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 #1

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General comments:

Overall, this is a well-written paper describing the large scale validation of the GRASP-AOD product. Considering this product is able to retrieve total column size distribution and some optical properties without the need of having sky radiance measurements, GRASP-AOD will provide valuable information for atmospheric research and will certainly be widely used. I consider that this manuscript fits perfectly into the scope of AMT. I recommend publishing the manuscript, but there are some minor/technical details that I would like to be addressed in this discussion process.

We would like to thank the anonymous referee 1 for reviewing the manuscript. We are glad for the overall positive assessment regarding the manuscript.

As general comment, this paper presents a very comprehensive and compelling study on the validation of the GRASP-AOD product. A similar study was already performed by Torres et al. (2017). However this new paper is approached as a large-scale validation using AERONET as the most widespread operational network for ground-based aerosol observation. The use of thirty sites and some million of observations world-wide provides robustness to this analysis. However, the results have been listed in this work as a pure sequence of 20 pages with numbers and some partial conclusions that are very difficult for a reader to follow. I therefore suggest that the authors make a synthesis effort so that the results are clearer for the reader.

Thank you for your comment. We agree that this study may provide more robustness to GRASP-AOD application beyond the results obtained by Torres et al (2017). The large number of parameters analyzed may have difficulted to follow the description of the comparisons presented. However, we think that no table has been "just listed" without its particular analysis (more or less detailed, depending on the relevance of its results) and conclusions. Nevertheless, we have tried to emphasize some of the results in the along the manuscript since we agree that some descriptions were a bit tedious.

Specific and technical comments:

Page 4, line 120: This is not the first time that GRASP has been mentioned in the text. Therefore, I recommend including the acronym once GRASP-AOD product is referred.

Thank you for your suggestion. Even though GRASP-AOD is forementioned previously in the manuscript, this is actually the first time that the whole GRASP project is mentioned. The logic of the introduction made us to mention the whole GRASP project at the end, even though GRASP-AOD is previously defined (with the reference of Torres et al. 2017).

Page 5, line 153: Please, correct the typo “teen”.

Thank you very much for your comment. We have corrected the mistake.

Page 6, footer line: Please correct the Cimel version. It is not CE-310 but CE-318.

Yes, you are right. Thank you again for the correction.

Page 8, first paragraph: In this part of the text the authors stated that the priority was the selection of sites with high aerosol loads. However, some lines below, they stated that the GRASP-AOD products do not depend on aerosol load. This sentence seems confusing for the reader. It is also confusing the fact that, if your aim is including sites with predominantly clean conditions, why selecting only two among some hundred stations? Please clarify.

Validation and climatology studies are normally carried out at key AERONET sites which are determined by its aerosol load and the availability of a long-term time-series data (see for instance Dubovik et al 2002 or Gilles et al. 2012). In this sense, our work has tried to be consistent with previous studies and 28 sites out of 30 accomplish these requirements. The inclusion of Lanai and St. Denis was done to present a couple of examples of marine aerosol sites. These sites (as all marine aerosol sites - Smirnov et al. 2002) do not fit into the general rule of high aerosol load. Please also note that these two specific sites were selected since they presented the longest time series data of all marine aerosol sites.

Page 11, line 255: Are you using the AERONET Version 2 instead of Version 3? Is this specific process you are talking about in this paragraph not provided in Version 3?

As stated in section 2.1, all the data used in the study have been taken from AERONET Version 3. We just wanted to indicate here that this specific routine was implemented in Version 2 and has been kept like this in Version 3. In AERONET Version 1, the mode separation was different (fix cutoff at 0.6 μm). Anyway, to make it clearer for the

reader we have added that this routine has been kept in Version 3 (our only data source).

Page 19, second paragraph: The reason for having higher on-average AERONET retrievals in comparison to SDA and GRASP-AOD is attributed by the authors to the radius cut-off used in AERONET to define the two modes. I suggest the authors to describe briefly the differences between the three compared techniques. This description would be more enlightening than attributing beforehand the problem to the AERONET's cut-off.

Thank you for your comment. The effect of the cutoff was previously discussed in O'Neill et al. 2003 though we have agreed to recall it here. Following the logic of the manuscript structure, we think that the explanation about the separation of fine/coarse mode by the different methodology fits better in section 2 (and then we refer to it in section 3.1). In this sense, we have added the following paragraph in subsection 2.3:

"The mechanical separation fine/coarse mode in the detailed size distribution is used as well to estimate the optical thickness for fine and coarse mode at 440, 675, 870 and 1020nm, from the AERONET aerosol retrieval algorithm outputs. The particular values at 500nm, $\tau_f(500)$, have been interpolated for our validation study. Note that the way that the two modes are separated by the AERONET aerosol retrieval algorithm represents itself an inherent source of error to estimate fine/coarse mode optical thickness. In fact, the distribution of fine and coarse particles are continuous entities which overlap between them and they spread beyond the border established by the separation point or cutoff. As explained by O'Neill et al. (2003), a simple analysis of Mie kernels would show that the optical depth due to coarse particles for radii smaller than the cutoff (wrongly included in $\tau_f(500)$ calculations) is larger than the optical depth due to fine particles for radii larger than the cutoff (wrongly excluded from $\tau_f(500)$calculations). Therefore, the fine mode optical depth is overestimated while the coarse mode optical depth is underestimated. This effect is typically small, and it is more significant if the coarse mode dominates. Neither SDA nor GRASP-AOD application present this issue since the two modes can overlap in both algorithms. In the case of GRASP-AOD, the primary outputs are two independent log-normal functions which represent separately the fine and coarse mode as aforementioned. The values of $\tau_f(500)$ and $\tau_c(500)$ are derived from the aerosol optical depth values calculated individually for each log-normal function."

Page 23, last line: Is there a typo or a lost sentence within the text? Please, correct.

Thank you very much. It was part of the following sentence before modification. It's removed now.

Page 24, line 424: “the interest of presenting”

Yes, thank you again.

Page 36, second paragraph: In this part of the text is stated that the main interest of having aureole measurements is adding extra information for improving the coarse mode characterization in situations of partial cloudiness. However, I consider that this improvement cannot be linked only to conditions of partial cloudiness. Furthermore, there are other possible and important applications in the use of this type of measurements, such as quality control, cloud screening, among others, that should be acknowledge. Regarding the use of aureole measurements to improve the aerosol characterization, there are published papers that have also followed this philosophy, such as the work published by Román et al. (2017). These authors proposed the use of an all sky camera to add aureole information into the GRASP code. Please acknowledge in this Section.

Thank you for your comments here. However, we believe that there are some misunderstandings at this point.

First of all, when we talk about aureole measurement, we do it from the point of view of the retrieval improvements (information contained). In these regards, we can only conclude that the characterization of coarse mode is considerably better as shown in the section later on. Certainly, the use of new specifically designed aureole scenarios is helpful for other aspects as the referee stated. Nevertheless, these aspects are out of the scope/interest of this paper, moreover, considering that we have used aureole measurements from existing almucantar and not any kind of new specific scenarios. On the other hand, the mention of “cloudy conditions” refers to the fact that if there is clear sky conditions we can directly benefit from the use of the whole almucantar measurements (AERONET aerosol retrieval) instead of performing GRASP-AOD or GRASP-Aureole inversion.

Respect to the citation to the work by Roman et al. (2017), please note that both authors were co-authors of the study. Therefore, we are aware of the strengths and limitations of that study, especially in those aspects related to the use of the GRASP code. In this sense, the use of GRASP forward module to validate the normalized radiance measurements obtained from the sky camera was revealed as a solid tool. However, the results obtained by the use of GRASP retrieval in that work were quite criticized (internally by coauthors and by external referees) and still lack validation as highlighted in the conclusions of the paper. Nevertheless, the work was already cited in other parts of the study (overall we have a quite positive vision of that scientific work) but we do not acknowledge it intentionally in this section since we reckon it would be

misleading for the readers. Further details are given in the answer to the following comment since we think both comments are related.

Page 36, line 645: Refractive indices are necessary to run the GRASP-AOD, even when aureole measurements are performed. But, taking into account that aureole measurements are relatively insensitive to chemical composition, do the authors consider is still relevant the use of climatological data, or the effect of the uncertainty on the refractive index in this case is less important?

We do agree with the referee that aureole measurements are not sensitive to refractive index. This affirmation can not be concluded at all by the work of Roman et al. (2017). In that work, the first author proposed to retrieve the refractive index only with relative aureole measurements and aerosol optical depth measurements. It should be noted that the election of that inversion strategy was exclusively chosen by the first author and was done against the advice of GRASP retrieval experts of that study: O. Dubovik, D. Fuertes, T. Lapyonok and B. Torres. The use of pre-fixed refractive index (and sphericity parameter) was highly recommended, and this recommendation was done based on previous sensitivity studies.

Thus, the study “Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements” by Dubovik et al. (2000) concludes that to accurate retrieve the refractive index (and the derived single scattering albedo) scattering information between 3°-150° is needed. The study points out that if the scattering information is reduced to 3°-100° there is already some loss of information, but the results are still acceptable (especially if there is enough aerosol load). If the scattering information is smaller than 3°-100°, there is a dramatic decrease in retrieval accuracy for the refractive index (see for instance figures 4, 8, 10 and 12 of that study). As a result, traditionally AERONET only gives the label of quality assured if the solar zenith angle of the almucantar measurement is higher than 50° (which assures scattering information between 3°-100°). Other studies that came after as Torres et al. 2014 (figures 3 and 10) or the more recent Sinyuk et al. 2020 are in line with the aforementioned results. The latter study points out that the use of Hybrid scans reduces the requirement of solar zenith angle to 25°, but because in this new configuration a scattering range between 3°-100° is assured. It should be also remarked that all the aforementioned well-conceived sensitivity studies were based on well calibrated absolute radiances (with an estimated 5% error) while the study by Roman et al. (2017) made use of normalized radiance measurements with errors up to 10-14% in a much shorter spectral range (469 - 608 nm against 440 - 1020 nm), which implies much less information contained. For all these reasons, we think that to retrieve the aerosol refractive index with the methodology proposed by Roman et al. (2017) does not respond to a sufficient scientific evidence. That’s why

we believe that to cite the study by Roman et al. 2017 here could mislead the readers and make them believe that aerosol refractive index could be actually retrieved by aureole measurements (added to aerosol optical depth measurements) while several other studies have shown the opposite.

To answer specifically the referee's question, there is a strong correlation between the real refractive index and the fine mode characteristics due to the anomalous diffraction theory of Van de Hulst (Van de Hulst, 1957) as primarily discussed in Yamamoto and Tanaka (1969) and later by King et al. 1978. In this sense, if the real refractive index is not correctly retrieved it represents a source of error in the retrieval of fine mode parameters. As previously commented, the aureole measurements used in this section do not present a particular advantage to retrieve the real refractive index, since we would need scattering information between 3°-100°. That's why we should provide exactly the same information (in terms of refractive index) as in the regular GRASP-AOD application.

Pages 38-40, lines 704, 711, 714 and 744: The statement about the excellent agreement for fine mode is repeated throughout the conclusion section. Please avoid using redundant conclusions in this section.

Thank you for your comment but we believe that we are talking about different things in each paragraph and we would like to keep as it is. First in line 704, we talk about the characterization of $\tau_f(500)$. In the paragraph from line 709 and 720, which includes lines 711 and 714, we summarize the results obtained for R_{Vf} and C_{Vf}. Finally, the line 744 corresponds to the paragraph of the comparison of GRASP-AOD with other codes that performs only with AOD measurements. We think that it is important to highlight the fact that it is the only existing code that gives a characterization of the fine mode radius and volume concentration. Maybe to say again that it works only in certain circumstances (AOD(440) > 0.2 and AE >1.2) could be avoided, however, please note that the other referee's comments demand to recall this result (the aforementioned conditions) and the consequent reduction of data in its applicability.

Page 39, line 739: Spectral Deconvolution Algorithm is written here without the acronym, as the first time in the conclusion section, despite "SDA" has been mentioned in previous lines. Please homogenize the use of acronyms in the text.

Thank you very much for your comment. We have corrected it.