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 ±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.