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_2) and methane (CH_4) are parameterized based on the present atmospheric pressure at the station (P) and at sea level (Po), following the methodologies outlined by Smirnov et al. (2004) and Giles et al. (2019), as detailed below:

$$AOD_{g,1060}=AOD_{CO2,1640}+AOD_{CH4,1640}+AOD_{PWV,1640}=0.0087\cdot(P/Po)+0.0047\cdot(P/Po)+(0.0014\cdot PWV-0.0003)$$

In order to consider the impact of CO_2 and CH_4 on the 1640 band, the solar absorption spectrum in this spectral region has been simulated for different atmospheric CO_2 and CH_4 contents at Izaña Observatory (IZO) using the line-by-line radiative transfer model PROFFWD (Hase et al., 2004). Only the CO_2 and CH_4 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_2 and CH_4 vertical concentration profiles as the baseline, and the simulated spectra for incremental increases of 5%, 10%, 25%, and 50% in the CO_2 profile (upper panel), 5%, 10%, 25%, and 50% in the CH_4 profile (middle panel), and 5%, 10%, 25%, and 50% in both the CO_2 and CH_4 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 CH4, 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_2 and CH_4 (lower panel of Figure 1), the intra-band variation remains limited to 0.25% for a 50% increase in CO_2 and CH_4 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_2 and CH_4 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.

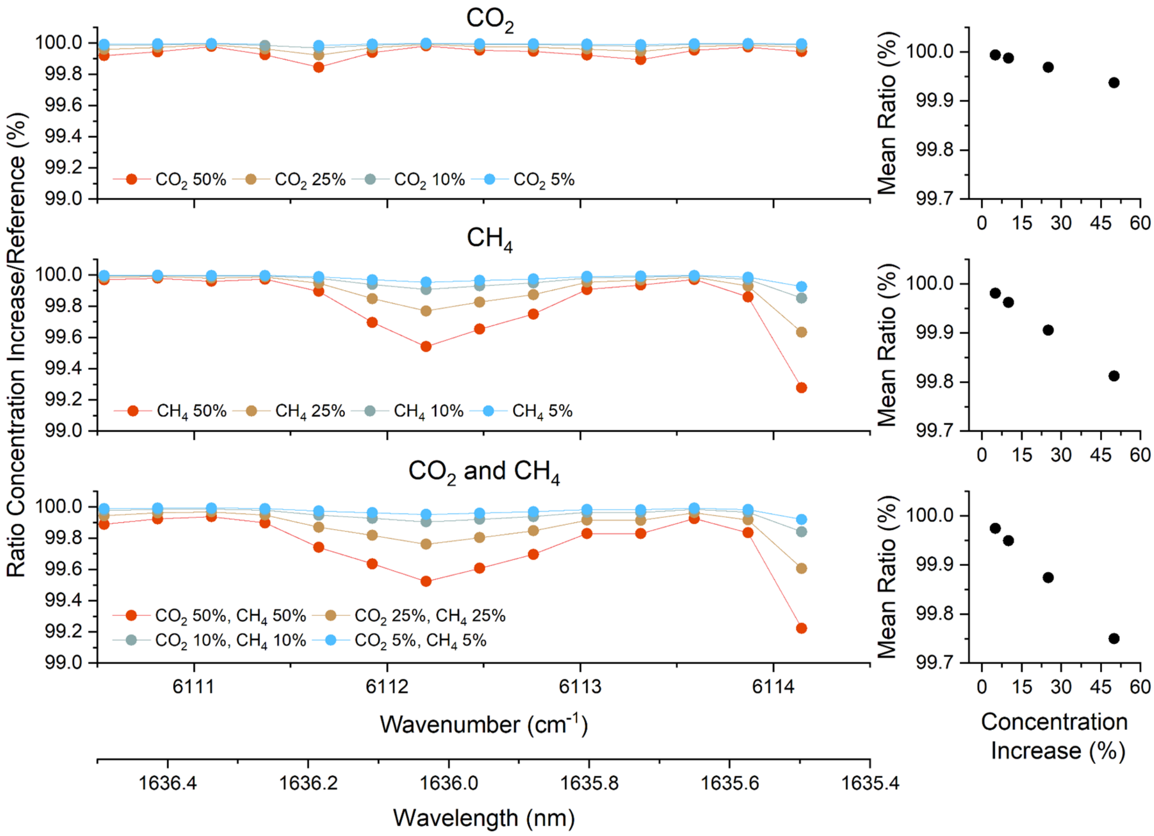


Figure 1. Ratio between the simulated solar absorption spectrum in the 1635.5-1636.5 nm spectral region, considering the WACCM-v7 CO_2 and CH_4 concentration vertical profiles as the reference, and the simulated ones for increases of 5%, 10%, 25% and 50% of the CO_2 profile (upper panel), of 5%, 10%, 25% and 50% of the CH_4 profile (middle panel), and of 5%, 10%, 25% and 50% of the CO_2 and CH_4 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."

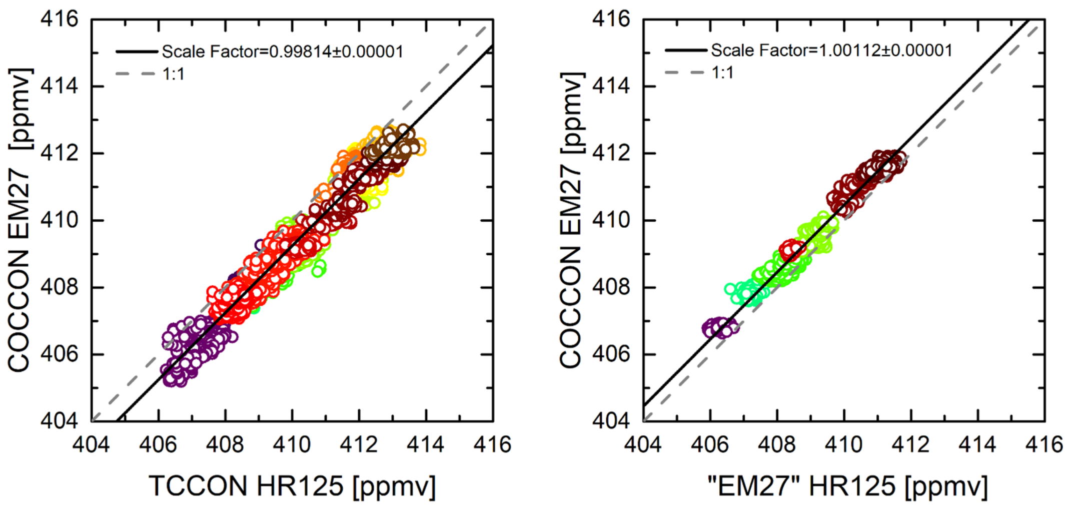
4. **Line 84: The comment in the parenthesis "an IFS 125HR", could be replaced by "an IFS 125HR spectrometer".**

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 XCO2, XCO, XCH4 and XH2O 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 empirical calibration factors for XCO2, XCH4, XCO and XH2O for the IZO COCCON spectrometer (SN85) are presented in Alberti et al., (2022) and correspond to values lower than 0.999 for XCO2, XCH4, and XH2O, 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).



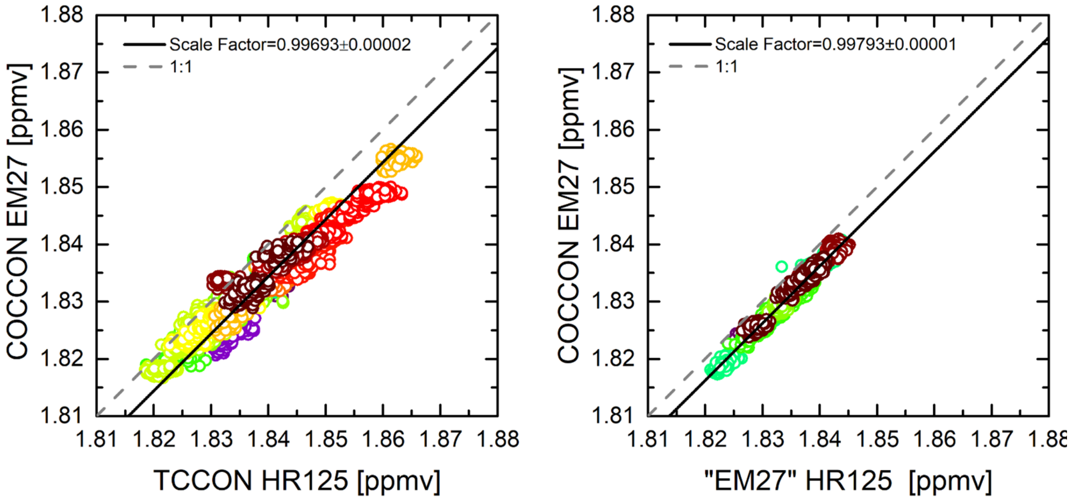


Figure 2. Comparison between the COCCON EM27/SUN and high-resolution (HR) IFS 125 retrievals for CO2 and CH4 at the Izaña Observatory. The HR IFS 125 observations are taken using the TCCON (TCCON HR12) and COCCON EM27/SUN measurement settings (EM27 HR125).

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 RockSolidTM pendulum interferometer, acquires solar absorption spectra in the near-infrared region from 4000 to 11500 cm−1 with a spectral resolution of 0.5 cm−1 (maximum optical path difference, OPDmax, of 1.8 cm), using a CaF2 beamsplitter and two InGaAs photodetectors. **The primary detector covers the**

spectral section between 5500 and 11000 cm−1, while the secondary detector covers the 4000–5500 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., XCO2, XCO, XCH4 and XH2O) 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.

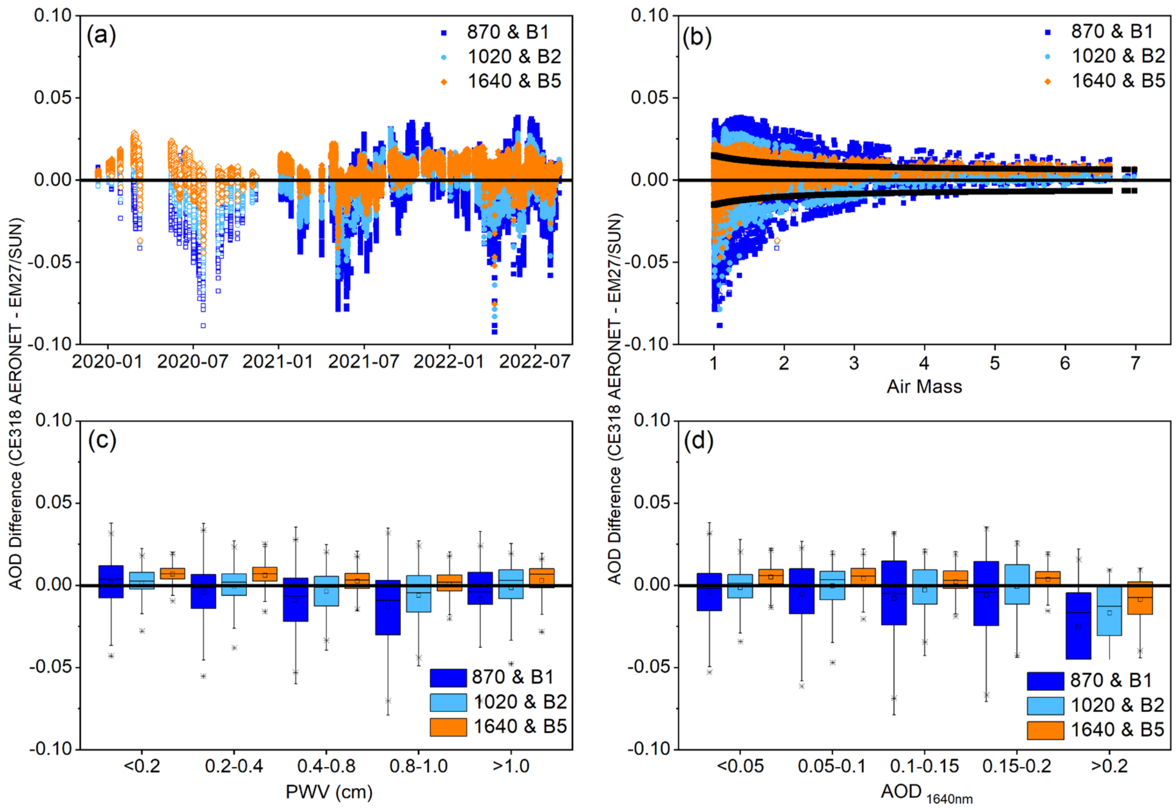


Figure 6: AOD comparison between CE318-AERONET and EM27/SUN (2021-2022) in the three coincident spectral bands for CE318-AERONET (870, 1020, and 1640 nm) and EM27/SUN (B1, B2, and B5) according to (a) time, (b) optical air mass, (c) PWV (cm) and (d) AOD at 1640 nm from CE318-AERONET (AOD_1640nm). Open 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 boarders.

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).

<u>Authors:</u> The authors' initials have been modified in the manuscript following the referee's suggestion.