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 Karagkiozidis 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 ±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 Roozendael 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., 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.

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 ~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 ±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. "I_0-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.