

Response to Referee #1

We would like to thank the reviewer for carefully reading the manuscript and for providing helpful comments, remarks and suggestions. You can find below our responses in red after each individual comment:

Review of the manuscript "Ground-based total ozone column measurements in the Huggins and Chappuis bands using Direct-Sun DOAS observations" by Karagkiozidis et al.

The manuscript describes measurements of total column ozone using a DOAS system from direct solar irradiance measurements in Thessaloniki, Greece. The retrieved ozone values are validated by comparison to two collocated instruments, a Brewer spectrophotometer and a Pandora system. The retrievals are performed both in the UV and VIS spectral regions and show consistent results with the collocated reference instruments, with the exception of the VIS retrievals during high aerosol contamination.

The manuscript is well written, the structure is clear and the results and conclusions follow from the discussions.

The figures are mostly informative and useful for the understanding of the arguments. Possibly the need for the SCD figures along with the corresponding TOC scatter plots are somewhat redundant, but I see the point of showing that the magnitude of ozone absorption has no systematic impact on the retrieval.

As indicated by the reviewer, the main purpose of presenting both the SCD comparisons and the corresponding TOC scatter plots is to demonstrate that the magnitude of ozone absorption does not introduce any systematic bias in the TOC retrievals. Although there is some redundancy in the information provided, the combined presentation allows for a clearer and more comprehensive assessment of the consistency between the UV and VIS retrievals across the full range of ozone absorption conditions. In particular, the SCD comparisons illustrate the agreement with the reference instruments at slant path geometry, while the TOC scatter plots confirm that this agreement is preserved after conversion to vertical columns. For these reasons, we believe that retaining both representations is necessary.

A point that needs to be clarified is the concept of "I0-correction" which is used without definition on line 266 and Table 1. While it may be familiar to the DOAS community, it is not a common term to the wider community.

We thank the reviewer for pointing this out. We agree that the terms "I0-correction" and "intensity offset", although commonly used within the DOAS community, may not be familiar to the broader community. Sect. 3.2 has therefore been revised to include a clear explanation of both terms and their physical meaning in the DOAS retrieval. We now explain that the I0 effect originates from inconsistencies between laboratory absorption cross sections measured with a smooth light source and the structured atmospheric absorption spectra recorded with lower spectral resolution, while the intensity offset compensates for potential stray light or dark signal that is not effectively removed from the measured spectrum.

The paper could also highlight the advantage of the TOC retrieval in the Chappuis band of not being sensitive to the stratospheric temperature in contrast to the Huggins band retrievals, which are an issue for TOC retrievals in the UV by some instruments (e.g.

Dobson and Pandora, see for example publications by Gröbner et al., 2021 amt-14-3319-2021 and Xiaoyi et al., 2016 amt-9-5747-2016).

We thank the reviewer for this helpful comment. The Introduction section has been revised to include a clearer discussion of the reduced sensitivity of TOC retrievals in the Chappuis bands to the effective ozone temperature. We now explain that ozone absorption cross sections in the UV exhibit a strong temperature dependence, which can introduce systematic uncertainties in TOC retrievals if the effective ozone temperature is not accurately represented. In contrast, ozone absorption in the Chappuis bands is only weakly temperature-dependent, making VIS-based TOC retrievals inherently less sensitive to temperature-related uncertainties.

In that respect, in Section 3.2 where the ozone layer temperature is discussed, I wonder how the tropospheric contribution of ozone could impact the retrieval due to its significantly different temperature than the stratospheric component?

We thank the reviewer for the comment. The influence of tropospheric ozone on the retrieval is accounted for by using in the retrieval methodology ozone cross sections at two temperatures, one for the stratosphere (223 K) and one for the troposphere (243 K) as used and suggested in previous studies (e.g., Van Roozendael et al., 2006; Wang et al., 2018). This has now been clarified in Sect. 3.2 (first paragraph). Tropospheric ozone can influence the TOC retrieval through variations in the ozone vertical profile, which affect the AMF calculation via changes of h_{eff} (see equation 5). The mean annual variability of h_{eff} is shown in Figure 3 and this variability is mainly caused by changes in the ozone profile during the year. The estimated effect on AMF is within $\pm 0.1\%$ for SZAs less than 70° and up to 0.8% at larger SZAs, as discussed in the last sentence of the 3rd paragraph of Sect. 3.3.

Van Roozendael, M., Loyola, D., Spurr, R., Balis, D., Lambert, J. -C., Livschitz, Y., Valks, P., Ruppert, T., Kenter, P., Fayt, C., and Zehner, C.: Ten years of GOME/ERS-2 total ozone data—The new GOME data processor (GDP) version 4: 1. Algorithm description, *J. Geophys. Res. Atmospheres*, 111, 2005JD006375, <https://doi.org/10.1029/2005JD006375>, 2006.

Wang, Y., Puķīte, J., Wagner, T., Donner, S., Beirle, S., Hilboll, A., Vrekoussis, M., Richter, A., Apituley, A., Piters, A., Allaart, M., Eskes, H., Frumau, A., Van Roozendael, M., Lampel, J., Platt, U., Schmitt, S., Swart, D., and Vonk, J.: Vertical Profiles of Tropospheric Ozone From MAX-DOAS Measurements During the CINDI-2 Campaign: Part 1—Development of a New Retrieval Algorithm, *J. Geophys. Res. Atmospheres*, 123, <https://doi.org/10.1029/2018JD028647>, 2018.

In section 3.2, two methods are discussed for the TOC retrieval. As briefly mentioned in the conclusion, one could also attempt a third method which would consist in using a reference top of the atmospheric reference solar spectrum, and retrieve the TOC from calibrated spectral measurements, as in Egli et al., 2022, amt-15-1917-2022. The advantage of this method would be that the reference spectrum obviously does not contain any residual ozone, and the method does not rely on zero-airmass extrapolations which require exceptionally stable conditions to produce reliable results, usually only found at high altitude, low latitude stations.

We thank the reviewer for this comment. Sect. 3.4 of the manuscript has been revised accordingly to further elaborate on the possibility of using an extraterrestrial reference

solar spectrum at the top of the atmosphere, which is free of atmospheric absorption, instead of a measured FRS, for TOC retrieval. We now clarify that this approach avoids the presence of residual ozone absorption in the reference spectrum and eliminates the need for Langley extrapolation methods, but that is also sensitive to the spectral resolution of the used solar spectrum which must be convoluted with the slit function of the measured spectra to achieve spectral matching. This discussion has been included to highlight this method as a potential alternative, however, it is not implemented in the present study, which focuses on retrievals based on measured reference spectra.

In section 4.4 on the AOD impact on the ozone retrieval in the VIS, AMF is used as a possible influencing factor. I am not sure if that argument is valid, since the AOD is predominantly in the low troposphere, where there is no ozone, so any path enhancement due to aerosol scattering would only have an effect due to the tropospheric ozone in that layer. Did the authors consider that?

We thank the reviewer for this comment. We agree that aerosols are predominantly located in the lower troposphere, where ozone concentrations are much lower than in the stratosphere, and therefore any aerosol-induced enhancement of the photon path would mainly affect the absorption signal of tropospheric ozone. As a result, the impact of aerosol scattering on TOC through a change of the AMF is expected to be relatively small, however not negligible. We have further elaborated on this topic in Sect. 4.4 of the revised manuscript. Under high aerosol load conditions, aerosol forward scattering, which is more pronounced in the visible spectral range, can modify the radiative transfer and hence the effective optical path length. In addition, part of the observed bias may arise from spectral fitting artefacts under enhanced aerosol loading. If not fully accounted for in the retrieval, these effects can propagate into systematic deviations in the retrieved TOC. A quantitative assessment of this effect would require dedicated RTM simulations, but such analysis is beyond the scope of this study. The discussion of Sect. 4.4 has been revised accordingly.

Response to Referee #2

We would like to thank the reviewer for carefully reading the manuscript and for providing helpful comments, remarks and suggestions. You can find below our responses in red after each individual comment:

Manuscript: *Ground-based total ozone column measurements in the Huggins and Chappuis bands using Direct-Sun DOAS observations* **ID:** egosphere-2025-5627

This manuscript presents direct-sun DOAS total ozone column (TOC) retrievals in both the UV (Huggins) and visible (Chappuis) spectral regions using the Delta UV-VIS DOAS system in Thessaloniki. The work is relevant for the AMT community, particularly as it explores the feasibility and performance of visible-band direct-sun ozone retrievals, which remain less established than traditional UV techniques. The manuscript is generally well written, and the reported agreement with collocated Brewer and Pandora observations is good. The study has the potential to make a valuable contribution to atmospheric measurement techniques, especially if methodological choices, calibration assumptions, and statistical aspects are documented more transparently. The comments below are therefore mainly requests for clarification, additional documentation, and improved transparency rather than indications of major methodological errors.

For transparency, I wish to inform the editor and authors that I am actively involved in ozone calibration and intercomparison activities related to Brewer. I believe this background is relevant to the comments provided.

In Section 3.4, the manuscript applies the Bootstrap Estimation (BE) method to derive the ozone reference slant column, representing a novel extension of an approach that has primarily been developed and applied for NO₂ retrievals (e.g. Cede et al., 2006; Herman et al., 2009). Given the fundamentally different vertical distribution, variability, and tropospheric contribution of total ozone compared to NO₂, this application would benefit from more explicit justification and validation. At present, BE results are shown mainly for the VIS channel (Fig. 4b), while the corresponding UV results are not presented (line 349), and no sensitivity analysis is provided regarding the choice of percentile or AMF binning. Including the UV BE results and a brief sensitivity assessment would strengthen confidence in the robustness of the BE approach when applied to ozone.

We thank the reviewer for this important comment. Sect. 3.4 has been revised to clarify both the methodology and the terminology used for the determination of the O₃ SCD of the FRS. We would like to clarify that the term "Bootstrap Estimation (BE)" was not used accurately in the original manuscript. The analysis in this study employs the Minimum-Amount Langley-Extrapolation (MLE) method (Herman et al., 2009), rather than a bootstrap estimation approach. The manuscript has been revised accordingly to use the correct terminology and to more clearly describe the implementation of this method.

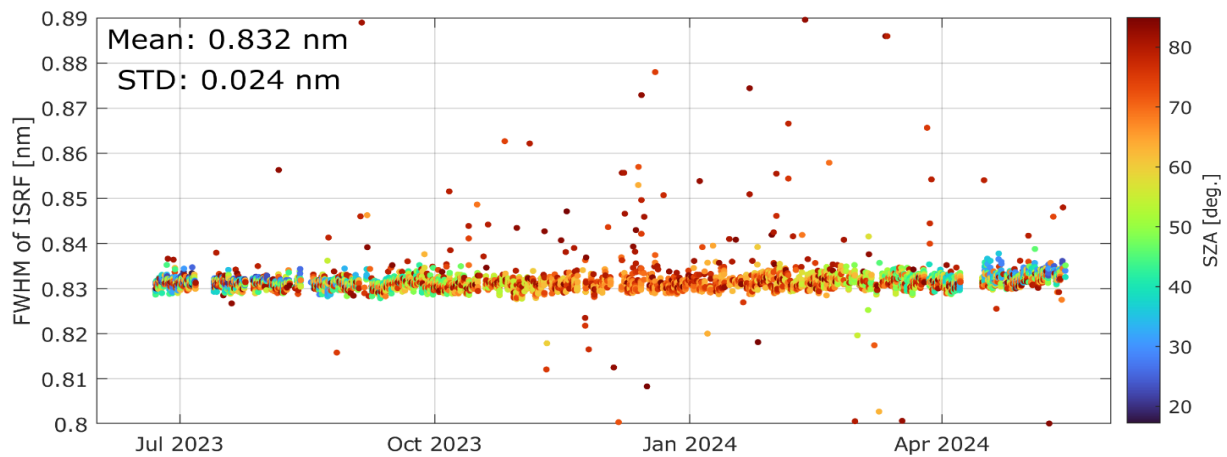
Sect. 3.4 has further been revised to clarify that MLE relies on the assumption that, if TOC was constant throughout all days in the period of study, dSCDs would exhibit a linear dependence on AMF (Eq. 3), and the extrapolated dSCD at AMF=0 would equal the SCD of the FRS. If the minimum vertical column amount of an atmospheric species is constant over part of the dataset, then this will be independent of AMF. Over Thessaloniki, the lowest TOC values typically occur during the summer and autumn months, hence cover the entire range of the AMF annual variability. Given that a sufficient number of

measurements is available for each AMF bin during these periods, the minimum dSCDs are expected to be similar across bins, thereby satisfying the assumption of the MLE method. A sensitivity analysis was performed to assess the robustness of the MLE method with respect to the choice of percentile. Percentiles ranging from the 2nd to the 5th, in steps of one, were tested. The resulting differences in the estimated SCD of the FRS were found to be up to 0.6% in the VIS and up to 2% in the UV. These differences reflect the statistical nature of the MLE method, which requires a sufficiently large number of measurements to yield stable results. The larger variability observed in the UV retrievals is attributed to the substantially lower number of available data in this spectral range (see Sect. 2.2). Additionally, care must be taken in the selection of the AMF bin width as overly broad AMF bins reduce the number of points used for the extrapolation, while excessively narrow bins may contain too few measurements for a reliable estimation of the selected percentile. UV data have now been included in Figure 4 as panel c. When the same MLE methodology is applied to data in the UV range retrievals, a comparable value of SCD_FRS=408.54 DU is obtained. However, the UV dataset covers a shorter time period and does not include the summer and autumn months, when TOC typically reaches its annual minimum. Furthermore, at larger SZAs the RMS filtering that is applied (see Sect. 4) removes a substantial number of measurements. As a result, the statistical assumptions of the MLE method are less optimally satisfied for the UV, which leads to a larger sensitivity to the choice of the percentile and the AMF range. In this case, to ensure statistical robustness, the AMF range was restricted to values below 4. Despite these limitations, the close agreement between the UV- and VIS-derived values of SCD_FRS indicates that the calibration of the reference spectrum is robust.

It is difficult to follow the different dataset lengths of the UV and VIS channels described in Sections 2 and 3. The manuscript would benefit from a summary table indicating the operating periods of each channel together with the main instrumental modifications. While the authors assume that the instrument remained stable over the analysis period, no supporting evidence is provided. Although it is stated that lamp spectra are used for accurate wavelength calibration (line 140), no results are shown demonstrating regular monitoring or otherwise documenting the long-term stability of the spectrograph.

We thank the reviewer for this comment. The manuscript has been revised to clarify the operating periods of the VIS and UV channels and to better document the stability of the instrument over the analysis period. We now explicitly state at the end of Sect. 2.2 that the VIS dataset spans from June 2023 to May 2024 (~11 months), while the UV dataset covers the period from January 2024 to May 2024 (~6 months). This clarification helps the reader to more easily follow the temporal coverage of the two datasets.

In addition, a description of the instrument stability monitoring has been added in Sect. 2.2. The stability of the instrument's slit function was regularly monitored by comparison of measured spectra with a high-resolution solar spectrum, convoluted with a Gaussian function (Fayt and Van Roozendaal, 2001; Aliwell et al., 2002; Danckaert et al., 2017). As shown in the figure below, the slit function remained stable throughout the study period, exhibiting a mean FWHM of approximately 0.83 nm (± 0.02 nm). This aligns closely within the 0.85 nm value derived from mercury lamp measurements. Most of the outliers are found at high SZAs, where reduced signal-to-noise ratios affect the slit function retrieval accuracy. The corresponding discussion has been added to Sect. 2.2 of the revised manuscript, while the figure itself was not included.



Although EuBrewNet is cited in Section 2, EuBrewNet ozone data are not used in the analysis presented in Section 4. Including EuBrewNet data—particularly the Version 2 total ozone product—would be a valuable addition, as Version 2 (Rimmer et al 2018) employs updated ozone absorption cross sections consistent with those used in the Pandora and Delta retrievals and applies a similar treatment of effective ozone height and effective temperature. In addition, it should be clarified whether the Brewer data used in the study include the straylight correction introduced during the 2021 calibration (WMO 2024, GAW Report No. 301, p. 43) and implemented on EuBrewNet processing.

We thank the reviewer for this thoughtful suggestion. We agree that the use of EuBrewNet Version 2 TOC product would potentially provide results more consistent with those of Pandora and Delta, as it employs updated ozone absorption cross sections and applies a similar treatment of effective ozone height and effective temperature. As shown in Figure 11 of the manuscript, the largest differences between Delta and Brewer occur at high SZAs ($> \sim 75^\circ$), where stray light effects become increasingly important, especially for the Brewer. Under these conditions, for our single-monochromator Brewer, stray light leads to an underestimation of TOC, as documented in the RBCC-E campaign reports. The positive drift observed in the Delta-Brewer differences at large SZAs is consistent with this known behavior. This indicates that the Brewer measurements are more strongly affected by stray light than the Delta retrievals, even though the measured spectra of Delta are not directly corrected for it. However, version 2 of the EuBrewNet TOC product excludes values for $AMF > 3$. This limits the AMF range available for comparison with Delta, which, as previously discussed, is important for our assessment. Nevertheless, we did compare Delta with the version 2 EuBrewNet dataset, and the statistics are practically the same, with a change in the slope from 0.98 to 0.97 and an increase in the mean difference from -0.08% to -0.23% . Therefore, we decided to keep our in-house processing rather than the EuBrewNet Version 2 data.

It is noted that no stray light correction was applied to the Brewer data used in this study. The stray light correction introduced during the 2021 calibration campaign was calculated as a power function of the O_3 SCD and for our instrument it was derived for SCD values below ~ 1400 DU. At larger SCDs (corresponding typically to larger AMFs), extrapolation of the power fit correction introduces additional uncertainties leading to over-correction for $SZA > 75^\circ$. Therefore, to avoid introducing additional assumptions, no stray light correction was applied to the in-house Brewer TOC processing.

The manuscript refers to Pandora “version 1.8.49” in Section 2; however, this designation corresponds to the processor version rather than to a specific ozone product definition.

For clarity and reproducibility, it would be helpful to explicitly state which Pandora ozone product is used (e.g. OUT2) and to briefly summarize the key retrieval assumptions relevant for comparison, such as the spectral window, ozone cross sections, and treatment of effective ozone temperature, with reference to the appropriate PGN documentation. The calibration of Pandora instrument is not commented.

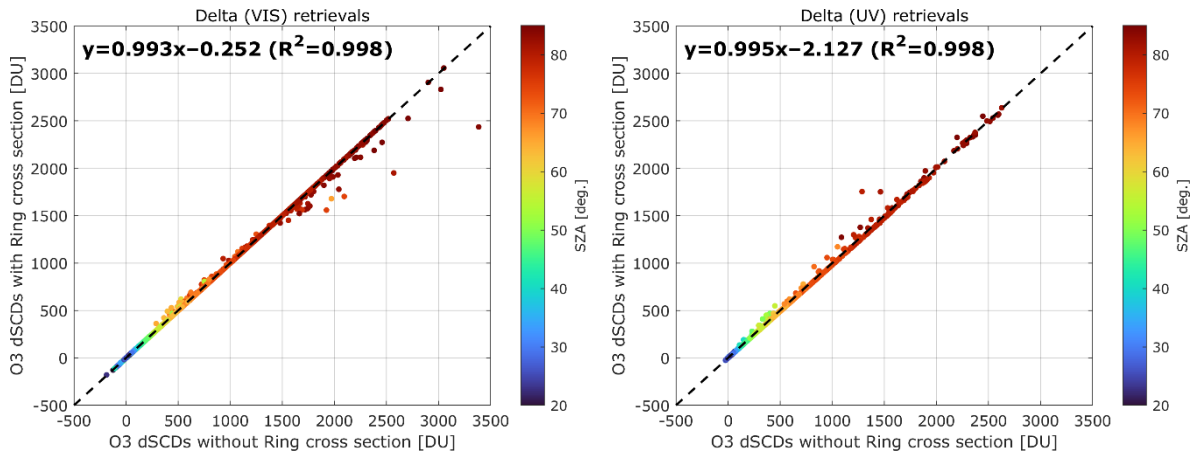
We thank the reviewer for this helpful comment. We agree that "1.8.49" refers to the BlickP processor version rather than to a specific ozone product definition. Sect. 2.4 has been revised to specify that the PGN TOC product used in this study corresponds to retrieval version "rout2p1-8", which is the current operational PGN TOC product. In addition, we now briefly summarize the key retrieval settings, including the spectral fitting window, the use of a high-resolution extraterrestrial reference spectrum, the O₃ absorption cross sections used in the analysis and the treatment of effective O₃ temperature. We also provide a reference to the corresponding PGN Data Products Readme document for additional retrieval details. Furthermore, the discussion of Sect. 2.4 has been slightly revised to clarify that the calibration procedures applied within the PGN framework are described in the official PGN manuals.

The UV channel inherits the absolute calibration from the VIS channel through the use of a common reference slant column, as described in Section 3.4. While this approach is acceptable, the manuscript would benefit from explicitly stating this assumption and from demonstrating that UV-based calibration estimates are consistent with the VIS-derived value, which is not shown in the current version. In this context, providing Figure 4c (UV Bootstrap Estimation), analogous to Fig. 4b, would significantly improve the work.

We thank the reviewer for this important comment. This point has been addressed in the response above (see first comment). Sect. 3.4 has been revised to clarify the calibration approach, and the corresponding UV results are now included in Figure 4c, demonstrating the consistency with the VIS-derived data.

The manuscript states in Section 3.3 that the Ring effect is not included in the DOAS fitting because direct-sun spectra are used. While this assumption is generally reasonable, a brief justification or sensitivity assessment would improve clarity, particularly in light of the discussion of thin cloud contamination and potential stray light effects in Section 4. Clarifying whether inclusion of a Ring pseudo-cross section has a negligible impact under the observed measurement conditions would strengthen the methodological description.

We thank the reviewer for this comment. To assess the potential impact of the Ring effect on the Delta retrievals, we performed a sensitivity test in which a Ring pseudo-cross section was included in the DOAS analysis. The comparison of O₃ dSCDs retrieved with and without the inclusion of the Ring cross section is shown in the figures below for the VIS (left) and UV (right). The data are colored by SZA.

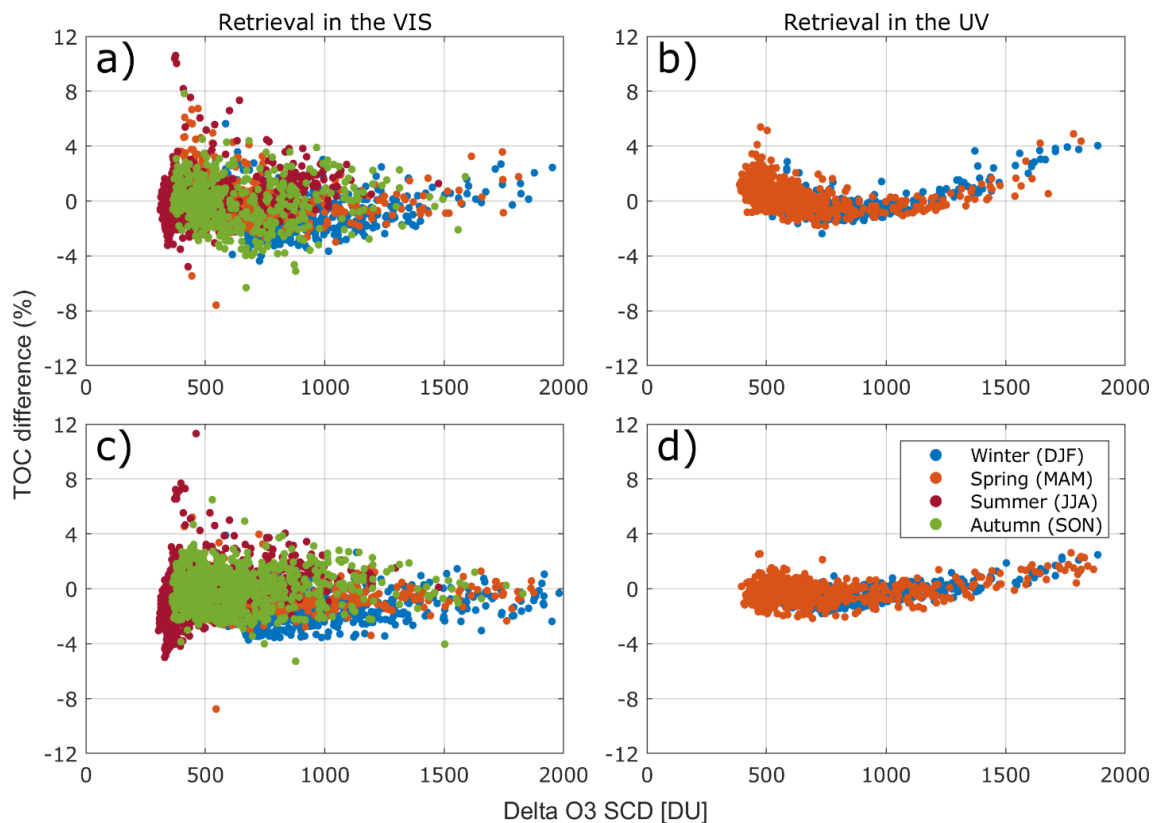


- For the VIS retrievals, the inclusion of the Ring cross section has a negligible effect on the retrieved O3 dSCDs, with a median difference of approximately -0.32% , confirming that the Ring effect has a limited impact under direct-sun measurement conditions. Most outliers are found at high SZAs.
- For the UV retrievals, the effect is slightly more pronounced, as expected due to the stronger Ring contribution at shorter wavelengths, but it remains small, overall, with a median difference of approximately -1.5% .

Nevertheless, the main focus of this study is on the VIS retrievals. We also note that the Brewer and Pandora instruments used in this study do not include a Ring correction in their operational TOC retrievals. For consistency with the reference datasets used in the intercomparison, we therefore chose not to include a Ring pseudo-cross section in the Delta retrievals.

For completeness, it may be helpful to further explore potential stray light effects by presenting the differences between Delta and the reference instruments as a function of the ozone slant column (SCD), used here as an indicator of high absorption conditions. Such a representation could provide additional insight into whether residual stray light effects contribute to the observed differences, particularly under high SZA or high aerosol conditions, and would complement the existing comparison plots shown in Figures 6–9.

We thank the reviewer for this helpful suggestion. We investigated the dependence of the differences between Delta and the reference instruments as a function of the ozone SCD, which can be used as an indicator of high absorption conditions. The corresponding analysis is provided in the figure below and is consistent with Figure 11 of the manuscript, except that the differences are shown as a function of SCD rather than SZA. Overall, the SCD-based representation yields results similar to those obtained as a function of SZA and essentially provides the same information. However, the SCD-based representation focuses on higher absorption conditions, while most of the observations correspond to typically lower SZAs (and hence lower SCDs). For clarity and to avoid redundancy, we prefer to show the SZA-based representation in the manuscript, as it provides a more direct interpretation of geometry-related effects.



The manuscript compares Delta with Brewer and Pandora separately in Section 4. Including a short Brewer–Pandora comparison, or referencing established agreement at the site, would provide useful context for interpreting the Delta validation and for assessing the consistency of the reference instruments themselves.

We thank the reviewer for this helpful suggestion. A brief comparison of collocated Brewer and Pandora TOC measurements has now been included in the revised Supplement (as we prefer to keep the focus of the paper to the Delta instrument) and a corresponding discussion has been added at the end of Sect. 4.2. The results show a high level of consistency between the two reference instruments at Thessaloniki, with a mean difference of less than 0.4% and a narrow distribution, with most differences confined within approximately $\pm 1\%$.

Throughout Section 4, intercomparison results are presented without reporting the number of collocated measurements (N), which is essential for assessing the statistical significance and robustness of the reported correlations and biases. Correlation coefficients and RMS values are given for several comparisons (e.g. Delta–Brewer, Delta–Pandora, UV–VIS) without specifying N, and qualitative statements about higher or lower numbers of collocations are not supported by quantitative values. Reporting N in the text and figure captions, and where appropriate providing confidence intervals for key metrics, would substantially improve transparency and interpretability.

We agree that showing and discussing the number of collocated measurements is essential for assessing the statistical significance and robustness of the reported correlations and biases and the number (N) was already included in the plots below the linear fit equation. All figure captions have been revised to explicitly state that N corresponds to the number of collocated measurements. Additionally, the number of collocations was already provided

in Sect. 4.2, but has now been further clarified, including a quantitative comparison of the number of VIS and UV collocations.

In relation to the calibration methodology described in Section 3.4 and the comparison results presented in Section 4, it would be useful to include uncertainty information for the Delta TOC retrievals. A concise discussion or table summarizing the main sources of uncertainty—such as DOAS fitting residuals, determination of the reference slant column (LE and BE), AMF/effective height assumptions, and sensitivity to atmospheric conditions (e.g. aerosols or thin clouds)—together with indicative uncertainty ranges, would improve the quantitative interpretation of the results shown in Figures 6–9.

We thank the reviewer for this suggestion. An error budget analysis has been included in the revised manuscript in a new section (3.5), where the main sources of uncertainty affecting the Delta TOC retrievals in both the UV and VIS ranges are discussed, along with a summary table.

Table 1 summarizes the DOAS fit settings used in the analysis; however, several key terms are not sufficiently defined in the manuscript. In particular, the meaning and implementation of the I_0 correction, the intensity offset term, and whether the absorption cross sections are orthogonalized are not clearly explained. Providing brief clarifications of these elements—either in the table caption or in the accompanying text—would improve transparency and reproducibility.

Sect. 3.2 has been revised to include a clear explanation of the “ I_0 -correction” and “intensity offset” terms, as well as their physical meaning in the DOAS retrieval. We now explain that the I_0 effect originates from inconsistencies between laboratory absorption cross sections measured with a smooth light source and the structured atmospheric absorption spectra recorded with lower spectral resolution, while the intensity offset compensates for potential stray light or dark signal that is not effectively removed from the measured spectrum. The use of orthogonalized absorption cross sections was already specified in Table 1.

Figure 4 would benefit from clearer labeling and explanation. It is not immediately evident whether the data shown correspond to the VIS or UV channel, particularly in panel (b). In addition, panel (a) refers to a specific clear-sky day used for the Langley analysis, but this day is not explicitly identified in the figure or clearly linked to the data shown in panel (b). The caption should explicitly state the spectral channel shown in each panel and identify the specific day used for the Langley extrapolation.

We thank the reviewer for this helpful comment. Figure 4 has been revised to improve clarity and help interpretation. In panel b, the measurements corresponding to the clear-sky day used for the Langley extrapolation in panel a are now explicitly highlighted. In addition, text has been included in the lower right corner of each panel to clearly indicate the dataset that is used (VIS or UV).

Minor points:

the use of MLS–GMI climatology for the effective height in lines 305–320 could be briefly justified, as Gröbner et al. (2021) uses local ozonesonde profiles.

Sect. 3.3 has been revised to clarify the use of the MLS–GMI climatology for the effective height. This approach was adopted because routine ozone soundings are not available at Thessaloniki, and therefore local ozonesonde profile measurements cannot be used.

Figure 7, please clarify how the Brewer slant column is calculated (e.g. $SCD = TOC \times AMF$, and which AMF is used);

Sect. 4.2 has been revised to clarify that for the Brewer and Pandora measurements, the AMFs provided in their respective operational products are used. The calculation of the SCD using the formula $SCD = TOC \times AMF$ (Eq. 2) is described at the beginning of Sect 4.2.

Figure 9 appears largely redundant with Figure 8 and could potentially be merged;

Even though Figs. 8 and 9 show very similar results, they present comparisons with the two independent reference instruments (Brewer and Pandora). Presenting these comparisons separately allows for a clearer evaluation of the Delta retrieval performance against each reference dataset. Since the intercomparison with both instruments constitutes a core result of this study, we believe that retaining the figures separately improves the clarity of the presentation.

Figure 10; Include UV data, is not shown on the paper and can illustrate the different periods used on the paper.

We thank the reviewer for this suggestion. Figure 10 has been revised to include the UV TOC retrievals in addition to the VIS data. This addition allows for a clearer illustration of the different time periods covered by the UV and VIS measurements and improves the overall completeness of the dataset presentation. The corresponding discussion in Sect. 4.2 has been updated accordingly.

line 182 (WMO 2024), please confirm whether updated stray light corrections reported in recent Brewer calibration reports were applied to the Brewer 005 data used in this study.

Please see our response to the comment on the usage of EuBrewNet data.

Response to Referee #3

We would like to thank the reviewer for carefully reading the manuscript and for providing helpful comments, remarks and suggestions. You can find below our responses in red after each individual comment:

Review of the manuscript: "Ground-based total ozone column measurements in the Huggins and Chappuis bands using Direct-Sun DOAS observations" by Karagiozidis et al.

This manuscript presents total ozone column (TOC) retrievals from a ground-based direct-sun DOAS instrument operating in Thessaloniki, Greece. Ozone columns are retrieved independently in the Huggins (UV) and Chappuis (VIS) absorption bands and are evaluated against co-located Brewer and Pandora observations. The study demonstrates very good agreement between the DOAS-derived TOC and the reference instruments and discusses the influence of aerosol loading on the VIS retrievals.

Overall, the paper addresses a relevant topic within the scope of AMT and related Copernicus journals. The extension of direct-sun DOAS TOC retrievals into the Chappuis band is of practical interest, particularly under conditions where UV-based retrievals are limited. The manuscript is generally well structured, scientifically sound, and clearly written.

I recommend publication after minor revisions, mainly to clarify methodological aspects and to strengthen the physical interpretation of some results.

Minor Comments:

Temperature effects and vertical ozone distribution

The treatment of ozone effective temperature in the UV retrievals is briefly discussed, but the potential influence of vertically inhomogeneous ozone distributions (e.g. enhanced tropospheric or UTLS ozone) is not fully addressed. Since tropospheric ozone resides at significantly higher temperatures than the stratospheric ozone maximum, it would be useful to comment on how such conditions could affect the UV retrievals and whether they could contribute to residual biases between instruments.

We thank the reviewer for this comment. The variability of the ozone profile can affect the TOC retrieval via the AMF calculation, specifically by affecting the effective height h_{eff} (see equation 5). The mean annual variability of h_{eff} is shown in Figure 3 and this variability is mainly caused by changes in the ozone profile during the year. The estimated effect on AMF is within $\pm 0.1\%$ for SZAs less than 70° and up to 0.8% at larger SZAs, as discussed in the last sentence of the 3rd paragraph of Sect. 3.3.

The effect of the ozone profile on the retrieval due to temperature dependent absorption cross sections is accounted for by using in the retrieval methodology cross sections at two temperatures, one for the stratosphere (223 K) and one for the troposphere (243 K) as used and suggested in previous studies (e.g., Van Roozendaal et al., 2006; Wang et al., 2018). This has now been clarified in Sect. 3.2 (first paragraph).

Van Roozendael, M., Loyola, D., Spurr, R., Balis, D., Lambert, J. -C., Livschitz, Y., Valks, P., Ruppert, T., Kenter, P., Fayt, C., and Zehner, C.: Ten years of GOME/ERS-2 total ozone data—The new GOME data processor (GDP) version 4: 1. Algorithm description, *J. Geophys. Res. Atmospheres*, 111, 2005JD006375, <https://doi.org/10.1029/2005JD006375>, 2006.

Wang, Y., Puķite, J., Wagner, T., Donner, S., Beirle, S., Hilboll, A., Vrekoussis, M., Richter, A., Apituley, A., Piters, A., Allaart, M., Eskes, H., Frumau, A., Van Roozendael, M., Lampel, J., Platt, U., Schmitt, S., Swart, D., and Vonk, J.: Vertical Profiles of Tropospheric Ozone From MAX-DOAS Measurements During the CINDI-2 Campaign: Part 1—Development of a New Retrieval Algorithm, *J. Geophys. Res. Atmospheres*, 123, <https://doi.org/10.1029/2018JD028647>, 2018.

Chappuis-band retrieval advantages and limitations

The manuscript could more explicitly emphasize that Chappuis-band ozone absorption is only weakly temperature dependent, which is an inherent advantage compared to UV-based TOC retrievals. At the same time, the discussion of aerosol sensitivity could be expanded to better separate radiative transfer effects (e.g. path length changes) from true ozone-related effects.

We thank the reviewer for this helpful comment. The Introduction section has been revised to include a clearer discussion of the reduced sensitivity of TOC retrievals in the Chappuis bands to the effective ozone temperature. We now explain that ozone absorption cross sections in the UV exhibit a strong temperature dependence, which can introduce systematic uncertainties in TOC retrievals if the effective ozone temperature is not accurately represented. In contrast, ozone absorption in the Chappuis bands is only weakly temperature-dependent, making VIS-based TOC retrievals inherently less sensitive to temperature-related uncertainties. The discussion of aerosol-related effects has been addressed separately in response to the corresponding reviewer comment and clarified in Sect. 4.4 of the revised manuscript.

Aerosol impact interpretation

In the discussion of high-AOD conditions, the manuscript suggests changes in air mass factor as a possible explanation for observed deviations. Given that most aerosols are confined to the lower troposphere where ozone concentrations are relatively small, it would be helpful to clarify whether the observed effects are driven primarily by tropospheric ozone, by spectral fitting artefacts, or by radiative effects not fully captured by the AMF formulation.

We thank the reviewer for the comment. We agree that aerosols are predominantly located in the lower troposphere, where ozone concentrations are much lower than in the stratosphere, and therefore any aerosol-induced enhancement of the photon path would mainly affect the absorption signal of tropospheric ozone. As a result, the impact of aerosol scattering on TOC through a change of the AMF is expected to be relatively small, however not negligible. We have further elaborated on this topic in Sect. 4.4 of the revised manuscript. Under high aerosol load conditions, aerosol forward scattering, which is more pronounced in the visible spectral range, can modify the radiative transfer and hence the effective optical path length. In addition, part of the observed bias may arise from spectral fitting artefacts under enhanced aerosol loading. If not fully accounted for in the retrieval, these effects can propagate into systematic deviations in the retrieved TOC. A quantitative

assessment of this effect would require dedicated RTM simulations, but such analysis is beyond the scope of this study. The discussion of Sect. 4.4 has been revised accordingly.

GCOS ECV requirements

The performance of the presented total column ozone retrievals could be more explicitly discussed in the context of the GCOS ECV requirements for ozone (GCOS-245, 2022), which specify target uncertainty and stability levels for climate applications. The reported agreement of the UV retrievals with Brewer and Pandora measurements is generally consistent with the GCOS goal uncertainty of $\sim 1\%$, while the VIS retrievals meet this level primarily under low-aerosol conditions. A brief statement placing the results within the GCOS goal/breakthrough/threshold framework would strengthen the climate relevance of the study.

We thank the reviewer for this suggestion. Sect. 4.2 of the manuscript has been revised to place the performance of the retrieved TOC products within the GCOS ECV requirements framework (GCOS-245, 2022). We now state that the agreement of the UV-based TOC retrievals with the collocated Brewer and Pandora measurements is within approximately 1%, which is consistent with the GCOS target uncertainty for TOC. We also clarify that the VIS-based retrievals exhibit a somewhat larger spread, with most differences remaining within approximately $\pm 2.5\%$. As discussed in the revised manuscript, part of this increased variability is attributed to aerosol-related radiative transfer effects that influence VIS retrievals differently than in the UV.

Technical and editorial corrections

Some sentences, particularly in the Introduction and Methodology sections, are rather long and could be simplified for readability.

Several sentences in the revised manuscript, particularly in the Introduction (Section 1), Instrumentation and Data (Section 2), and Methodology (Section 3), have been revised and split into shorter sentences to improve clarity and readability.

Acronyms and technical terms (e.g. "I₀-correction") should be defined at first occurrence.

Sect. 3.2 has been revised to include a clear explanation of the "I₀-correction" and "intensity offset" terms, as well as their physical meaning in the DOAS retrieval. We now explain that the I₀ effect originates from inconsistencies between laboratory absorption cross sections measured with a smooth light source and the structured atmospheric absorption spectra recorded with lower spectral resolution, while the intensity offset compensates for potential stray light or dark signal that is not effectively removed from the measured spectrum.

Figure 7, 8, 14: X-axes labels are cut, please fix.

We thank the reviewer for pointing this out. The apparent cut of the X-axes labels is likely related to the PDF conversion process. The original figures contain the complete axis labels. This can be verified by zooming in on the figures of the initially submitted manuscript (e.g., at 200%). The issue is expected to be corrected during the typesetting stage if the manuscript is accepted for publication.

Ensure consistent notation and terminology for ozone columns (e.g. TOC vs. total ozone column) throughout the manuscript.

Done.

Line 16: "Delta TOC" is used without defining what this is.

Line 22: This time "Delta" is used.

Delta is now more clearly introduced at its first occurrence in the manuscript (Line 12)

Line 39: I wouldn't say that the Dobsons were "superseded" by the Brewers, this statement is not quite accurate scientifically or historically. It is true that in many networks, Brewers have become the primary operational instrument, while Dobsons continue to provide critical long-term reference measurements.

We thank the reviewer for this comment. We agree that the term "superseded" is not scientifically or historically accurate, as Dobson instruments continue to provide essential long-term reference measurements and remain part of ozone monitoring networks. The corresponding discussion in the Introduction has been revised.

Line 194: "44°C" should be " -44°C"

Done.

Line 523: "suffers by straylight" -> suffers from straylight

Done.