

## (1) Comments from referees/public & Author's response

**Manuscript: amt-2023-106**

**Title: Aerosol properties derived from COCCON ground-based Fourier Transform spectra**

### **Response to Referee#1**

The authors appreciate the overall positive response of the Referee #1 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. In general, I would recommend to try to have an uncertainty analysis of the retrieved AOD.**

**Authors:** We agree with this referee's comment. We have added in the new version of the manuscript a new section (Section 5.3) with the uncertainty analysis performed by means of the Monte-Carlo method. We have removed the previous estimation of the uncertainty based on the comparison of CE318-AERONET and EM27/SUN independent techniques.

**2. AOD at 1640 nm needs a GHG correction. Is this already done and do you use actual FTIR GHG concentrations to do it?**

**Authors:** As stated in the manuscript, AOD values have been calculated following the AERONET procedures and the methodology proposed by Barreto et al. (2020). Utilizing the same approach as AERONET serves two purposes: firstly, it ensures the derivation of reliable AOD values due to the well-established and widely recognized nature of the AERONET methodology within the aerosol community; secondly, it enables a meaningful comparison between the new EM27/SUN AOD products and the AERONET reference AOD data.

Consequently, in this study, AOD at 1640 nm has indeed been adjusted for gaseous absorption through the use of the AERONET climatology correction. This correction is contingent upon the current Precipitable Water Vapor (PWV) content, while the quantities of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) are parameterized based on the present atmospheric pressure at the station (P) and at sea level (P<sub>o</sub>), following the methodologies outlined by Smirnov et al. (2004) and Giles et al. (2019), as detailed below:

$$\text{AOD}_{g,1060} = \text{AOD}_{\text{CO}_2,1640} + \text{AOD}_{\text{CH}_4,1640} + \text{AOD}_{\text{PWV},1640} = 0.0087 \cdot (P/P_o) + 0.0047 \cdot (P/P_o) + (0.0014 \cdot \text{PWV} - 0.0003)$$

In order to consider the impact of CO<sub>2</sub> and CH<sub>4</sub> on the 1640 band, the solar absorption spectrum in this spectral region has been simulated for different atmospheric CO<sub>2</sub> and CH<sub>4</sub> contents at Izaña Observatory (IZO) using the line-by-line radiative transfer model PROFFWD (Hase et al., 2004). Only the CO<sub>2</sub> and CH<sub>4</sub> concentration vertical profiles have been varied among simulations for typical

measurement conditions at IZO, considering the Whole Atmosphere Community Climate Model (WACCM)-v7 climatological values as the reference (<https://www.cesm.ucar.edu/>). All remaining model inputs have been kept identical, and the effect of aerosols and clouds have not been taken into account in the simulations.

Figure 1 presents a summary of the test results, illustrating the ratio between the simulated solar absorption spectrum using the WACCM-v7 CO<sub>2</sub> and CH<sub>4</sub> vertical concentration profiles as the baseline, and the simulated spectra for incremental increases of 5%, 10%, 25%, and 50% in the CO<sub>2</sub> profile (upper panel), 5%, 10%, 25%, and 50% in the CH<sub>4</sub> profile (middle panel), and 5%, 10%, 25%, and 50% in both the CO<sub>2</sub> and CH<sub>4</sub> profiles (lower panel). The subplots on the left depict the mean ratio within the spectral bands as a function of the concentration increments. Note that EM27/SUN AOD is estimated from the averaged solar absorption spectra in the micro-windows defined in Table 1 of the manuscript.

The most pronounced interference arises from CH<sub>4</sub>, resulting in a variation of 0.40% for the most extreme scenario (a 50% increase relative to the climatological value) at the center of the absorption line. This variation translates to a mere 0.20% mean ratio difference across the entire 1640 band. When considering the combined influence of CO<sub>2</sub> and CH<sub>4</sub> (lower panel of Figure 1), the intra-band variation remains limited to 0.25% for a 50% increase in CO<sub>2</sub> and CH<sub>4</sub> vertical concentration profiles. This estimation is a quite conservative estimation of the difference between utilizing a climatological approach like AERONET and utilizing actual gas concentration observations at the site.

Comparing the observed intra-band variations in the simulations with the intra-band coefficient of variation (CV) values derived from the measured solar absorption spectra within the 1640 band (0.67%, as depicted in Table 1 of the manuscript), we can deduce that the impact of CO<sub>2</sub> and CH<sub>4</sub> on AOD estimations is expected to be insignificant. Thus, the AERONET approach proves adequately precise for acquiring dependable AOD values. This assertion finds support in the uncertainties assessed through the Monte-Carlo method within this spectral band, as well as the minimal mean differences, standard deviations, and root-mean-squared errors observed in the CE318-AERONET and EM27/SUN comparison detailed in Section 5.4.

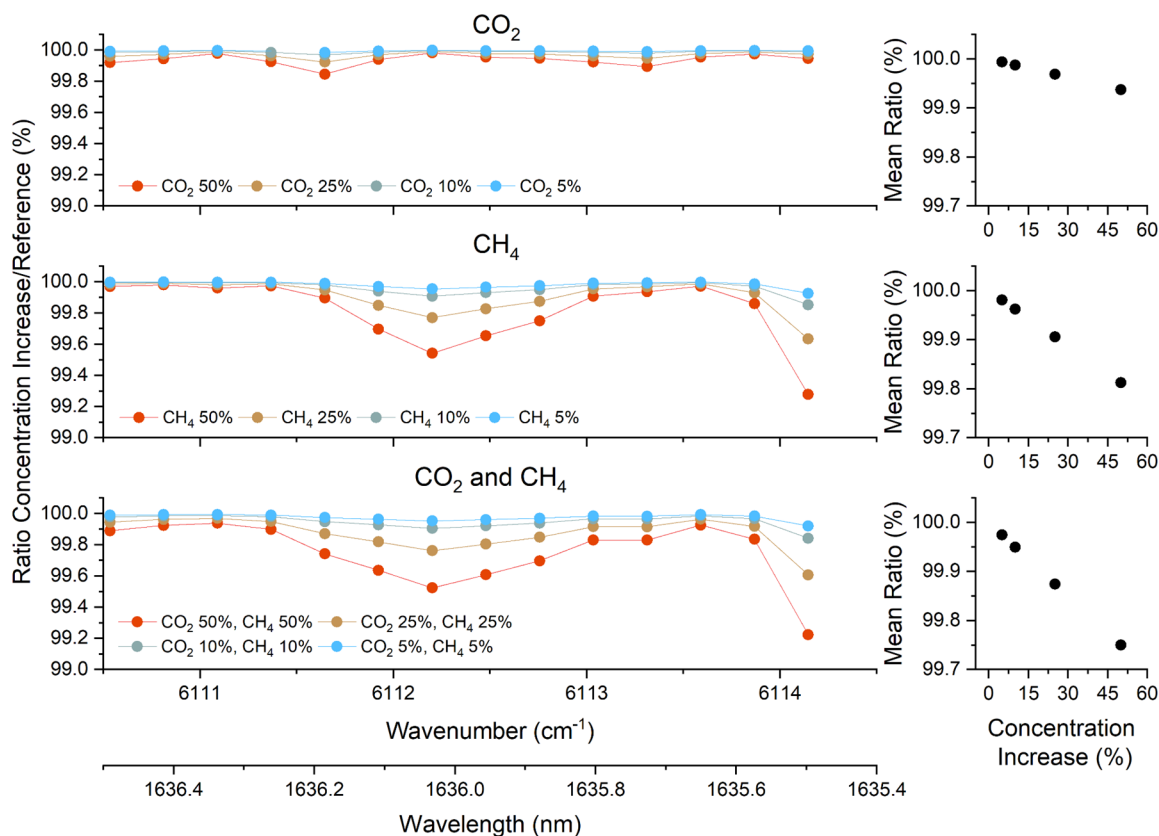


Figure 1. Ratio between the simulated solar absorption spectrum in the 1635.5-1636.5 nm spectral region, considering the WACCM-v7 CO<sub>2</sub> and CH<sub>4</sub> concentration vertical profiles as the reference, and the simulated ones for increases of 5%, 10%, 25% and 50% of the CO<sub>2</sub> profile (upper panel), of 5%, 10%, 25% and 50% of the CH<sub>4</sub> profile (middle panel), and of 5%, 10%, 25% and 50% of the CO<sub>2</sub> and CH<sub>4</sub> profiles (lower panel). Sub-plots on the left show the intra-band mean ratio as a function of the different concentration increases.

#### References:

Barreto, Á.; García, O.E.; Schneider, M.; García, R.D.; Hase, F.; Sepúlveda, E.; Almansa, A.F.; Cuevas, E.; Blumenstock, T. Spectral Aerosol Optical Depth Retrievals by Ground-Based Fourier Transform Infrared Spectrometry. *Remote Sens.* 2020, 12, 3148

Giles, D.M.; Sinyuk, A.; Sorokin, M.G.; Schafer, J.S.; Smirnov, A.; Slutsker, I.; Eck, T.F.; Holben, B.N.; Lewis, J.R.; Campbell, J.R.; et al. Advancements in the Aerosol Robotic Network (AERONET) Version 3 database—Automated near-real-time quality control algorithm with improved cloud screening for Sun photometer aerosol optical depth (AOD) measurements. *Atmos. Meas. Tech.* 2019, 12, 169–209.

Smirnov, A.; Holben, B.; Lyapustin, A.; Slutsker, I.; Eck, T. AERONET processing algorithms refinement. In Proceedings of the AERONET Workshop, El Arenosillo, Spain, 10–14 May 2004.

### 3. Finally, is the purpose of the paper the investigation of using FTIRs for AOD retrieval? Would that help increasing AOD networks or would it have an added value for COCCON?

**Authors:** As mentioned in the manuscript, EM27/SUN observations are expected to expand the monitoring of atmospheric composition to new stations (COCCON) and to the NIR and SWIR spectral ranges. This extension is expected to improve our understanding of atmospheric processes due to its ability to simultaneously retrieve column-integrated aerosol and trace gas information.

In the conclusion section, it is stated that “*This portable instrument is highly versatile and can be deployed at numerous stations worldwide to meet specific measurement needs. Therefore, it has the potential to serve as a crucial tool for densifying current ground-based networks for observing aerosols and gasses, as well as for validating satellite-based gas and aerosol products.*”

**Specific Comments:**

**1. Lines 24-25: “...the most recent assessment report by the Intergovernmental Panel on Climate Change (IPCC)” – If possible, please provide a reference in order to support this statement.**

**Authors:** The reference to Forster et al. (2021) is already included at the end of this specific sentence.

**2. Line 31: “SI-traceable measurement technique” (the acronym SI is not defined).**

**Authors:** The acronym SI has been defined in the manuscript as follows:

“To further advance our understanding of atmospheric aerosols, WMO considers the development of new, reliable, and **International System of Units (SI)**-traceable measurement techniques, and non-conventional measurement methods with open availability of validation data, as a core activity (WMO, 2010, 21017).”

**3. Lines 50-51: If possible, please add a brief comment introducing the near-infrared (NIR) and short-wave infrared (SWIR) spectral regions at this point.**

**Authors:** We agree with the referee and therefore we have included the following information to the manuscript:

“The near-infrared (NIR) and short-wave infrared (SWIR) spectral regions are portions of the electromagnetic spectrum that extend beyond the visible light range. NIR refers to wavelengths between approximately 700 to 900 nanometers, while SWIR refers to wavelengths between approximately 900 to 2500 nanometers. These regions have unique properties that make them valuable for various applications, including remote sensing, spectroscopy, and imaging.”

**4. Line 84: The comment in the parenthesis “an IFS 125HR”, could be replaced by “an IFS 125HR spectrometer”.**

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

**5. Lines 131-132: Is there any further information about the uncertainty of the instrument or the performance according to the last calibrations available?**

**Authors:** According to the COCCON protocols (Frey et al., 2019; Alberti et al., 2022), to harmonize the retrieved species when using any COCCON spectrometer, empirical instrument-specific calibration factors for XCO<sub>2</sub>, XCO, XCH<sub>4</sub> and XH<sub>2</sub>O are calculated from the side-by-side

solar measurements with the COCCON reference spectrometer (SN37). The instruments are set up on the seventh floor at the Meteorology and Climate Research – Atmospheric Trace Gases and Remote Sensing (IMK-ASF) building located at KIT Campus North (49°050 38.7 00 N, 8°260 11.5 00 E, 134 m a.s.l.). The correction factors are calculated by comparing a defined gas retrieved with any EM27/SUN instrument with the reference instrument; a linear fit forced to zero intercept is performed, and then the slope is taken as its value.

The empirical calibration factors for XCO<sub>2</sub>, XCH<sub>4</sub>, XCO and XH<sub>2</sub>O for the IZO COCCON spectrometer (SN85) are presented in Alberti et al., (2022) and correspond to values lower than 0.999 for XCO<sub>2</sub>, XCH<sub>4</sub>, and XH<sub>2</sub>O, and lower than 0.980 for XCO (see Figure 24 or Table S2 in the Supplement of Alberti et al., 2022). In addition, the IZO COCCON instrument is continuously compared with the IZO TCCON IFS 125HR instrument, corroborating the calibration factors computed with respect to the COCCON reference (Figure 2).

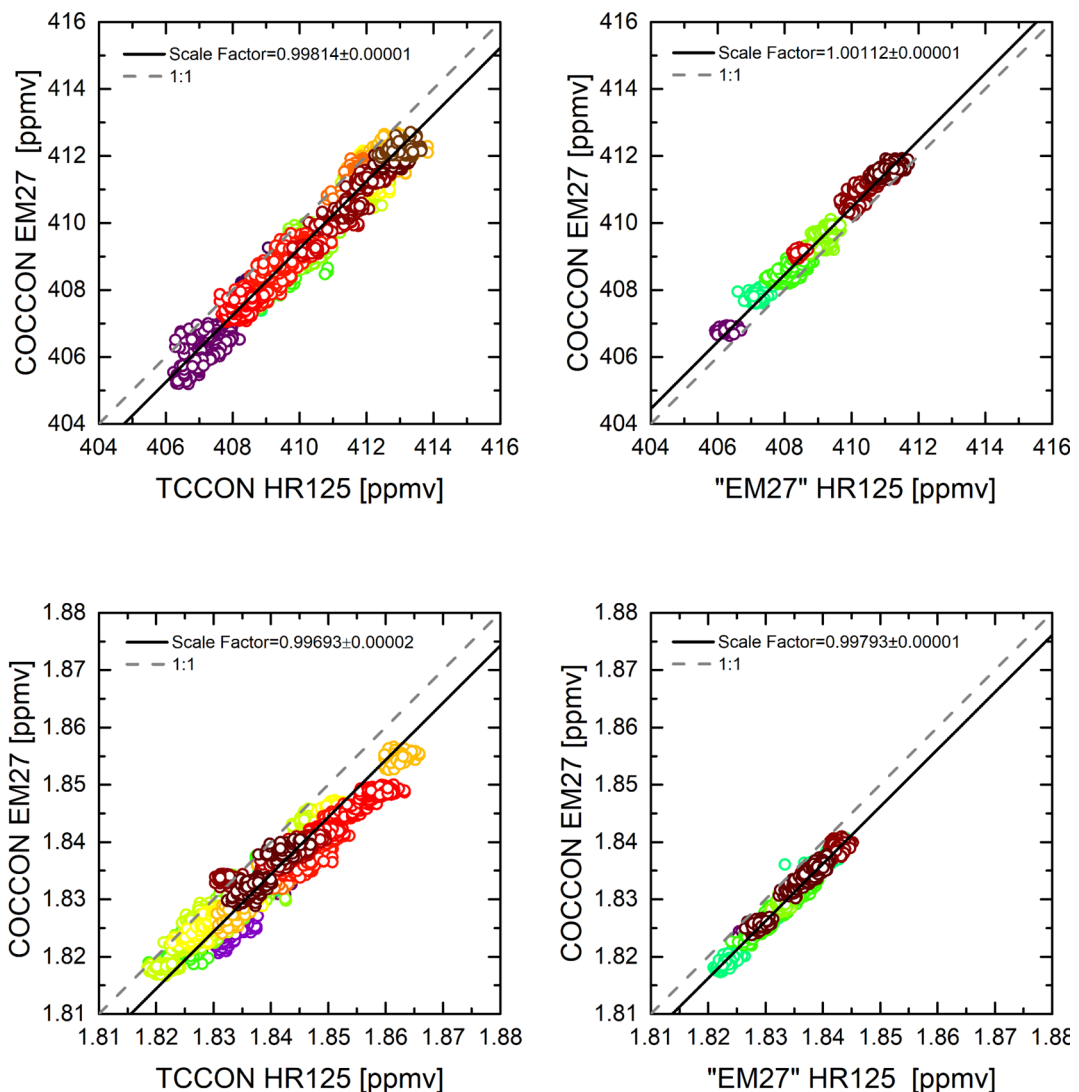


Figure 2. Comparison between the COCCON EM27/SUN and high-resolution (HR) IFS 125 retrievals for CO<sub>2</sub> and CH<sub>4</sub> at the Izaña Observatory. The HR IFS 125 observations are taken using the TCCON (TCCON HR12) and COCCON EM27/SUN measurement settings (EM27 HR125).

## References:

Alberti, C., Hase, F., Frey, M., Dubravica, D., Blumenstock, T., Dehn, A., Castracane, P., Surawicz, G., Harig, R., Baier, B. C., Bès, C., Bi, J., Boesch, H., Butz, A., Cai, Z., Chen, J., Crowell, S. M., Deutscher, N. M., Ene, D., Franklin, J. E., García, O., Griffith, D., Grouiez, B., Grutter, M., Hamdouni, A., Houweling, S., Humpage, N., Jacobs, N., Jeong, S., Joly, L., Jones, N. B., Jouglet, D., Kivi, R., Kleinschek, R., Lopez, M., Medeiros, D. J., Morino, I., Mostafavipak, N., Müller, A., Ohyama, H., Palmer, P. I., Pathakoti, M., Pollard, D. F., Raffalski, U., Ramonet, M., Ramsay, R., Sha, M. K., Shiomi, K., Simpson, W., Stremme, W., Sun, Y., Tanimoto, H., Té, Y., Tsidu, G. M., Velazco, V. A., Vogel, F., Watanabe, M., Wei, C., Wunch, D., Yamasoe, M., Zhang, L., and Orphal, J.: Improved calibration procedures for the EM27/SUN spectrometers of the COllaborative Carbon Column Observing Network (COCCON), *Atmos. Meas. Tech.*, 15, 2433–2463, <https://doi.org/10.5194/amt-15-2433-2022>, 2022.

Frey, M., Sha, M. K., Hase, F., Kiel, M., Blumenstock, T., Harig, R., Surawicz, G., Deutscher, N. M., Shiomi, K., Franklin, J. E., Bösch, H., Chen, J., Grutter, M., Ohyama, H., Sun, Y., Butz, A., Mengistu Tsidu, G., Ene, D., Wunch, D., Cao, Z., Garcia, O., Ramonet, M., Vogel, F., and Orphal, J.: Building the COllaborative Carbon Column Observing Network (COCCON): long-term stability and ensemble performance of the EM27/SUN Fourier transform spectrometer, *Atmos. Meas. Tech.*, 12, 1513–1530, <https://doi.org/10.5194/amt-12-1513-2019>, 2019

### **6. Figure 1 (page 6): The exact spectral range that each detector is sensitive in (InGaAs-1 and InGaAs-2), could be introduced in the instrument’s technical description in Section 1.**

**Authors:** The definition of spectral range covered by each detector has been included in Section 1 as follows:

“This instrument, based on a RockSolid™ pendulum interferometer, acquires solar absorption spectra in the near-infrared region from 4000 to 11500  $\text{cm}^{-1}$  with a spectral resolution of 0.5  $\text{cm}^{-1}$  (maximum optical path difference, OPD<sub>max</sub>, of 1.8 cm), using a CaF<sub>2</sub> beamsplitter and two InGaAs photodetectors. **The primary detector covers the spectral section between 5500 and 11000  $\text{cm}^{-1}$ , while the secondary detector covers the 4000–5500  $\text{cm}^{-1}$  region (Hase et al., 2016) (hereafter referred to as InGaAs-1 and InGaAs-2, respectively)**”.

### **7. Figure 1 (page 6): “Note that both detectors have different gains ...the measured radiation.” This part could be included in the main passage instead.**

**Authors:** This statement has been included in the main text of Section 4.1 following the referee’s suggestion. This is the sentence included in the text (in line 169):

“Note that both detectors have different gains (greater for InGaAs-2, i.e., B6-B8 micro-windows in SWIR), therefore the observed spectral behaviour is not the one expected for the solar radiance: the higher the wavelength, the lower the measured radiation.”

### **8. Lines 166-167: The explanation of the selection of these specific wavelengths was essential and well placed here by the authors. However, the sentence “In this study, an additional channel (B1) ...” might firstly give the impression that another channel (apart from the 8 already mentioned) was added. To avoid confusion, this sentence could be rephrased**

as: “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.”

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

**9. The names of the two detectors are introduced in lines 168-169: “hereafter referred to as InGaAs-1 and InGaAs-2, respectively”, however, they have already been mentioned as “InGaAs-1” and “InGaAs-2” in Figure 1. A suggestion would be to move Figure 1 below this paragraph.**

**Authors:** To avoid confusion, the name of the two detectors have been defined in the EM27/SUN technical description given in Section 1 (see 6th question).

**10. Lines 170-171: “in this study the EM27/SUN solar spectra were neither calibrated nor referenced to any traceable lamp”. In section 3.1 it is mentioned that proper calibration of all COCCON spectrometers is performed.**

**Authors:** The calibration of the COCCON spectrometer is performed in terms of the standard retrieved species (i.e., XCO<sub>2</sub>, XCO, XCH<sub>4</sub> and XH<sub>2</sub>O) by using side-by-side solar measurements with the COCCON reference spectrometer (SN37) (see comment 5). As stated by Barreto et al. (2020), the Langley-Plot calibration of Fourier Transform spectrometer (FTS) is only necessary for detecting atmospheric constituents with broadband signatures (e.g., atmospheric aerosols or water continuum) for measuring lunar absorption spectra or atmospheric emissions. For standard trace gas retrievals, such calibration is dispensable, since high-resolution solar absorption spectra are self-calibrating in the sense that the absorption signature is referenced to the surrounding continuum. This is a relevant advantage of ground-based FTS systems for atmospheric trace gas monitoring provided that the instrument is optically well-aligned and well-characterized.

To make it clearer in the revised manuscript, the following statement has been modified in Section 3.1. as follows:

“This guarantees strict common methods for ensuring the quality of measurements (evaluation of the optical alignment and instrumental line shape), proper calibration of all COCCON spectrometers with respect to the TCCON site Karlsruhe and the COCCON reference EM27/SUN spectrometer operated permanently at KIT (**in terms of the standard retrieved species**), and adherence to the COCCON data analysis scheme ensures the generation of precise and accurate data products.”

**Table 2 (page 14): could be moved after the end of the paragraph (line 303).**

**Authors:** Table 2 has been placed at the end of the paragraph following the referee’s suggestion.

**11. Equation 2 (page 14): It was mentioned previously that “m” is the air mass, please define if “ma” stands for a different parameter.**

**Authors:** The term “m” in Eq. 2, as defined by WMO, stands for optical air mass. In our study we have calculated the U95 traceability limit using the equation of Kasten and Young (1989). This information will be included in the manuscript.

**12. Figure 5 (page 15): It is a bit puzzling why during the first period (2020 – 2021) the uncertainty is higher than the second one. Is it a matter of calibration?**

**Authors:** The period showing the highest AOD differences, as depicted in Figure 5, is the 2019-2020 timeframe. Initially, this behaviour was attributed to inadequately conditioned smoothing spline functions resulting from the absence of calibration during the 6-month period. Although this explanation seemed plausible, subsequent detection and rectification of a computational error have revealed only minimal discrepancies in AOD differences during this particular time span. These findings are presented in the revised figures and tables. Corresponding adjustments have been made to the text and tables accordingly.

All figures have undergone correction, with the exception of Figure 1 and the former Figure 7, now Figure 8. The good agreement observed between the AOD retrieved from CE318-AERONET and EM27/SUN in the new Figure 6 (former Figure 5, placed below) over this specific period, despite the gap of 6 months in the calibration due to COVID restrictions, has ensured the robustness of our calibration approach.

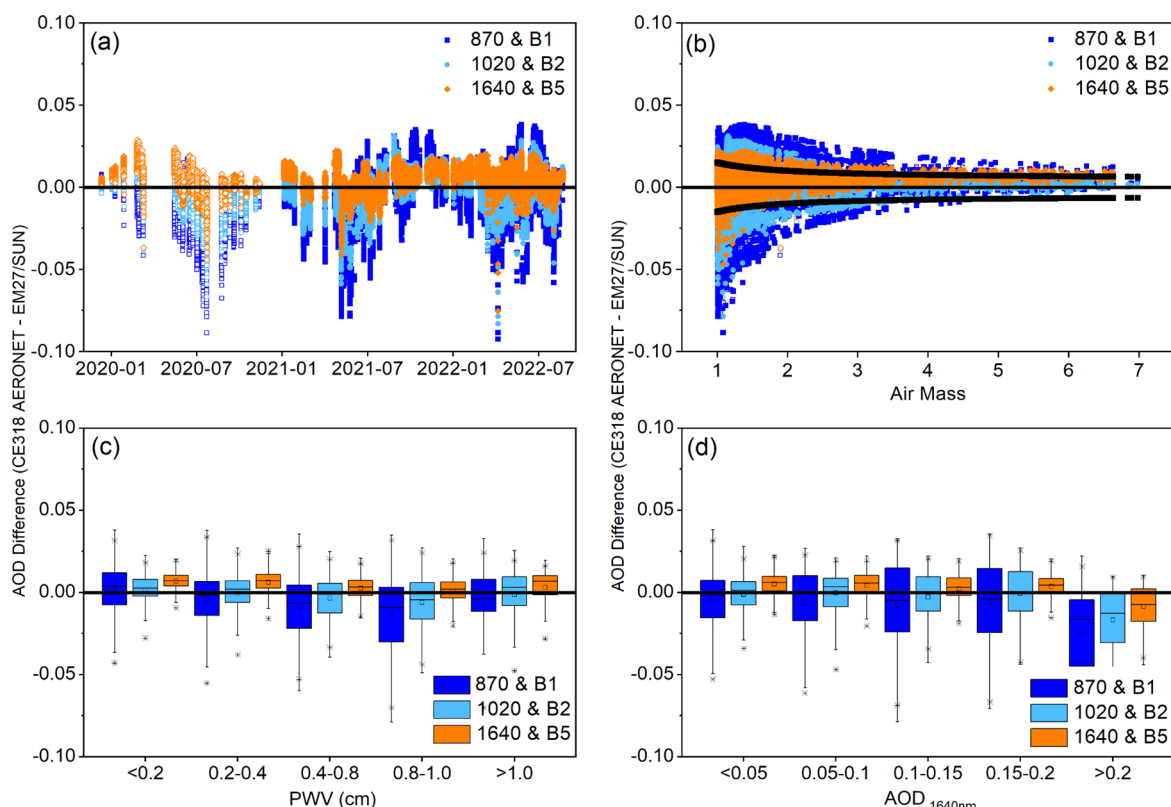


Figure 6: AOD comparison between CE318-AERONET and EM27/SUN (2021-2022) in the three coincident spectral bands for CE318-AERONET (870, 1020, and 1640 nm) and EM27/SUN (B1, B2, and B5) according to (a) time, (b) optical air mass, (c) PWV (cm) and (d) AOD at 1640 nm from CE318-AERONET (AOD<sub>1640nm</sub>). Open circles in (a) and (b) correspond to the September 2019 - December 2020 period. The solid curves in (b) represent the U95 uncertainty limit. The AOD differences in (c) and (d) are displayed as box plots, where the lower and upper boundaries for each box are



the 25th and 75th percentiles, the solid line is the median value, the hyphens are the maximum and minimum values and the asterisks indicate the 1th and 99th percentiles. The number of cases in each box was 3191, 3729, 3690, 773 and 238 for PWV <0.2, 0.2-0.4, 0.4-0.8, 0.8-1.0 and >0.1 cm, respectively. In the case of AOD differences with respect to CE318-AERONET AOD, the number of cases was 9757, 929, 542, 364 and 29 for AOD\_1640nm <0.05, 0.05-0.1, 0.1-0.15, 0.15-0.2 and >0.2, respectively.

**13. Figure 6 (page 16): I would recommend to be presented in Section 5.4, probably after line 346.**

**Authors:** Figure 6 has been located at the end of Section 5.4 following the referee's suggestion.

**14. Figure 8 (page 20): the numbering of each cell is in the same position (bottom right) except for 8f & 8g which are on the bottom left side.**

**Authors:** The numbering of each cell has been located in the same position for all subplots following the referee's suggestion.

**15. Figure 8 caption is not inside the page borders.**

**Authors:** The figure caption has been adapted to fit in the page borders following the referee's suggestion.

**16. Author contributions: It is suggested to change O.G. to O.E.G. (Omaira E. Garcia) and S.L. to S.F.L.L. (Sergio F. Leon-Luis).**

**Authors:** The authors' initials have been modified in the manuscript following the referee's suggestion.

**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 acronym 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  $1.61\% \text{month}^{-1}$  (B8) and  $1.75\% \text{month}^{-1}$  (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<sup>-1</sup>" 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<sup>-1</sup>. 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<sup>-1</sup> (i.e. OPD of 45 cm, reference of TCCON network) and truncated a posteriori at 0.5 cm<sup>-1</sup> 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<sup>-1</sup>. 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<sup>-1</sup> (0.5 cm<sup>-1</sup>). 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<sup>-1</sup>"**

**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) | $\Delta\lambda$ (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  $\sim 1.3^\circ$  (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  $V_\lambda = V_{0,\lambda} * d^{-2} * \exp(-m * \tau_\lambda)$  -> Since  $V_{0,\lambda}$  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  $V_{0,\lambda}$ , since  $V_{0,\lambda} = V_{sun,\lambda} * d^{-2}$  (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  $V_{0,\lambda}$  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 too 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 ( $\ln(I)$  vs 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 ( $V_{0,\lambda}$ )**

**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 interpret the graphic.

**Authors:** The authors agree with this comment. A Figure 4 has been replaced by this figure:

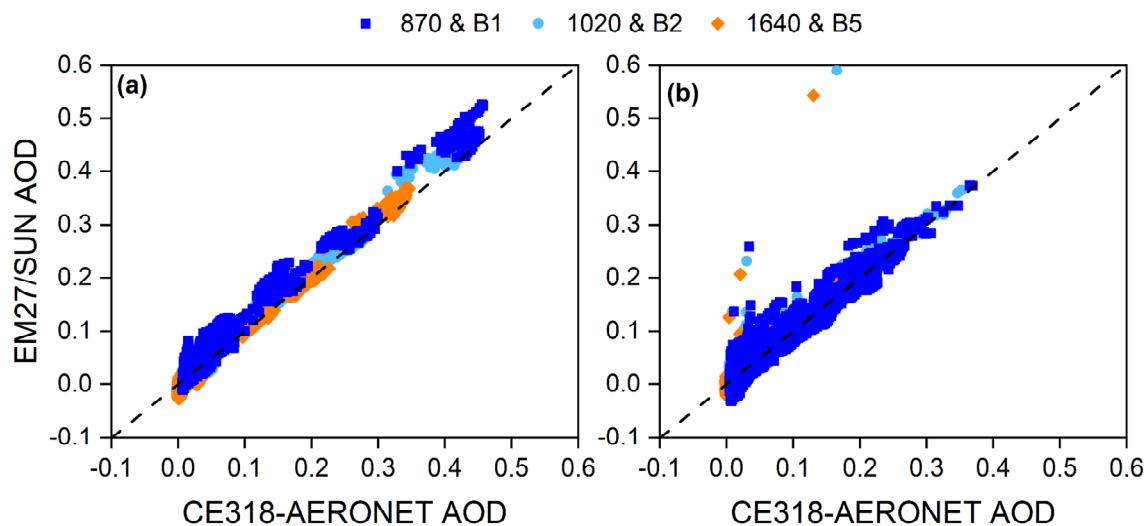


Figure 4. Scatterplot for the coincident EM27/SUN-AERONET AOD values from (a) December 2019 to December 2020 and (b) January 2021 to September 2022 considering the EM27/SUN B1 (870 nm), B2 (1020 nm) and B5 (1640 nm) micro-windows. The number of coincidences is 14575 and 2863 in the period January 2021 - 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 recommend 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  $\text{cm}^{-1}$ . *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 occurrence 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<sub>95</sub> 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<sub>95</sub> 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 Angstrom 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<sub>2</sub>O 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<sup>-1</sup> (B8) and 1.75%month<sup>-1</sup> (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.

## (2) Author's changes in the manuscript

Please, find below the following changes performed by the authors different than those made as part of the review process by reviewers 1 and 2.

### Spelling and grammar

We have corrected the following spellings: analyze, centered, colour and dashed-coloured (caption Fig. 1), and U95.

sulfur: lines 373, caption Fig. 7.

line 159: as a conservative estimation -> a conservative estimation.

line 209: New information has been added about the equation to retrieve the AOD from Eq. 1. This information will be necessary for the new section 5.3 with the uncertainty estimation.

line 245: "backward arrows" has been replaced by "black arrow".

line 267: added the meaning for arrow (1): "It should be noted that there was a lack of calibration during the 6-month period from March to September 2020 (**black arrow (1)**), as mentioned in Section 5.1".

line 359: artefact -> artifact

line 231: "constant" has been replaced by "stable".

line 239: The word "day" has been removed in this sentence.

line 242: "some improvements" in this sentence seems to be a mistake. This sentence has been corrected as follows:

"Temporal degradation and continuous drift of the EM27/SUN calibration coefficients are observed in Figures 2(a)-(b)."

line 316: "for air mass equals to 1" is replaced by "for air mass equals 1".

line 437: "despite of observing" has been replaced by "despite observing".

The name given in the text to the Cimel AERONET instrument has been homogenized to "CE318-AERONET".

### Other changes

Competing interest have been included since one of the (co-)authors is a member of the editorial board of Atmospheric Measurement Techniques/Atmospheric Chemistry and Physics. Updated reference has been introduced for Stremme et al. (2023). Three other changes have been made to the text:

1. The authors have ensured that the term "calibration" is not confused in the manuscript with the standard COCCON protocols for XGAS calibration.

Line 12: "This indicates that the low-resolution COCCON instruments are suitable for detecting the aerosol broadband signal contained in the IR spectra in addition to the retrieval of precise trace gas concentrations provided a robust calibration procedure (**Langley-based or absolute calibration procedures**) is used to compensate for the optical degradation of the external system (~0.72% per month)".

Line 246, caption Fig. 2, line 255, line 290, line 461: "**Langley-Plot** calibration".

Line 454: "**It is important to note that this Langley-Plot calibration is dispensable since high-resolution solar absorption spectra are self-calibrating in the sense that the absorption signature is referenced to the surrounding continuum**".

2. A calculation error has been identified in the script used to compute the smoothing splines, which has led to an erroneous calculation applied to the splines during the 2019-2020 period. This error has resulted in the negative AOD values retrieved by the EM27/SUN during the spring and summer of 2020, as well as the poorer agreement observed between CE318-AERONET and EM27/SUN for this specific period. Initially, this behaviour was attributed to the inadequately conditioned smoothing spline functions arising from the absence of calibration during the 6-month period. While this explanation appeared plausible, subsequent detection and rectification of the computational error have revealed minimal discrepancies in AOD differences, as presented in the revised figures and tables. All figures have undergone correction, with the exception of Figure 1 and the former Figure 7, now Figure 8. Corresponding adjustments have been made to the text and tables accordingly. The good agreement observed between the AOD retrieved from CE318-AERONET and EM27/SUN over this specific period, despite the gap of 6 months in the calibration due to COVID restrictions, has ensured the robustness of our calibration approach.

3. The authors have eliminated the dependence of the Earth-Sun distance in the EM27/SUN measurements, as initially outlined in Equation 1 and noted on line 203. This consideration is unnecessary in the context of EM27/SUN measurements due to the instrument's reduced Field of View (FOV), which is smaller than the solar disk's dimensions. 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 it 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. To express this differently, brightness level on a detector generated by an extended source overfilling the FOV does not change with the distance to the source (see Figure 2 in Blanc et al., 2014).

The impact of eliminating the distance factor in our dataset has been demonstrated to be negligible in terms of the computed AOD, owing to the application of a "quasi-continuous" Langley calibration approach. This approach involves utilizing every single day of measurements an

estimated  $V_{0,\lambda}$  for each specific day from its Langley (in a Langley day) or from the smoothing spline functions derived from the 31 Langleys performed over the entire period (in a non-Langley day). Consequently, the Earth-Sun distance does not exhibit significant variability between consecutive EM27/SUN measurements (a total of 56190 solar measurements spanning the 3-year duration, corresponding to 187 days).

The distance effect on the AOD retrieval using our calibration procedure has been estimated according to the following table, showing the AOD differences ( $\Delta AOD$ ) computed between the AOD without and with the distance correction:

| $\Delta AOD$ | 870 nm | 1020 nm | 1640 nm |
|--------------|--------|---------|---------|
| 2019-2022    | 0.003  | 0.002   | 0.002   |

Examining this table, we can deduce that the differences attributed to the distance term in the AOD retrieval with our "quasi-continuous" Langley calibration approach can be deemed negligible. This effect is approximately within the range of  $\pm 3\%$  in terms of  $V_0$ , as evidenced in the changes observed in the new Figure 2.

#### 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.