Response to the Anonymous Referee #3 comments for the manuscript "Retrieval of aerosol properties from zenith sky radiance measurements" By Sara Herrero-Anta et al. in AMT

First of all, we would like to thank the time and effort of the referee for their detailed review of the manuscript. Reviewer comments (RC) are in black font and author comments (AC) are in red.

Author's answer to Anonymous Referee #3

RC: The manuscript presents a new methodology to calibrate ZSR from a ZEN-R52 instrument and a new method based on GRASP to retrieve aerosol optical properties from the calibrated ZSR values. The new methodologies present advantages with respect to previous works cited in the manuscript, namely the calibration without laboratory measurements and the retrievals without the need of local lookup table. The obtained results show a strong potential of both methodologies to make this instrument useful in different locations. Therefore, I clearly agree that this study fits within the scientific innovation, quality and the scope of Atmospheric Measurement Techniques.

The manuscript presents high scientific level, significance and potential for application of the work. It is well written and well structured, the objective and applicability is clear and the approach is technically well justified and validated. The abstract is accurate and concise, the introduction properly presents the topic background, previous works on the subject are properly cited and the new points are clearly indicated. The data and methods are well explained, and the rest of the sections are comprehensive and detailed. To my view, sections 3 and 4 have the same weight as section 5 and contain technical results as it is expected in this kind of works, therefore I do not consider that the results are only in section 5.

I propose that this article is accepted for publication, after improving some minor aspects that, to my view, will make the work more robust.

Minor aspects:

RC: Line 34: please give a short indication on the conclusions obtained for volume concentrations.

AC: A short indication has been added in the abstract:

L34-36: 'The comparison against independent values from AERONET presents r^2 values of 0.57, 0.56 and 0.66, and uncertainties with values of 0.009, 0.016 and 0.02 $\mu m^3/\mu m^2$ for VCT, VCF, VCC respectively'.

RC: Line 263: It is stated that a "clear trend" is shown in Figure 3 e and g. However, those fittings only present r2 = 0.01, so in my view this is not a clear trend at all. I agree that 675 nm and 870nm channels show such a trend once the median values are taken for the 2°C-bins, i.e., in Figure 3 f and h. Please rephrase those sentences to make this clearer.

AC: We agree with the referee, and we consider that it is not a clear trend. The new sentences are:

L270-275: "In the left panels (a, c, e and g) of Figure 3 all data points are represented together with the linear fit, showing a negligible dependence on temperature for 440 and 500 nm. For 675 and 870 nm channels this dependency presents slopes of the linear fitting of 0.008 °C⁻¹ and 0.0036 °C⁻¹, respectively. These values are higher than the 0.0002 °C-1 obtained for the other two channels, which led us to consider a temperature correction for 675 and 870 nm."

RC: Figure 4 and line 861: Here the acronym ZSRDSC_TC is used for the Dark signal and Temperature corrected ZSR. However, in the text (e.g. in Equation 2 and lines 269 or 278) this is simply called ZSRTC. Please homogenize the acronyms to avoid confusion.

AC: It has been homogenized to ZSRTC following the referee comment.

RC: Line 291: This seems a bit confusing here. I agree that the proposed calibration method is better in the sense that the same introduced ASTM-E490 solar spectrum to calculate ZSRSIM will be then applied to the calibrated ZSRZEN before using them as input for GRASP inversion. Therefore, the statement "since there is no need for extraterrestrial spectrum normalization" may be misleading. Please rephrase those sentences.

AC: This sentences have been rephrased to make it more understandable as:

L299-305: "However, when using the calibration method developed in this study, the same normalization factor applied to the ZSR simulated by GRASP (ZSRSIM) can be applied to the calibrated ZEN-R52 measurements when using them as input to GRASP for the inversion. This way it can be avoided the introduction of a systematic error due to the normalization required by GRASP inversion algorithm. It means that this calibration method is better suited when using the ZSRZEN values as input for GRASP to retrieve aerosol properties, since we could work directly with the normalized radiances from GRASP."

RC: Section 3.5: it is stated that the ZSRZEN observations are compared against two different scenarios of CE318. However, the comparisons in 3.5.1 and 3.5.2 are not "treated" equally, or used for the same purposes. Only the bias statistics (MBE, Md, SD) from PPL comparison are considered and actually used as GRAPS "noise" input for each channel, while the same statistics (Md and SD) from CM are not used later. It seems that the CM comparison was actually more used to confirm the that the SZA and ZEN error criteria are successfully filtering out cloudy cases, but the deviations are not used as the reliability of each channel. If this is the case, please rephrase some parts of this section so that this is clearer.

AC: This comment is entirely correct. We primarily used the CM comparison to verify the source of errors for some SZA when the sun is not obstructed by clouds and observe the calibration behaviour qualitatively. However, to accurately quantify the uncertainty of the ZEN measurements we believe it is essential to exclusively consider measurements taken in the absence of clouds. This is mainly because, in this study, we are only utilizing measurements under cloud-free conditions for the retrieval of aerosol properties. In addition, the ZSR measurements strongly vary in short time periods under the presence of clouds due to the variability of clouds. This fact Implies that for a good comparison between ZEN and CIMEL measurements under cloudy conditions, the time of both measurements should be exactly the same. Unfortunately, it is not possible, the ZEN measurements are an average along one minute, while CIMEL measurements are quasi-instantaneous.

To clarify this, the following paragraph has been added in section 3.5.1:

L338-344: "This comparison against the cloud mode measurements will not be used to quantify the uncertainty of the ZEN measurements; it is because clouds are very variable and, therefore, the recorded signal. Therefore, we should need to compare both measurements carried out at exactly the same time; but this is not the case since ZEN measurements are 1-min averages while CE318 photometer measurements are quasi-instantaneous. In addition, for the retrieval of aerosol properties, it is necessary to employ measurements under cloud-free conditions, therefore, the results obtained in following comparison will be the reference ones."

RC: Line 404: are the values of MBE 0.23 to 0.11 correct? These are not the values shown in Figure 7.

AC: Yes, they are correct, a mistake was in the figure. It has been changed in the new version of the manuscript.

RC: Line 418: SD value of 33.2% is not matching with the value appearing in Fig. 8.

AC: The correct value is the one appearing in the Fig. 8. This error has been corrected in the new version of the manuscript.

RC: Lines 466-468: is this statement correct? In the previous paragraphs, you explain that the AOD values are clearly overestimated by GRASP-ZEN and also in Figure 10 the VCF, VCT and VCC show the highest deviations in % (12 to 70%).

AC: Regarding extensive properties, such as volume concentration, we are focusing on absolute differences rather than relative differences. This is because these magnitudes can reach very small values, leading to significant percentage differences even when the actual difference is relatively small. Thus, if these values are considered, the results are acceptable. However, for the radius and standard deviation, the possible values are bounded, making it more appropriate to analyse the results considering percentage differences.

RC: Lines 502-504 and Figure 12e: I do not think this is needed, it is redundant plot.

AC: This has been removed in the new version of the manuscript.

RC: Lines 516-520: since the sensitivity study in section 4 was used to set the accuracy and precision of the proposed method, I would emphasize here in the results (section 5) whether the observed differences are within those accuracy and precision values or not.

RC: Line 550: again, I would not speak here about "uncertainty", as it is understood that you obtain such uncertainty from the sensitivity study in section 4. I would better say that the dispersion of the differences are (or not) within the uncertainties obtained in section 4 and the uncertainties offered by AERONET.

AC: We understand the comments, but the uncertainties obtained through the synthetic analysis may not correspond to the actual uncertainties. In the synthetic analysis, for instance, the same radiative transfer model is used in both the forward and inversion parts. As a result, the uncertainty in the radiative transfer model itself is not taken into account in the synthetic study, but this uncertainty is propagated in the results when real measurements are inverted. Therefore, we consider it more appropriate to discuss about 'uncertainty' when comparing against AERONET products, which are realistic, especially AOD. To differentiate between the uncertainties in both cases, we have renamed the uncertainty obtained in the synthetic study by "theoretical uncertainty" and we have tried to add comments about if the obtained results are within or without the theoretical uncertainty.

RC: Line 563: the sentence is not clear, what did the methods reduce?

AC: This sentence has been modified as next in order to be more clear:

L585-587: "None of these methods significantly improved the retrieval of aerosol properties; but they did reduce the computation time (the data of a full day are inverted all at the same time)."

RC: Section 5: because of the strong importance of sections 3 and 4, that already contain technical results within the scope of the work, I will not call section 5 "results". I propose to change the name of the section to something more describing the application of the methodology to measurements database.

AC: We agree with the reviewer, hence we have renamed 'Results' Section as 'GRASP-ZEN application to ZEN-R52 database'.

RC: Other corrections or typos:

-Line 111: "an uncertainty" instead of "and uncertainty"

-Line 224: "On the contrary"

-Line 234: "represents"

-Line 296: "whole"

-Lines 324-325: "at the same fixed angles regards the SZA" could be removed, it seems redundant.

-Line 426: "with a"

-Line 467: "but not"

-Line 517: maybe better "sensitivity study" as it was previously called like that.

-Line 551: 0.020 μ m3/ μ m2 according to the plot

AC: All these typos have been corrected.