasa.govâĂÍReceived and published: 25 January 2017 Review for Atmospheric Measurement Techniques Title: Advanced characterization of aerosol properties from measurements of spectral optical depth using the GRASP algorithm. Authors: Benjamin Torres, Oleg Dubovik, David Fuertes, Gregory Schuster, Victoria Eugenia Cachorro, Tatsiana Lapyonok, Philippe Goloub, Luc Blarel, Africa Barreto, Marc Mallet, Carlos Toledano, and Didier Tanré

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Discussion paper

General Comments: This paper describes a new application of the GRASP algorithm that utilizes accurate spectral AOD measurements from sunphotometer instruments (both ground-based and airborne) to retrieve aerosol size distributions. The paper is clearly organized and very well written. This new algorithm provides inversions that may be extremely useful in analysis of particle size related aerosol parameters, since these retrievals can be made much more frequently from AOD spectra alone than from the combined input of AOD spectra and sky radiance distributions, such as in almucantar inversions in AERONET (the Dubovik and King (2000) algorithm). The sensitivity analysis is clear and thorough, however I am surprised that the perturbations in real refractive index applied in this section are so small. Real refractive index variations of +-0.02 (for fine mode dominated cases; section 3.5.1) are somewhat trivial and it would seem very difficult to provide with such accuracy when day-to-day (and seasonal) variation in real refractive index can be expected at some sites (such as at GSFC). This is due to hygroscopic growth of particles related to variation in relative humidity and low altitude cloud interaction with aerosol, plus relative changes in aerosol species composition variation. Ideally, the analysis should include much larger uncertainty/variation in the initial guess of the real refractive index.

We have carried out a series of test with real refractive index variations up to ± 0.05 for the example of GSFC. We have observed a linear behaviour between the real refractive index variation and the error produced in the retrieved parameters that characterize the fine mode. Thus, it is possible to approximate the error in r_{V_f} and C_{V_f} as: $\Delta r_{V_f} = -0.04\ \Delta n$ and $\Delta C_{V_f} = -0.27\ \Delta n\ \tau_a(440)$. In relative terms, these differences in r_{V_f} represents between $\mp 12-13\%$ while in C_{V_f} between $\mp 8-10\%$ for the maximum variation considered of $\pm 5\sigma_n$.

I agree with the editor and other reviewers that you should add the case of AOD(440 nm)=0.1 to your sensitivity analysis section, since most data in the AERONET network are for AOD levels <0.3. Especially for the Lanai site the AOD=0.3 is much too high to be representative. This is a background marine site where AOD is predominately

<0.15. The monthly mean AODs at Lanai do not exceed 0.08 at 440 nm except for the dust transport season in spring when they reach a monthly average maximum of 0.12 in April. I also agree with the editor that it would be useful to include very high AOD cases in your sensitivity analysis (1.5 or 2 at 440 nm) for dust and also fine mode smoke, since these cases are important for analysis of major aerosol events.

The estimation of uncertainty in AERONET measured AOD for field instruments of 0.01 in the visible and NIR and increasing to 0.02 in the UV wavelengths should be mentioned on Page 13 or elsewhere in the manuscript. This wavelength dependent uncertainty was taken into account for operational SDA inversions where only the 380 to 870 nm wavelengths (5 channels) were utilized as input to the algorithm in order to avoid somewhat higher uncertainties in the 340 nm AOD due to both calibration and filter instability (degradation in time), and for 1020 nm due to temperature sensitivity of the detector at this wavelength and water vapor absorption. It seems that you should make some tests of wavelength range used as input to the code (with accounting for realistic uncertainty for each wavelength) to determine the optimal set of AOD spectral data that should be used as input, plus characterize how the uncertainty changes when specific wavelengths are not available. In particular some AERONET Cimels operated by PHOTONS only have the 440, 675, 870 and 1020 nm channels for AOD measurement. How is the algorithm performance affected when only these 4 wavelengths are available as input to the GRASP-AOD algorithm?

The multiterm LSM formulation allows to account for the uncertainty of each wavelength. Therefore, in real applications of the code, there may be two alternatives: to introduce the known uncertainties of each wavelength in the covariance matrix (Eq 4 and 5) or to reduce the spectral range. Following your advices, we have repeated the tests in section 3.4 (Simulation of aerosol optical depth errors) assuming double uncertainty for the wavelengths 340, 380 and 1020 nm. At the same time, we have re-done the study for the spectral range of the old polarized photometers 440 to 1020 nm (new section 3.4.3).

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For the Real Cases (Section 4) analysis it is important again that you include cases with AOD of 0.1 at 440 nm for all sites, and for the Lanai site a case of AOD(440)=0.07 since this is the climatological mean AOD (for non-dust months, i.e. excluding spring season) for that site. Additionally you should include some analysis of sites that are not included in the Dubovik et al. (2002) Table 1. The vast majority of AERONET sites are not characterized in that table so if the algorithm is to be operationally applied a strategy for selection of first guesses of the input parameters is needed.

The new sites considered in the Section 4, Lampedusa and Rome Tor Vergata, include study cases with $\tau(440) < 0.2$. Thus, four of the eight analysed sites include one case with $\tau(440) < 0.2$ and all the sites has at least one case with $\tau(440) \leq 0.3$. In the simulation study for low aerosol conditions, we point out that the uncertainty for the parameters representing the bimodal log-normal size distribution increase a lot when the aerosol load is low. For practical uses of GRASP-AOD, it would be convenient to establish a lower limit in the τ_a values for quality assured retrievals. On the other hand, the capacity to discriminate between fine and coarse mode extinction remains stable, in absolute terms, and it is related to the value of τ_a-errors.

The way the paper is written it sometimes implies using the almucantar retrievals as a source of first guesses for refractive indices and size, therefore it is presented (in some sections) as a companion retrieval to the almucantar sky radiances retrievals and not a fully independent retrieval such as SDA. This should be discussed or clarified in the paper.

To run the GRASP-AOD inversions, we have assumed climatological values of the refractive index for the different sites in Table 13. This is now clearly stated in the text. In any case, in real applications of the code, we would suggest exploring other alternatives (to use the data from the closest AERONET standard inversion (same day), monthly climatologies, etc.)

Specific Comments: Title: You say ". . .characterization of aerosol properties. . ." but

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shouldn't this be more specific, since only aerosol size related properties are retrieved as the refractive indices are assumed a priori. Therefore it seems that perhaps the word 'size' should be inserted between 'aerosol' and 'properties' in the title.

Changed, thanks.

Abstract lines 10-11: "Differences in the fine mode volume median radii for the GRASP-AOD and AERONET inversions are less than 0.02 μm at sites dominated by the fine mode for all cases. . ." Is this the maximum difference or mean difference?

We have redone the sentence: Differences in the fine mode volume median radii for the GRASP-AOD and AERONET inversions are in average 0.010 μm at sites dominated by the fine mode and 0.013 μm including all cases.

Abstract line 16: I suggest removing the word 'advance' in this sentence since it is confusing.

Removed, thanks.

Introduction, Page 2, line 4: I suggest that you remove 'Unfortunately' from this sentence, as it is somewhat confusing.

Removed, thanks.

Page 3, lines 4-8: Please provide more details and justification for utilizing bi-modal assumption with rv and sigmas for each mode to describe the size distributions rather than use 22 size bins as is done in the AERONET standard retrieval. I assume that it is a more stable retrieval than the 22 bins, given only the AOD spectra as input, but it would be informative to provide some more details on why you selected this way to make the retrievals (either here or in Section 3).

Now it is shown in Section 3. Initially we have tried with 22-bis size distribution but we rapidly observed that strong smoothness constraints, in terms of size distribution, were required to assure realistic retrievals. That is why we changed our strategy to a log-normal approximation.

Page 3, lines 11-13: “For instance, AERONET includes around forty direct Sun measurements per day in its standardized sequence of measurements (in cloudless conditions); only about eight of these sequences are coincident with sky-radiance measurements that are suitable as input to the AERONET inversion code.” It would be useful to include information on the new Cimels here, where Hybrid scans make sky radiance retrievals possible each hour throughout the day thereby doubling or more the total number of sky-radiance retrievals. Also many new Cimels are run in the turbo mode for AOD measurement where the time interval of AOD sampling is 3 minutes rather than the 15 minutes for the standard scenario in a Cimel sun-sky radiometer. Therefore the âĹij40 measurements per day increases to about 200 AOD spectra per day (depending on latitude and season, therefore day length) with new Cimels.

We have added a sentence including the possibilities of the new instruments.

Page 3, lines 13-14: “. . .a large amount of data containing only direct Sun measurements which are not used apart from the characterization of the aerosol load.” I disagree, since the Angstrom Exponent and it’s spectral variation are also useful parameters providing basic aerosol size information that are computed by AERONET from the spectral AOD, and are utilized in many studies.

Yes, we agree. We have added: and to obtain some basic aerosol size information computed from its spectral variation

Page 3, lines 14-15: “Moreover, many AERONET sites are plagued by several months of partial cloudiness (especially in wintertime).” I think this is somewhat misleading, since some sites have the seasonal opposite and some tropical sites are cloudy for most seasons. Therefore I suggest dropping the phrase ‘(especially in wintertime)’ from the sentence.

Thanks, removed.

Page 3, lines 14-15: “As a result, the aerosol loads are the only data reported at many sites. . .” Please insert after ‘aerosol loads’ something like: ‘and Angstrom Exponents that parameterize the relative fine versus coarse mode optical influence, depending on the wavelength range used in the computation of AE (Eck et al. 1999; Schuster et al., 2006).’

Thanks, added.

Page 4, lines 11-12: Please include the reference of van Donkelaar et al. (2010).

Thanks, added.

Page 6, lines 4-6: For GRASP-AOD please discuss whether there would be any difference in the retrievals if spherical particle shape assumption was utilized/assumed versus spheroidal shape.

We have added in the inversion strategy description that we assume the sphericity parameter. At the same time we have included the subsection 3.6 Variations on the sphericity parameter, were the effects of this assumption are described.

Page 7, lines 7-9: In most cases this is true, that lognormal bi-modal size distribution assumptions are sufficient. However, for cloud-processed aerosol a third middle mode sometimes exists, see Eck et al. 2012 for Dubovik almucantar retrievals that are tri-modal, not bi-modal, and these middle modes are supported by independent in situ measurements of fog/cloud processed aerosols. This should be mentioned as a relatively rare case that does occur however.

We have added the following paragraph: However, some AERONET retrievals suggest that the particle size distribution is not always perfectly log-normal, as some size distributions are characterized by asymmetrical mode shapes (e.g. Dubovik et al., 2002a; Eck et al., 2005, 2010). Moreover, some size distribution retrievals have a pronounced tri-modal structure, such as observed in volcanic aerosol plumes (Eck et al., 2010) or aerosol retrievals located near fog or cloud (Eck et al., 2012; Li et al., 2014). Obviously, our strategy that is based upon simplified bimodal size distributions would not provide correct retrievals in these specialized situations.

Page 8, lines 12: Note that Dubovik 2002 presents an AERONET Version 1 database climatology that has been refined significantly with the AERONET Version 2 reprocessing of the entire database that occurred in 2006.

We have modified the paragraph as follows: The numerical tests in this paper are based upon the climatology provided by Dubovik et al. (2002a), which utilizes about 10 years of real aerosol retrievals with AERONET's Version 1 processing. (We note that AERONET has subsequently implemented a Version 2 aerosol retrieval product, but the single-scatter albedo climatology that is based upon this newer processing scheme is within 0.02 of the Dubovik et al. (2002a) climatology for the same aerosol type; Giles et al., 2012).

Page 8, lines 16-17: This is weakly absorbing aerosol at GSFC, non-absorbing aerosol would have SSA=1.

Changed, thank you.

Page 8, lines 17-18: It should be mentioned that this is strongly absorbing biomass burning (BB) aerosol at Mongu, Zambia and that BB aerosol range from very weakly absorbing to strongly absorbing (see Eck et al, 2003, GRL) depending on fuel types and phase of combustion (flaming versus smoldering).

Added, thank you.

Page 8, lines 27-28: Why would you assume that for a mixed case (Bahrain) that all particles are non-spherical? Solar Village also has many mixed mode aerosol days with predominately spherical fine mode particles from pollution.

We use a sphericity parameter of 0 (i.e., all particles are non-spherical) for all the Solar Village cases except the one with $\tau_a(440) = 0.3$ (SOLV1), which has an Ångström exponent greater than 0.6 and can not be considered as pure desert dust. We use a linear approximation with respect to the Ångström exponent to select intermediate values of the sphericity parameter for the three cases with Ångström exponents between 0.6 and 1.1 (SOLV1, BAHR2, and BAHR3). That is, we use sphericity parameters of 0 and 100 for Ångström exponents of 0.6 and 1.1 (respectively), and linearly interpolate the intermediate values. The rest of the examples have Ångström exponents greater than 1.1, so we fixed the sphericity parameter at 100 (considering all the particles as spheres).

Page 8, lines 12-13: Surprising that you do not show the case of AOD=0.9 in Figure 2 since this is the one where the curvature for the fine mode cases would become the most obvious, since the fine mode radius is largest. Note that the 1640 nm AOD showing a departure from a 2nd order fit of AOD versus WL in Figure 2 is not observed in the real GSFC site AERONET measurement data. Perhaps you can show the 2nd order fit to your simulated data in Figure 2 and also show the delta AOD departures from the fit.

The curvature for case with AOD=0.9 is just a little bit more obvious than for the other two cases and it does not include Lanai case. Therefore, we prefer to keep the selected figures in the work. Note also that the purpose of figure is just to offer a graphical idea of the tendencies from the AOD simulated values. A study of the 2nd order fit and the differences would be interesting but we are aware of the length of the article and this analysis is out of the scope of the work.

Page 9, lines 8-9: Note that the Angstrom Exponents you computed for the Lanai site are much too high, you have 1.22 in Table 2, while the measurement data yield averages of âĹij0.6 to 0.8 for Lanai data (see the AERONET climatology tables). This possibly suggests an issue with the Dubovik 2002 climatology parameters for this site. Also note that for the Solar Village site only the 0.6 and 0.9 AOD cases can be considered dust dominated, since the AOD=0.3 case is mixed with Angstrom Exponent=0.84 in Table 2.

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It is true that values of Angstrom Exponents for Lanai site are higher than the data averages in AERONET climatology table. This may be due to an overestimation of the concentration of fine mode in the analysis of Dubovik 2002. At the same point, we would like to note that values of 1.2 are registered and they are not rare at Lanai site (see figure 9 in Smirnov 2001 or figure 1 in Smirnov 2003). On the other hand, we totally agree with the comment regarding Solar Village. In the new version, when we introduce the sphericity parameter, we comment that SOLV1 cannot be studied as pure desert dust and should be considered as mixed desert dust.

Page 13, line 29-31: The estimation of uncertainty for AERONET measured AOD for field instruments is 0.01 in the visible and NIR and increasing to 0.02 in the UV wavelengths (Eck et al. 1999). This spectral variation in AOD uncertainty should be mentioned here or elsewhere in the paper.

We have included it and redone all the tests taking into the account this fact in the covariance matrix.

Page 14, line 8: This is an awkward way to say that uncertainties increase at lower AOD. For the reader it would be much clearer if you did not use abbreviations such as GSFC1 and GSFC3 in the text but instead referred to the AOD levels and associated uncertainties.

Thank you, done.

Page 19, line 5: Should insert the Dubovik et al. (2006) reference here since these significant algorithm advances were incorporated in the AERONET operational inversions in 2006.

Thank you, added.

Page 19, line 6: Please add the reference Smirnov et al. (2000) here for Level 2 data in Version 2.

Thank you, added.

Page 19, line 22-23: Should say that these sites did not have the 1640 nm channel of the newer Cimels, as many readers will not know what you mean by 'extended' photometer.

I have added a reference to table A1.

Page 19, line 31-33: I suggest that an additional, perhaps more robust way to compare the retrievals of the fine mode fraction (FMF) of optical depth is to provide a scatterplot of all individual (not just daily means) GRASP-AOD retrievals regardless of SZA (not just time matched to almucantars) for many sites (including sites not in the Dubovik 2002 table) compared to the AERONET almucantar retrievals of FMF. This should include all AOD levels measured at each site. Both can be plotted as a function of AE(440-870) on the x-axis. Regression fit and statistics can then also be shown. Other parameters such as volume median radius could also be compared as scatterplots using time-matched individual retrievals from many sites, with the AERONET standard sky-radiance inversion on the x-axis and the GRASP-AOD inversion on the y-axis.

We consider that the paper is already quite overloaded, and that the main point of section 4 is to do a first validation with the daily means of retrieved products. As a next step, we would like to do a large validation with AERONET data, with individual (or point by point) studies like the one proposed here. Ideally, we would like to have some members of AERONET staff involved in that future analysis.

Page 20, lines 10-11: Need to mention the likely reason for this large difference for the GSFC-A case. It is the lowest AOD of all sites with AOD(440)=0.166. This is important and needs to be quantified further with cases that have lower AOD values such as 0.10 or lower.

The new analysis in subsection "3.4.2. Low and high aerosol load conditions" does show the large uncertainties of the retrieved parameters when the aerosol load is low. Given the characteristics of actual sun-photometer measurements, and particularly AERONET $\tau - errors$, the quality of the retrieval of the size distribution parameters can only be assured from $\tau(440) > 0.2, 0.3$.

Page 20, line 13: This is an anomalously large rv fine for a low AOD case at GSFC. Note that the climatology of GSFC size distributions (as a function of AOD) in Eck et al. (2012; Figure 17 b) and in Dubovik et al. (2002) strongly suggest that this case is an anomaly, therefore a relatively poor choice for a case study. The actual values of the almucantar retrieved individual rv fine at GSFC for this date (Nov 22, 2009) range from 0.21 to 0.26 micron (still very high for this AOD level) so I don't know where you got the 0.27 micron number from.

The value of 0.27 micron was a typo error in the text but not in (-old) Table 11 where the value of rv was 0.23 (now in table 14), which agrees with the values pointed out by the referee. On the other hand, we could find a day where fine rv fits better with the AOD values following the climatological functions. However, we reckon that the case is of a great interest since it shows that rv value retrieved by GRASP-AOD is not far from the value obtained by AERONET standard inversion, even thought, it is very different from the "initial guess" used (around 0.138 applying equations in Table 4, which are based on climatology results from Dubovik 2002).

Page 21, lines 17-20: Should note here in the text that differences in the definition of modes, radius cutoff in AERONET standard retrievals versus tails of modes included in SDA explain a least a portion of these differences.

This was written as a footnote and we have kept it

Page 22, lines 9-10: Need to note that the moon measurements have a spectral range from 440 to 1640 nm, while earlier in the paper you made the sensitivity analysis for the 340 nm to 1640 nm spectral range (see Table 2).

Done.

Page 23, lines 20-21: Are you referring to pollution or smoke from biomass burning or a mixture of the two types of aerosols?

I’m referring to only pollution.

Page 24, lines 8-9: Please give the rv for both the fine and coarse modes for the AERONET standard almucantar inversion, for comparison purposes to the GRASP-AOD inverted size distributions.

Added, thanks.

Page 28, lines 2-3: Calibration uncertainty for AERONET master instruments is better than you stated here. The uncertainty in Vo due to calibration by Langley analysis at Mauna Loa is $\sim 0.25\%$ in the visible and NIR and 0.5% in the UV. The resulting total uncertainty in AOD (additionally including some other sources of uncertainty) is ~ 0.002 to 0.009 for Master Cimels (lower values in visible and NIR and higher in UV ; see Eck et al. 1999). This is for overhead sun (SZA=0 and optical airmass=1) and the uncertainty in measured AOD is less as solar zenith angle increases (optical airmass increases).

We have modified it, thank you.

Page 28, lines 8-9: Should cite Smirnov et al. (2000) here since the Version 2 Level 2 data are cloud screened and quality controlled as described in this reference.

Added, thank you.

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