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The description of the effect of the ozone profile on UV radiation should be modified

Anonymous Referee #1

Referee comment on "Variability and trends in surface solar spectral ultraviolet irradiance in Italy: on the influence of geopotential height and lower-stratospheric ozone" by Ilias Fountoulakis et al., Atmos. Chem. Phys. Discuss.,
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The manuscript by Fountoulakis et al. correlates total ozone columns (TOC) and surface UV radiation at 307.5 and 324 nm with geopotential height at 250 hPa (GPH). While the anti-correlation between tropopause height and TOC has long been known, using GPH instead of tropopause height is a novel idea. Using GPH, the authors then explore the effect of atmospheric pressure patterns on short- and long-term variations in ozone and UV radiation. The results are interesting and worth publishing in ACP. In the second part of the paper, the authors then analyze UV measurements at three Italian stations for long-term trends and use TOC and GPH data to interpret these trends.

Most of the analyses are sound, with the exception of two issues: the effect of the atmospheric ozone profile on UV radiation and the effect of changes in aerosol and clouds on trends, as described in “Mayor Comments” below. The presentation is generally clear, although the Summary at the end is too long; too much detail distracts from the main messages. I recommend publication of the manuscript, provided that my Mayor and Minor comments below are addressed appropriately.

Mayor Comments:

The authors mention multiple times that the vertical redistribution of ozone in the atmosphere has an important effect on the global irradiance at 307.5 nm and that changes of this distribution over time can explain some trends in their UV measurements. My model calculations (see below) suggests that this effect is not important for the solar zenith angles (SZAs) of 45° and 67° considered in this study. The authors should perform their own model calculations and adjust their assertions accordingly.

Along the same lines, the authors imply that clouds and aerosols have a larger effect on surface UV radiation at a SZA of 45° compared to 67°. For example, they report that there are no significant trends in UV radiation at 67° at Rome while significant trends were calculated for 45° (their Figure 7). They attribute this difference to cloud and aerosol effects. My model calculations (also below) confirm that aerosol effects are larger at 67° than 45°. It is therefore unlikely that there is no trend at 67° but a signficant trend at 45° because of cloud or aerosol effects. Instead, I suspect that there is a problem in the UV radiation data at 45°. However, the data presented do not allow me to confirm this suspicion.

Minor Comments:

L54: Regarding "avoided extremely high levels of solar UVB radiation". Perhaps give a number here.

L56: The recent *Nature* paper by Young et al. (https://doi.org/10.1038/s41586-021-03737-3) could also be cited here.

L70: You could also cite: Ohring & Muench 1960: (https://doi.org/10.1175/1520-0469(1960)017<0195:RBOAMP>2.0.CO;2).

L72: Please provide a reference that the tropopause rises with warming of the troposphere.

L152: regarding "standard uncertainty for spectral UV measurements … of the order of 5%" So that would be 10% at the 2-sigma level?

L156: "smaller than 2.5%" is only half as large as the 5% quoted above. Why?

L168: Regarding "available for at least 15 days.": That's only half the days in a month. Significant error in monthly averages could occur if the missing days are biased towards either the start or end of a month. How was this problem addressed? Could this have caused spurious trends?

L225: The study focuses on anomalies in GPH at 250 hPa rather than tropopause altitude, the parameter frequently in other studies (see line 70). Figure S1 in the Supplement shows that there is a good correlation between the two parameters. Please describe the advantage of using GHP instead of tropopause height considering that the tropopause separates tropospheric and stratospheric ozone and therefore might be the more important parameter.

L253: Attributing correlation coefficients between GPH and the other parameters shown in Table 1 to "dynamical stratospheric processes" and "tropospheric processes" is a bit of a stretch. It would be more appropriate to say that a higher GPH emphasizes processes in the troposphere while a lower GPH emphasizes those in the stratosphere.

L263 - 285: Please structure better: First describe changes at SZA=67° based on Fig 3, then do the same for SZA=45°, based on Fig 4. Lastly, highlight the differences between trends at the two SZA.

L263-270: The text does not fit Figure 3. The whole paragraph is questionable, and while it identifies Figure 3, it should be mentioned that this description refers to the analysis of data at SZA=67°. Specifically, “307.5” in line 263 should be “324”; “324” in line 264 should be “307.5”. Regarding “The overall increase of the 307.5 irradiance for 2006 – 2020”: Do you refer to an increase averaged over all months? If so, how was the annual anomaly calculated considering the large difference in winter and summer UV radiation?

Lines 271 - 285: The text should be better structured. First, it should be said that this analysis is now for SZA=45° and that data from Aosta, Lampedusa, and Rome are discussed sequentially. For example: "We now discuss changes observed at a SZA of 45°. At Aosta, the irradiance at 324 nm increased by 0.6%/year in August...”

L275 - 277 and L351 - 353: As already noted above, I find it hard to believe that there were no trends at 67° but significant trends at 45° due to cloud and aerosol effect. Clouds and aerosols typically have a larger effect at 67° compared to 45°. To quantify this, I modeled spectra of global irradiance for 45° and 67°, either without aerosols or by assuming an aerosol layer. I parameterized the aerosol optical depth with Angström’s formula, setting alpha=1 and beta=0.25. This is a rather dense aerosol layer. At SZA=45°, global irradiance with aerosols was suppressed relative to the no-aerosols case by 17.5% at 307.5 nm and 16.3% at 324 nm. At 67°, global irradiance was lower by about 20% at both wavelengths, confirming that the effect of aerosols increases with SZA. Hence, I do not believe that clouds or aerosol are responsible for the different trends at 45° and 67°. Instead, I suspect some problems in the data, e.g., due to gaps. This issue should be explored further by the authors with their own model calculations.

L284: Considering that the changes for April and May are so different, can it be ruled that problems in the data, such as data gaps affecting the calculation of monthly averages, led to the high value in April?

L288 and L294: Why suddenly "SZA of 67.5°"? Up to now the SZA was 67°.

L300: Again, I find it hard to believe that the trends for a SZA at 45° and 67° (or 67.5° ?!?) are so different, in particular for April, and to a lesser extent for August and September. The fact that the large trend for April is also present at 324 nm suggests that the trend in ozone (Fig 8a) is not the driving factor.

L310: Regarding: "As the SZA increases the role of ozone at the middle and upper atmosphere becomes more important regarding the attenuation of the UV-B irradiance relative to ozone at the lower stratosphere." I presume that you refer to the Umkehr effect here. However, that effect is only significant for SZA > 80°. To confirm that the ozone profile has only a minor affect on UV irradiances at 307.5 and 324 nm for SZA of 45° and 67°, I ran model calculations (LibRadtran/UVSPEC) using either the standard mid-latitude profile (afglms.dat) or a modified profile where I increased the ozone concentration by 5% in the upper troposphere and lower stratosphere (between 13 and 20 km). I then scaled the original afglms profile and this modified profile to a TOC of 315 DU. This scaling effectively increased the ozone of the modified profile by 3.8% between 13 and 20 km and lowered it by 1.1% at all other altitudes. The global spectral irradiance at 307.5 nm calculated with the modified profile for SZA=45° was 0.06% larger than the irradiance calculated with the standard profile. The difference for 67° was 0.09%. At 324 nm, the difference was basically zero. These calculations show that the effect of the profile at these SZAs is negligible. So the sentence in line 310 should be removed.

L326-375: The summary is too long. Please shorten and emphasize the essential numbers and messages rather than repeating the Results section.

L355: The sentence "The increase … at Rome" is one example of a sentence that does not add much and could be deleted to make Section 4 more focused.

L358: As mentioned above, my model calculations do not support the assertion that the difference at 45° and 67° is due to clouds and aerosols. If the authors feel otherwise, they should support their assertion with their own calculations.

L359: "SZA decreases" > "SZA increases". ("SZA decreases" means that the Sun is closer to the zenith and the contribution of the direct irradiance becomes larger, not "less significant".)

L364: My model calculations above strongly suggest that the following sentence is either incorrect or greatly overstates the effect: "The difference between the observed and the expected change in the irradiance at 307.5 nm can be attributed to the fact that ozone

changed differently at different levels in the atmosphere."

L372: The same can be said about the sentence "… can be explained by the decreasing ozone in the lower stratosphere and the increasing ozone in the upper stratosphere."

L379: Again, at 67° the effect of the profile is negligible and the sentence "of upper stratospheric ozone to the attenuation of UVB irradiance becomes more significant with increasing SZA" should be removed.

L380: I agree that. "More robust statistical analyses and radiative transfer modelling are necessary in order to quantify the relative contribution of different factors to the short- and long-term changes of the surface solar UV irradiance in Italy" but I disagree that these calculations are "out of the scope of the present study." The authors assert multiple times that the vertical redistribution of ozone has an import effect on global UV irradiance for SZA of 45° and 67°, contrary to my calculations. They should make their own calculations to look into this issue in more detail than I did, and perhaps add an additional figure to the manuscript summarizing their results.

Technical suggestions / grammar / typos:

L13: Delete "In this study" (It is obvious that the abstract refers to this study.)

L14: over Italy > across Italy (Otherwise readers might think that UV was measured in the air above ground.)

L14: "located at quite different" > "characterized by quite different"

L16: "307.5 nm, 324 nm" > "307.5 nm and 324 nm"

L18: "geopotential height at 250 hPa (GPH)." > "geopotential height (GHP) at 250 hPa."

L28: "It was also showed that" > "It was also shown that" or "We also showed that"

L31: “period which” > “period, which” ; “aerosols were” > “aerosols, were”

L42: “is also absorbed” > “is absorbed”

L43: “ozone dominates on scattering” > “ozone has greater importance than scattering

L46: “leading to reduced” > “ but also reduced” (otherwise it sounds as if low- and mid-latitude ozone changes are only the result of high-latitude processes)

L52: “resulted on decreasing” > “resulted in decreasing”

L89: “parameters which affect significantly” > “parameters that significantly affect”

L96: “different latitudes and environmental conditions” > “and affected by differing environmental” (a site cannot be "located at an environmental condition”)

L97: “extent at which” > “extent to which”

L118: “referred as IOS standard.” > “referred to as IOS standard.”

L188: “GPH at 250 hPa” > “GPH at 250 hPa and at 850 hPa”, and delete "and ...850 hPa."

L243: “For 45°” > “For a SZA of 45° “

L279: “none” > “any”

L286: “3.2.1 Long-term variability in the period 1996 – 2020” > “3.2.1 Long-term variability at Rome for the period 1996 - 2020”

L348: “of the GPH” > “in the GPH”

L349: I don’t understand “was depicted to”

Supplement, line 3: “Figure A1” > “Figure S1”

Supplement, line 4: GHP at 250 hPa and 850 hPa are correlated, not “anti-correlated”

Supplement, Caption Figure S1: “tropo-pause” > “tropopause”; delete extra space after 850 hPa