Response to the Referee Francisco Molero 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 his detailed review of the manuscript. Reviewer comments (RC) are in black font and author comments (AC) are in red.

Author's answer to Referee Francisco Molero

RC: The manuscript titled "Retrieval of aerosol properties from zenith sky radiance measurements" by Herrero-Anta et al., presents the procedure to extract aerosol properties from zenith sky radiances measured by a newly developed instrument. The inversion algorithm is performed with GRASP and the results are compared with AERONET products. The methodology is well explained, providing details of the calibration, inversion strategy and sensitivity analysis. The manuscript is well written and it presents a substantial contribution to scientific progress within the scope of Atmospheric Measurement Techniques. There are several issues that require a better explanation, and also some minor issues and typos that should be corrected, highlighted in the attached file.

Firstly, the main difference of this measurement strategy respect to AERONET is the use of vertical data instead of that derived by multiple-angles. The AERONET instruments perform principal plane and almucantar measurements to obtain angle information, while the ZEN instrument only measures vertically. Please comment on these different strategies and their effect on the inversions. For instance, would CIMEL vertical measurements (like "Cloud mode" but on clear-sky days) be as reliable as PPL and almucantar?

AC: The reliability of the sky radiance measurements relies on the instrument configuration, not on the geometries. The cloud mode measured by the CIMEL is as reliable as any point of the PPL or the Almucantar scans. However, for the inversion purposes is much more useful to have a wide range of scattering angles to have more information about the aerosol's scattering.

The "cloud mode" but on clear-sky conditions could be implemented as a routinary measurement in the photometers, and these measurements could be used to retrieve AOD and other aerosol properties applying the same method used in this work with the ZEN-R52 instrument. However, we observed that this method is mainly useful to retrieve AOD values, and the photometer is capable to obtain AOD values in a simple way only pointing to the sun. Then, it has not too much sense to configure photometers to carry out only ZSR measurements.

RC: The authors conclude that the GRASP-ZEN measurements produce reliable volume concentration estimations, but not radii estimations. Is it related with the limitation on the measurements (only vertical) or the inversion procedure (limited to five size distributions)?

AC: In this case is related with both. The scarce of scattering angles (only in the zenith), led to the need to apply an inversion procedure very constrained. This inversion procedure limits the retrieved size distribution (radii and standard deviation) to linear combination of five size distribution. It implies that plausible solutions can be rejected if they are not a combination of these five distributions.

This was mentioned in:

L470-476: *"The reason for the observed overestimation could be in the limitations of the GRASP-ZEN method based on the 'models' approach, which only allows to retrieve aerosol properties within the properties of the five aerosol types. It means that, for example, if the real aerosol has a median radius of fine mode bigger than the ones of the five 'models', then the GRASP-ZEN retrieval will underestimate the real median radius of fine mode and this difference will be compensated unbalancing other aerosol properties to fit the measured ZSR and the synthetic ZSR values of the retrieved aerosol scenario (to reduce the residual differences in ZSR values)."*

We could consider a different inversion procedure, as a bimodal size distribution for the inversion, that could improve the retrieval of aerosol properties. But this possibility is not feasible for the current work since this would imply the retrieval of many parameters, increasing the unknowns, and we still have only four ZSR measurements.

The possible alternatives were also discussed in:

L576-588: *"All the results of this paper have been obtained using the GRASP-ZEN methodology based on the 'models' approach, which is a suitable option for the current study due to the reduced number of radiometric observations provided by the ZEN-R52. However, the versatility of GRASP code allows different strategies for the retrieval of aerosol properties. In this sense, we have considered other strategies in this study to choose the one which provides the best results. These strategies are based on the temporal multi-pixel approach offered by GRASP (Lopatin et al., 2021), that constraints the variation of aerosol properties in time, forcing them to vary smoothly. The multi-pixel approach was firstly used in combination with the 'models' approach. In order to avoid the problems derived of having fixed aerosol models with fixed aerosol properties, the temporal multi-pixel was also used assuming the size distribution as a bimodal (fine and coarse modes) log-normal distribution and the refractive indices have no dependence on wavelength. 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). Nevertheless, these strategies could be considered for future aerosol retrievals."*

RC: Secondly, the normalization by extraterrestrial spectra is not clear. The authors conclude (lines 578 & 579) that "proposed methodology incorporates the advantage that it includes the normalization used by GRASP and therefore there is not any need to use extraterrestrial spectra to normalize the data when they are used as input in GRASP". However, section 2.2 "GRASP methodology" seems to imply that normalization is required. Please clarify this issue.

AC: The radiances in GRASP are normalized, so we could directly work with the GRASP normalization when using ZEN-R52 measurements which were calibrated using the methodology described in Section 3 (by comparison with measurements simulated by GRASP). When applying the calibration, the ZSR simulated by GRASP are multiplied by the extraterrestrial irradiance to convert them into radiance units, but we could avoid this step, or revert it applying the same factor to recover the normalized GRASP radiance.

If we had to use sky radiances measured by other instruments or methods, then yes, an extraterrestrial normalization must be applied to the measurements if we want to use them as input to GRASP.

It was mentioned also in lines 298-304, which has been rephrased to:

L298-303: "*This fact can increase the relative differences between the two calibration methods, together with the lack of temperature correction in the second one. However, when using the calibration method developed in this study, the same normalization factor applied to the ZSR simulated by GRASP (ZSR_{SIM}) 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.*"

Also a clarification is included on section 2.2:

L158-162: "*The standard ASTM-E490 solar spectrum has been used in this work for the normalization of Eq. (1). This spectrum was calculated for moderate solar activity and medium Sun-Earth distance; therefore, it has been corrected from Sun-Earth distance for each day of the year. This way, the normalization factor must be applied when using data in radiance units as input to GRASP and to transform the output normalized radiances from GRASP into radiance units.*"

RC: Finally, some technical issues:

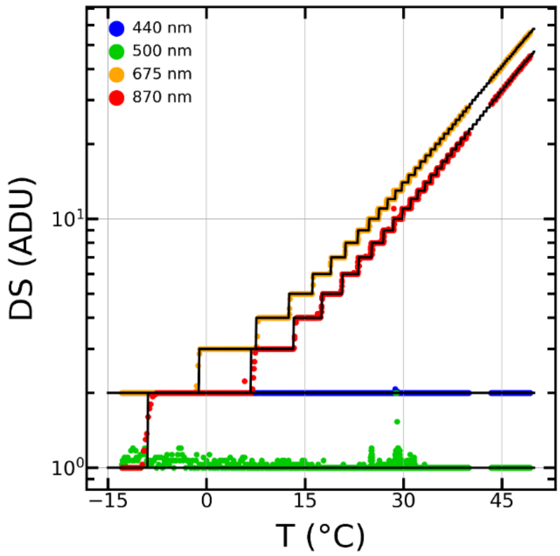
- The procedure described in Section 4.2 AERONET scenarios is not clearly explained. Does the AERONET inversions determine the five aerosol types concentrations? Or the AERONET information is input into GRASP directly?

AC: The AERONET information is input into GRASP directly. This has been added in the new version of the manuscript:

L452-453: *"In this case, the AERONET retrieved aerosol properties (size distribution, refractive indices, etc.) are used directly as input in the GRASP forward module to simulate the ZSR values."*

- Details in Figure 1 are difficult to see. Maybe a logarithm Y-axis would help. Also, the details of the fitting may be added, and a residuals graph, to check randomness.

AC: In order to improve this, Figure 1 has been updated with an y-logaritmic scale as can be seen next.



Regarding the residuals, they have been included in the supplementary material as Figure S2 (see the figure below). The results about residuals have been also discussed in the new version of the manuscript by these new sentences:

L234-236: "*The residuals between the modelled and real DS are shown in the supplementary material (Figure S2); these residual values are within the instrument resolution for all channels.*"

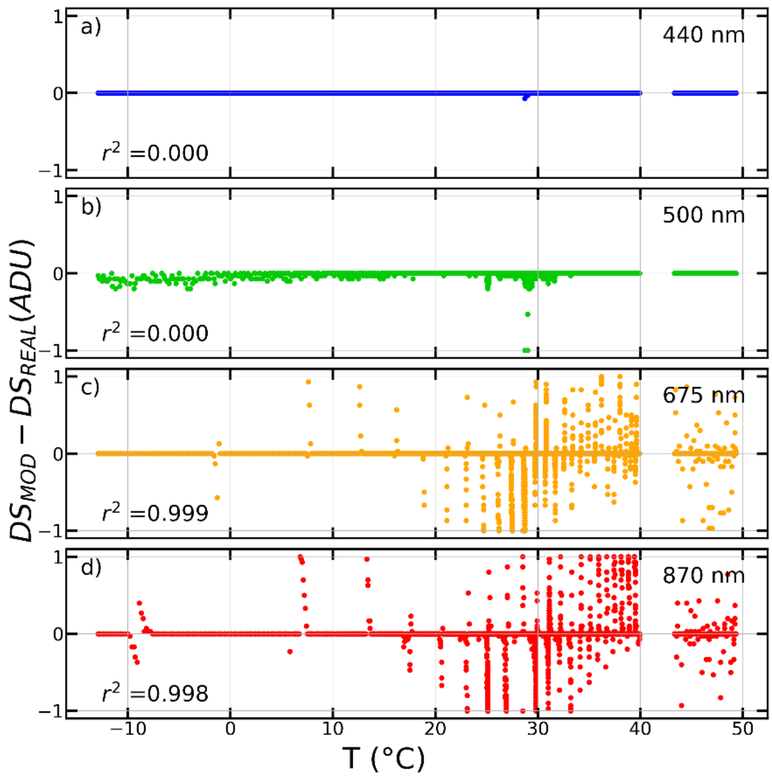


Figure S2. Residuals graph for the dark signal (DS) correction for the thermal chamber period. The differences between the modelled dark signal (DS_{MOD}) and the correspondent real dark signal recorded by the ZEN-R52 (DS_{REAL}) are plotted against the temperature at a) 440nm, b) 500nm, c) 675 nm and d) 870 nm. The determination coefficient (r2) obtained for the direct comparison of the DS_{MOD} against the DS_{REAL} is also included.

RC: L193 How are the fine and coarse modes defined?

AC: The inversion procedure retrieves the proportion of 5 different aerosol size distributions. Each one of these five size distributions is defined for fine and coarse modes, as can be observed in Figure S1. Then, the retrieved fine parameters are the combination of the fine mode of the five aerosol models; the same for the coarse.

To clarify this issue, we added the next in the new version of the manuscript:

L195-199: "*The size distribution of the five models is defined for fine and coarse modes, hence the retrieved parameters are also calculated for these modes. Then, the obtained size distribution parameters are volume median radius of fine (RF) and coarse (RC) modes, standard deviation of lognormal distribution for fine (σF) and coarse (σC) modes, and aerosol volume concentration for fine (VCF) and coarse (VCC) modes and the total value (VCT).*"

RC: L199 But non-convergent cases don't produce simulated ZSR. How is the residual information calculated for those cases?

AC: The non-convergence issue was not clear. All the retrievals provide simulated ZSR. However, we assume that if the residual is higher than a threshold, the retrieval does not show a good convergence (non-convergence cases). Regarding how the residual is calculated, we add the cite of Román et al. (2022), where the residual formula is written. We have tried to clarify these issues about convergence, thresholds and residuals adding the next text in the new version of the paper:

L199-205: "*Each output, one per retrieval, provides the relative residual differences between the measured ZSR (input) and the ones generated after the inversion (simulated by GRASP forward module under the retrieved scenario) for each wavelength (Román et al., 2022). This residual information will be used to evaluate the goodness of the retrievals; if the residual at one or more wavelengths is above an established threshold, the inversion is rejected (assumed as non-convergent). This threshold, which varies with the wavelength, has been set as the absolute value of the accuracy plus the precision for each channel of the ZEN-R52 (see Section 3.5.2).*"

And also the next:

L372-373: "*These accuracy and precision values will be used in the convergence criteria mentioned in Section 2.2.2.*"

RC: L203-204 Is this study published? In such case, add reference

AC: It is not published, it is just a simple monthly climatology calculated for this study. It has been obtained using all data available in AERONET in Valladolid for the 2012-2021 period as it is mentioned in the manuscript.

RC: L225-226 Add details of fitting to the figure. Also, figure 1 may be more interpretable with y-axis in logarithm scale.

AC: These changes have been applied as has been commented above.

RC: L309-310 Why is it necessary to download them from AERONET webpage? Aren't they available from the instrument memory?
& L334-335 Again, why download from webpage is required? Shouldn't these data be available within the instrument?

AC: The memory within the instrument is limited, especially older instruments, which are the ones measuring at these geometries. Anyway, the raw data is frequently stored and sent to CAELIS and AERONET databases. These instruments record only raw data, it implies that we cannot obtain the sky radiance directly from the instrument memory. Then, the radiances cannot be directly obtained from instrument memory since we need to know the calibration coefficients. The data of all photometers belonging to AERONET is sent to AERONET server; AERONET also knows the calibration coefficients since these photometers are calibrated following the AERONET procedures. AERONET provides the well calibrated photometer products like sky radiance. Therefore, it is more comfortable and simpler to download the data directly from AERONET (warrantying the data quality assurance), specially for researchers that do not participate in the photometer calibration tasks. Fortunately, our group, within ACTRIS/AERONET-Europe branch, participates in the AERONET calibration tasks of part of the photometers of the network. It helps to know the calibration coefficients, which are stored in CAELIS; this makes that we have access to sky radiance values in the principal plane directly from CAELIS.

RC: L316 Only the sun, or the sky?

AC: Clouds in the sky may affect the ZSR measurements, especially if they are in the zenith. For that the ZEN error (now renamed as ZEN variability as suggested by the reviewers) mentioned in section 3.2 is used, since clouds strongly affecting the measurement would produce a high ZEN variability, due to the high variability of the measurements within the minute, and therefore the consequent high standard deviation. But in this case, we are referring to the observed outliers for SZA<30º in Figure S3, that are due to sun stray light as confirmed here since we do not observe those outliers when there is an obscured sun like in the Cloud Mode observations. Therefore, we assume it is only the sun.

RC: L380 This sentence isn't clear. An extreme AOD event should be AOD@440nm above 0.7, not below. Please rephrase more clearly.

AC: This sentence has been rewritten as next:

L404-407: "*Nevertheless, for lower SZA conditions (Figure S5; panels a-d) there is a clear sensitivity to type and aerosol load for AOD at 440 nm, at least for values below*

0.7; values above 0.7 are assumed for extreme AOD events (Mateos et al., 2020) and therefore are unusual".

RC: L410 Are all the scenarios real, in terms of aerosol concentration of all types? Would a classification of the scenarios (Saharan intrusions for those with high dust concentration, Urban pollution event for those mostly influenced by urban aerosol type etc...) assist in the interpretation of convergent inversions?

AC: This should not be a problem for the convergence. In this section we have used the same scenarios for different SZA and the amount of convergence values has changed, so it should to be related with the SZA. Nevertheless the 'models' strategy from GRASP does not really care about the amount of each model, it just uses common aerosol properties and tries to replicate the observed measurements adjusting the fraction of each model and the total concentration. If it is able to reproduce the forward, it should be able to conduct the inversion.

RC: L425-427 This procedure is not clearly explained. Does the AERONET inversions determine the five aerosol types concentrations? Or the AERONET information is input into GRASP directly?

AC: AERONET information is input into GRASP directly. It has been clarified in the text with this new sentence:

L452-453: "*In this case, the AERONET retrieved aerosol properties (size distribution, refractive indices, etc.) are used directly as input in the GRASP forward module to simulate the ZSR values*".

RC: L443-445 But if the five aerosols types can explain the AERONET measurements, the scenario should be reproducible. Please describe this procedure in more detail, as mentioned in the previous comment.

AC: When we do the forward simulations with a RTM we are in a direct problem, that has only one solution. This correspond to the sentence added in the comment before: "*In this case, the AERONET retrieved aerosol properties (size distribution, refractive indices, etc.) are used directly as input in the GRASP forward module to simulate the ZSR values*".

But when we do the inversion there might be a high number of scenarios that would reproduce the same measurements, hence the importance of constraining the solutions. In this case we can reproduce the sky radiance measurements and even the AOD, but the microphysical properties are not reproducible because we are employing only five fixed aerosol types for the inversion. For example, if the coarse radius of the AERONET scenario is out of the achievable values using the combination of the five models, this under or overestimation could be compensated with a under or overestimation of the refractive index (or any other combination of aerosol properties providing an effective solution); this retrieved scenario could reproduce the measurements, even the AOD, but the aerosol microphysics properties are only effective, not necessarily the real. Anyway, the five aerosols are not completely explaining the AERONET measurements since an overestimation of the inverted AOD values can be seen.

This limitation is explained in:

L470-476: *"The reason for the observed overestimation could be in the limitations of the GRASP-ZEN method based on the 'models' approach, which only allows to retrieve aerosol properties within the properties of the five aerosol types. It means that, for example, if the real aerosol has a median radius of fine mode bigger than the ones of the five 'models', then the GRASP-ZEN retrieval will underestimate the real median radius of fine mode and this difference will be compensated unbalancing other aerosol properties to fit the measured ZSR and the synthetic ZSR values of the retrieved aerosol scenario (to reduce the residual differences in ZSR values)."*

RC: L474 Please relate with dates and number of measurements

AC: Dates are the correspondent to the total available period of ZEN-R52 measurements, April 2019 to September 2021. It has been added in the new manuscript version:

L500-504: "*Once the ZSR_{ZEN} measurements have been calibrated, and the GRASP-ZEN method has been proved in Section 4 as capable to retrieve aerosol properties, the GRASP-ZEN methodology has been applied to the whole available dataset of ZEN-R52 measurements at Valladolid at the moment of the study. As result, a total of 222663 GRASP-ZEN retrievals have been obtained between April 2019 and September 2021.*"

RC: L525-531 The AERONET instruments perform PPL and almucantar measurements to obtain angle information, while the ZEN instrument only measure vertically. Please comment on this different strategies and their effect on the inversions. For instance, would CIMEL vertical measurements (like "Cloud mode" but on clear-sky days) be as reliable as PPL and almucantar?

AC: This comment has been answered above.

RC: L577-579 This is not mentioned before. It is not clear why no extraterrestrial spectra is needed since it is mentioned in section 2.2. Please clarify this statement

AC: This comment has been answered above.

RC: L583 On section 4, lines380, says below 0.7. Why has it changed?

AC: On section 4 it referred to SZA = 70º, and in this sentence it is for SZA≤50º. It was not clear then now it is detailed:

L603-606 "*An analysis with synthetic data has concluded that ZSR measurements are useful to derive aerosol optical depth (AOD), since these measurements are sensitive to aerosol load and type for the ZEN-R52 channels, at least for AOD at 440 nm below 1 for SZA ≤ 50º*".

RC: L606 Expanding from the previous comment about the difference in strategies between the AERONET (PPL and almucantar measurements to obtain angle information) and ZEN (only measure vertically), can the authors comment on the properties (volume concentrations, refractive indices, asymmetry parameters) that can be obtained and possible limitations?

AC: As mentioned on section 4.2, the retrieval of aerosol properties is limited to the properties of the five types of aerosols used for the inversion. They possess fixed properties, so the resulted aerosol must be a linear combination of them. For the volume concentration there should not be more limitations than the one derived from the fact of using fixed types of aerosol that can influence the inversion as mentioned on lines 469-475.

RC: General typing errors:

AC: All these typing errors have been corrected.