Manuscript: amt-2023-106
Title: Aerosol properties derived from COCCON ground-based Fourier Transform spectra

Response to Referee#2

The authors appreciate the overall positive response of the Referee #2 and we would like to thank for his/her constructive comments. In the following, the Referee suggestions (in bold) are in detail addressed (the author's responses are below).

General comments:

1. I would recommend to add an accronym tables, in order that the readers can find quickly what the different paragraphs are talking about, since there are a lot of accronyms used in the manuscript.

Authors: The acronym table has been included at the end of manuscript following the referee's suggestion.

2. I recommend to develop more information about the reference Fourier Transform Spectrometer: High resolution IFS 125 HR presented and validated in Barreto et al. 2020, and not only to cite Barreto et al. 2020 each time IFS 125 HR is mentionned. For instance, explain the resolution of IFS 125 HR when the resolution of EM27/SUN is discussed. Also during the validation of the AOD retrieval with EM27/SUN, since IFS 125 HR is presented as reference and Barreto et al. 2020 continuously mentioned, the authors should better give some values of the statistics of Barreto et al. 2020 regarding intercomparison AERONET vs. IFS 125 HR, and discuss and interprete these results to the intercomparison results of AERONET vs. EM27/SUN presented in this manuscript.

Authors: We agree with the referee that it would be appropriate to directly introduce some of the results presented in Barreto et al. (2020) instead of referencing this article. We have included the following information in the manuscript:

Section 4.1:

"**Seven of the presented spectral bands (B2-B8) were selected with respect to those presented in Barreto et al. (2020), while an additional channel (B1) has been incorporated for the purposes of this study due to the wider coverage range of the EM27/SUN InGaAs detector.**".

Section 5.1, line 261:

"**These values are relatively low compared to that of the high-resolution IFS 125HR system at the same station, which ranged between about 1.61%month^{-1} (B8) and 1.75%month^{-1} (B2), reaching a total decrease of 14.5% (B8) and 15.8% (B2) from May 2019 to February 2020 (Barreto et al., 2020)**".

Specific comments/questions

1. Line 4 or Line 13: You mention "low resolution" -> Maybe specify "0.5 cm-1" in brackets

Authors: This statement has been modified in the manuscript following the referee's suggestion.

2. Line 42 and Line 51: Specify the resolution of IFS 125 HR (line 42) and of "low resolution" EM27/SUN (line 51).

Authors: The spectral resolution of the Fourier Transform spectrometers (FTS) depends on the optical path difference (OPD) used to measure the interference pattern associated with the solar beam. It is estimated as the ratio between 0.9 and OPD (Griffiths and de Haseth, 2007). Therefore, it has not a fixed value, but it depends on the measurement configuration, ranging from almost zero to the maximal spectral resolution given by the maximal OPD.

In the case of the EM27/SUN FTS instruments, they are operated at their maximal OPD (i.e. 1.8 cm) within COCCON, therefore their spectral resolution can be considered fixed at 0.5 cm-1. This information has been included in the Line 51 following the referee's suggestion.

Nevertheless, the high-resolution IFS 125HR spectrometer referred to in the Introduction section was operated at 0.02 cm^{-1} (i.e. OPD of 45 cm, reference of TCCON network) and truncated a posteriori at 0.5 cm^{-1} for the AOD analysis presented in Barreto et al. (2020). However, similar results would be expected if the spectral resolution had been increased until the maximal OPD of the FTS spectrometer (180 cm), resulting in a spectral resolution of 0.005 cm-1. Therefore, to avoid confusion, the information about spectral resolution of the high-resolution IFS 125HR is not included in the Introduction section.

Griffiths, P. R. and de Haseth, J. A.: Fourier Transform Infrared Spectrometry, John Wiley & Sons, Inc, New Jersey, USA, 2007.

3. Line 114: You give the instrumental resolution in cm-1 (0.5 cm-1). Maybe specify how it is in nm (for SWIR and NIR bands), since the rest of the study and the comparison with AERONET is given with wavelength and spectral band width in nm. -> This inconsistency is very visible in the legend of Figure 1: "EM27/SUN solar spectrum for the 870-2500 m ... resolution of 0.5 cm-1"

Authors: Following the referee's suggestion, Figure 1 has been modified by including an auxiliary y-axis with the spectral range in wavenumber. In addition, the equivalence of the spectral resolution in nm has been included in the figure caption for the coincident AERONET channel as a reference. Table below lists the equivalent spectral resolution in wavelength for all EM27/SUN micro-windows.

Band	Central Wavelength (nm)	Δλ (nm)
B1	872.55	0.038
B2	1020.90	0.052
B3	1238.25	0.077
B4	1558.25	0.121
B5	1636.00	0.134
B6	2133.40	0.228
B7	2192.00	0.240
B8	2314.20	0.268

4. Line 140: "Solar/lunar and sky measurements are normally taken every ~15 minutes" -> Can you verify this information, in my opinion it is more often (every 5 minutes)

Authors: We agree with this referee's comment. We have corrected the manuscript with the following information:

"Solar/lunar and sky measurements are normally taken every ~15 minutes or at fixed air mass intervals at specific wavelengths with a FOV of ~1.3◦ (Holben et al., 1998; Torres et al., 2013). **In the case of photometric information used in this paper, Cimel solar observations have been retrieved with a higher frequency, between 2 and 6 min.** The instrument is equipped with Silicon and InGaAs detectors and…"

5. Line 181: Typo: "3-year period" -> "3 years period"

Authors: This statement has been modified in the manuscript following the referee's suggestion.

6. Line 201: Formula V_lambda = V_0,lamdda * d-2 * exp(-m*tau_lambda) -> Since V_0,lamdda is later (Line 203) defined as the "instrument's signal at TOA", and not at the sun, the term "d-2" has to be cancel from the formula of Line 201 and from the description of Line 203.
d-2 is already integrated in V_0,lamdda, since V_0,lamdda = V_sun,lambda * d-2 (signal measured at the sun)

Authors: The authors agree with this comment. In the text should be stated that the $V_{0,\lambda}$ term represents the instrument's signal measured at TOA at the Earth-Sun distance of 1 UA, and therefore the distance correction term in Eq. 1 should clarify the ratio as $(1AU/d)^2=d^{-2}$, as it is written. However, further corrections in the manuscript have led us to eliminate the distance correction term. This correction is not necessary in the case of the EM27 observations considering the reduced FOV of the instrument (much smaller than the solar disk). In this case, our source can be considered not only as uniform but also as an extended source, distinct from what a photometer capable of measuring the entire solar disk can detect. The authors admit that is well known the existence of center to limb variations (CLV) that could cause changes in the measured radiance and correspondingly in the estimated AOD. However, according to previous studies (Blanc et al., 2014; Bernhard and Petkov, 2019), these variations are quite small when measuring away of the solar limb, as is our case. This statement can be also supported considering this effect is less pronounced in the NIR region and taking into account the pointing accuracy of the EM27/SUN. In this scenario, EM27/SUN measurements are not a function of the distance between the source and the observer. This is because both the solid angle subtended by the source and its flux density fall off as the inverse square of its distance, so their ratio is constant.

Section 4.2 has been changed accordingly.

References:

P. Blanc, B. Espinar, N. Geuder, C. Gueymard, R. Meyer, R. Pitz-Paal, B. Reinhardt, D. Renné, M. Sengupta, L. Wald, S. Wilbert: Direct normal irradiance related definitions and applications: The circumsolar issue, Solar Energy, Volume 110, Pages 561-577, ISSN 0038-092X, https://doi.org/10.1016/j.solener.2014.10.001, 2014.

Bernhard, Germar & Petkov, Boyan: Measurements of spectral irradiance during the solar eclipse of 21 August 2017: Reassessment of the effect of solar limb darkening and of changes in total ozone. Atmospheric Chemistry and Physics, 19, 4703-4719, 10.5194/acp-19-4703-2019, 2019.

7. Line 251-254: Why should an event increasing atmospheric turbidity lead to a lower V_0,lamdda TOA signal? The aim of the langley-plot method is to get rid of the atmospheric turbidity. It can be, that because of these events, the turbidity is to high and unstable, and then we cannot do Langley-Plot, but if we can do it (not too much turbidity and stable during sun rise / sun set) = langley plot (ln(I) va airmass) is a straight line, then the result should not be lower because of it. -> Can you please consider this question and give explanation. If not I do not agree with "... could also cause a loss of signal" (Line 253), at least if "signal" = TOA signal (V_0,lamdda)

Authors: The calibration performed with the EM27 at Izaña, as described in Section 5.1 (first and second paragraphs), was carried out under pristine conditions, following the criteria presented in Toledano et al. (2018). In total, 31 high-quality Langley plots were retrieved at the eight EM27/SUN spectral bands between December 2019 and September 2022 for our analysis, and these values are presented in Fig. 2 (a) and (b).

The significant EM27/SUN loss explained in line 250 is due to the environmental exposure of the EM27 tracker. Unlike what happens with the Cimel or other photometers with a protected solar tracking within the optical head, our EM27/SUN has a set of external mirrors that are constantly exposed to environmental conditions, such as dust or volcanic aerosols, which deposit on their surface every time the instrument is in operation. This degradation, estimated to be 24% as average across all bands during the entire period, must be taken into account in the AOD calculation process through "quasi-continuous" Langley calibrations. This process includes not only the Langley calibration (performed under pristine conditions in a Langley day) but also the "quasi-continuous" Langley calibration approach (in a non-Langley day) utilizing an estimated $V_{0,\lambda}$ from the smoothing spline functions derived from the 31 Langleys performed over the entire period (in a non-Langley day). Consequently, the calibration approach used in this study has been proved to follow the observed optical degradation of the system.

8. Figure 4:

8.1. please make two figures, one with 2019-2022 (whole period) and the other one with Dec 2019 - Dec 2020, with the open markers and the plain one, it is too confusing to interprete the graphic.

__Authors:__ The authors agree with this comment. A Figure 4 has been replace by this figure:

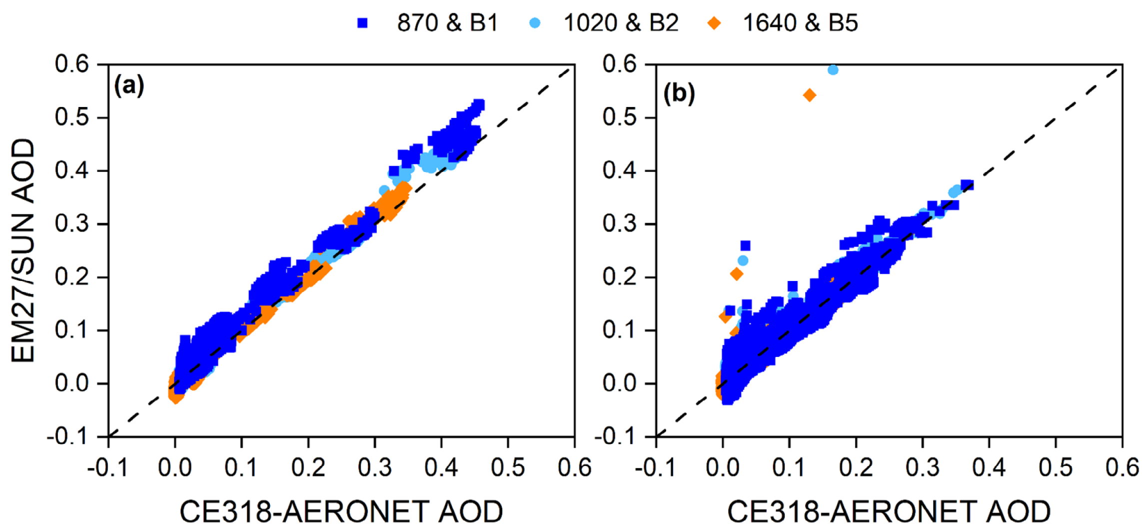

Figure 4. Scatterplot for the coincident EM27/SUN-AERONET AOD values from (a) December 2019 to December 2020 and (b) January 2021 to September 2022 considering the EM27/SUN B1 (870 nm), B2 (1020 nm) and B5 (1640 nm) micro-windows. The number of coincidences is 14575 and 2863 in the period January 2021 - September 2022 and December 2019 - December 2020, respectively.

8.2. In the legend "open circles" -> "open markers" (there are other open symbols than circles)

Authors: This statement has been modified in the manuscript following the referee's suggestion.

9. Lines 301-302: You recommand ideally one calibration / month -> Then you cannot use the system as an operational system on a site without opportunity of Langley-Plot calibration (urban areas, turbid areas, not high mountains, ...) -> Do you have to suggest other methods of calibration for these non langley-plot compatible sites?

Authors: Yes, according to the calibration methodology proposed in this paper, based on the Langley-Plot calibration procedure and smoothing spline functions to cover the calibration gaps, the EM27/SUN is not intended for AOD operational observations in polluted sites. However, we have demonstrated in this paper the potential of this system to provide simultaneous retrieval of column-integrated aerosol and trace gas information, which are important and complementary pieces of information for understanding atmospheric processes.

Further investigations must be undertaken to ensure EM27/SUN Langley-Plot calibration (compensation for the optical degradation of the system) when operating in non-pristine conditions. Possible solutions to this problem could include the design of protective domes to prevent system degradation during operation, or the use of high-intensity calibration sources and robust calibration transfers, as already implemented during sporadic field campaigns.

We have included this information in the manuscript in line 461 as follows:

"In this regard, our results demonstrate that the calibration approach used in this paper **based on Langley-plot regular calibrations and smoothing spline functions to cover the calibration gaps is adequate to compensate for the optical degradation of the system. Other possible solutions to address this issue could involve the design of protective domes to prevent system degradation during operation, or other absolute radiometric calibration procedures, such as using high-intensity calibration sources or robust calibration transfers, as already implemented during sporadic field campaigns by Gardiner et al. (2012), Menang et al. (2013) or Elsey et al. (2017)**."

References:

Elsey, J.; Coleman, M.D.; Gardiner, T.; Shine, K.P. Can Measurements of the Near-Infrared Solar Spectral Irradiance be Reconciled? A New Ground-Based Assessment between 4000 and 10,000 cm−1. Geophys. Res. Lett. 2017, 44, 10071–10080.

Gardiner, T.D.; Coleman, M.; Browning, H.; Tallis, L.; Ptashnik, I.V.; Shine, K.P. Absolute high spectral resolution measurements of surface solar radiation for detection of water vapour continuum absorption. Philos. Trans. R. Soc. A. 2012, 370, 2590–2610.

Menang, K.P.; Coleman, M.D.; Gardiner, T.D.; Ptashnik, I.V.; Shine, K.P. A high-resolution near-infrared extraterrestrial solar spectrum derived from ground-based Fourier transform spectrometer measurements. J. Geophys. Res. Atmos. 2013, 118, 5319–5331.

10. Lines 322-325: The authors seem to be satisfied with the agreement EM27/SUN to AERONET, even if the WMO criterium (U95) that has been mentioned is not satisfied. Maybe here it is worth to give some explanation about what values (percents of occurence in U95 or which softer criterium than U95) is expected from the authors to be satisfied. Here maybe it would be interesting to compare the performances of EM27/SUN to the one of IFS 125 HR, mentioning the values of the performances explained in Barreto et al. 2020.

Authors: In the submitted manuscript, we found traceability limits of 50%, 71%, and 84% for the respective EM27 coincident bands with AERONET. The new values, after correction, are quite similar (51.4, 70.4 and 82.2%). It is important to note that this study was not conducted with the aim of ensuring traceability between the two instruments (Cimel and EM27), but rather to provide additional comparison results that could be useful in understanding the performance of the EM27 spectrometer. As stated in the manuscript (line 326), it is important to consider that the EM27/SUN instrument was not specifically designed to offer the absolute photometric stability necessary for aerosol monitoring. Therefore, satisfying traceability limits in this specific case is not the purpose of our study.

We acknowledge the referee's suggestion that a comparison with the HR FTIR presented in Barreto et al. (2020) could add interesting information in this paragraph. We have included this information in the manuscript (line 330):

"**The low traceability identified in our study stands in contrast to the remarkable traceability established between CE318-AERONET and the IFS 125HR, as reported by Barreto et al. (2020). This disparity, evident despite employing identical methodology and spectral resolution, might indicate the existence of mechanisms introducing a variable spectral ordinate calibration in the case of the EM27/SUN,**."

11. Lines 328-330: I disagree with the assumption, that since U95 is defined for UV, it should be harder in SWIR+NIR. No, in the contrary: U95 is a criterium set in the absolute AOD difference that is larger in UV than in SWIR+NIR, since the AOD itself is larger. U95 should be in my opinion, from a statistically point of view easier to reach in SWIR+NIR since the AOD is lower. Of course, from an instrumental point of view it is different, but this has to be justified with other argument (signal noise ratios of the photometers, etc...)

Authors: In the manuscript, we have stated that U95 has been defined for the UV-VIS spectral range, and the uncertainty term has been considered wavelength-independent within this range. However, we have not mentioned that in the SWIR+NIR range, the U_{95} criterion may be harder/easier to achieve and that it needs to be re-defined. As far as the authors know, there is no detailed publication aimed at defining this U_{95} criterion beyond the UV-VIS spectral range. Considering this referee's comment and also the lack of investigations in this regard, we have decided to eliminate the sentence in lines 328-330.

12. Lines 341-342 vs Line 352-353: Line 341-342 mention that older studies (Toledano et al. 2019 and Barreto et al. 2020) say that for high dusty events, there is no Angstroem law, than at lines 352-353, you mention that this study has same results as older studies (the same: Toledano et al. 2019 + Barreto et al. 2020) and you have more interspectral correlation for high AOD and dusty. But: Angstrom law should not be a source of increasement of interspectral correlation? Can you develop / explain (not in manuscript but in comment) where should the higher interspectral correlation come from, if not from Angstrom law?

__Authors:__ In lines 341-342, it is asserted that the Angström Law is an unsuitable approximation for describing the spectral dependence of AOD in the SWIR (Short-Wave Infrared). This is particularly evident in the context of the referenced papers, which primarily focus on mineral dust and volcanic ash, the predominant aerosol species affecting this spectral range. The spectral variation of AOD arises from diverse interactions between atmospheric aerosols and solar radiation, contingent on their physical and chemical properties. Nonetheless, it is widely acknowledged that the Angström Law inadequately captures this spectral diversity, resulting in a notable overestimation of AOD in the infrared. Consequently, the Angström Law is eschewed in favor of a non-parametric Kendall rank correlation analysis to examine the spectral dependence of AOD obtained from the EM27/SUN instrument.

Our findings underscore spectral coherence among adjacent bands, particularly in high-AOD conditions characterized by elevated dust levels. Reduced correlations are attributed to instances of low AOD (owing to artifact presence) and potential inaccuracies in addressing absorption features, such as the H_2O absorption band in B3, in our analysis.

In general, we can conclude that there is a robust correlation between the AOD in proximate spectral bands (specifically selected considering their high atmospheric transmission), where the impact of aerosol is similar, and also (with the exception of B3), the impact of gaseous absorption is similar and low.

13. Lines 455-457: You give quantificated values of the evolution of calibration values for EM27/SUN and mention IFS 125 HR as reference... But without mentioning values of the stability/evolution of calibration values of IFS 125 HR.

__Authors:__ Following the referee's comment, for a better completeness of discussion, the reference values of the calibration coefficients evolution for the IFS 125HR have been included in the discussion of Section 5.1 as follows:

"**These values are relatively low compared to that of the high-resolution IFS 125HR system at the same station, which ranged between about 1.61%month^{-1} (B8) and 1.75%month^{-1} (B2), reaching a total decrease of 14.5% (B8) and 15.8% (B2) from May 2019 to February 2020 (Barreto et al., 2020).**"

14. Lines 462-463 vs Lines 483-486: At lines 462-463 you mention the need of monthly calibration of the system (that only works on some few calibration sites) and lines 483-486 you mention that the system should be applied in a measurements' network -> Most of the station of measurements' network are not compatible with Langley-plot

calibration -> Which method do you suggest for these stations to keep the instrument well calibrated without sending it to IZO or another calibration site every month?

Authors: We understand the referee's concern about the use of the EM27/SUN in an operational network, such as COCCON. This issue has already been addressed in the 9th question posed by this referee. As mentioned before, possible solutions to this problem could involve the design of protective domes to prevent system degradation during operation, or the development of absolute calibration procedures using high-intensity calibration sources.