

General comments

The paper by Czerwińska et al provides useful information regarding the effect of an urban agglomeration on the levels of the solar UV irradiance. It highlights the importance of the aerosol optical properties for the determination of the UV irradiance that reaches the earth surface and has the potential to contribute in the better understanding of the complex interactions between aerosols, clouds, surface albedo and UV radiation in an urban environment. The authors compare the erythemal and UV-A1 (340-400 nm) doses measured by the Brewer spectrophotometers in Warsaw (52.3°N, 21.0°E) and Belsk (51.8°N, 20.8°E) and are trying to quantify the effects of differences in surface albedo, cloudiness, aerosol optical depth and aerosol single scattering albedo.

However, the main problem in the data analysis is that the effect of different latitude (thus of different SZAs for the measured UV doses) has not been removed (or quantified) properly leading to biases in the quantification of the effect of other factors (such as the aerosol SSA and the cloudiness). The effect of different latitude changes periodically in the year and is more pronounced in winter (higher SZAs, which means that the difference of 0.5° becomes more important). The authors considered the effect of different SZAs to be invariant during the year which is not correct. Thus, I suggest to re-analyze the data and either quantify the effect of different latitude properly (e.g. with the use of a radiative transfer model) or perform the comparison for standard SZAs.

Answer:

We revised the modelling part. In the revised manuscript, we took into account the effect of different latitude and re-analysed data with the use of the LibRadtran simulations. Please see P7, L6-11:

“The difference in the geographical coordinates for the sites, which are based on the simulations of the erythemal and UV-A irradiances at 10:40 a.m. (i.e. near local noon) throughout 2015 leads to slightly higher values at Belsk. The modelled ratio changes with SZA (Fig. 6). The average ratio over the whole year is 1.03 ± 0.02 (1σ) for the erythemal irradiance and 1.02 ± 0.01 (1σ) for UV-A (324 nm). For the warm period (from 15 May to 14 September) modelled ratios were 1.01 ± 0.003 (1σ) and 1.01 ± 0.002 (1σ), but for the cold period (from 15 September to 14 May) modelled ratios were 1.04 ± 0.01 (1σ) and 1.03 ± 0.01 (1σ) – for erythemal and UV-A (324 nm) irradiances, respectively.”.

The second important issue that has to be solved prior to publication in ACP is the large number of editorial, grammatical and linguistic errors in the manuscript. The authors have to try hard to improve the manuscript. If possible, I suggest that the manuscript should be edited by a native English speaker.

Answer:

We did our best to improve the manuscript.

Additionally, I suggest reorganizing sections 3 and 4 as follows: 3.1. Comparison between measurements at Belsk, 3.2. Comparison between measurements at Belsk and Warsaw, 3.3. Quantification of the factors which are responsible for the differences, 3.4. Long-term change of the erythemal irradiance at Belsk. In section 3.3 you can include the numerical simulations (now section 3.3) and part of the discussion from section 4 (e.g. the results reported in P8, L15 – 31 regarding the effects of cloudiness). I also suggest moving figure 8 (and the relative discussion) in section 3.4 and expand the discussion. This way, I believe that it will be easier for the reader to follow the discussion.

Answer:

The paper was reorganised following the Referee's #1 suggestions.

The effect of different SZA (presented in paragraph 3.2) should be initially studied for UV-A1. Then, the combined effect of SZA and TO3 could be studied for the erythemal dose. Although the effect of SZA is stronger for lower wavelengths (due to stronger Rayleigh scattering and increased absorption by TO3 for larger SZAs) and the effects of SZA and TO3 on erythemal irradiance are not independent to each other, this way you could get a quantitative estimation regarding the effect of differences in TO3. However, the most effective way to quantify the effect of different TO3 is to study the ratios of UV-A1 and erythemal irradiance for specific SZAs (or small SZA intervals).

Answer:

We did the calculation for specific SZAs or time. Please see the results in section 3.3.

Specific comments

Please define a specific acronym to use in the document for each Brewer. For example, Brewer with Serial Number 207 is referred as BS No. 207, BS 207 and BS207 (without defining what the number 207 is) at different points of the document. Please choose a single acronym and use it everywhere (in the manuscript and the figures). E.g. you could define at the beginning of the methodology section that each BS with serial number xxx will be referred as BSxxx and then refer to each Brewer the same way.

Answer:

We changed the acronyms throughout the paper. They were defined on P3, L20-21:
“(…) by the single monochromator BS, serial number 64 (BS064), and in Warsaw since 2013 by the double monochromator BS, serial number 207 (BS207)”.

Abstract

P1, L7-8: replace “well-know” with “well-known”

P1, L8: replace “cleaner” with “less polluted”

P1, L9-11: replace the sentence:

“The present study focuses on differences in the erythemal and UV-A1 (340-400 nm) doses measured by the Brewer spectrophotometers in Warsaw (52.3°N, 21.0°E) and at Belsk (51.8°N, 20.8°E), which is located in a rural region at a distance of about 60 km in the south-west direction from the city.”

With

“The present study focuses on differences between the erythemal and UV-A1 (340-400 nm) doses measured by the Brewer spectrophotometers in Warsaw (52.3°N, 21.0°E) and Belsk (51.8°N, 20.8°E). The latter is a rural region at a distance of about 60 km south-west of the city of Warsaw.”

P1, L18: replace “by larger aerosol absorption” with “mainly by larger aerosol absorption over Warsaw”

P1, L18-19: The meaning of the phrase: “It appears that a slightly increased optical depth of the urban aerosols and properties of clouds generated over Warsaw are less important for the UV attenuation.” is not clear at this point. I would suggest replacing with something like:

“Differences between the aerosol optical depth and cloud optical properties over the two sites are found to be less important.”

P1, L19-20: replace “In this work we are showing that the higher city surface albedo compensates for the solar UV attenuation caused by urban aerosol load in the city of Warsaw.”

With something like:

“We show that the higher surface albedo in Warsaw compensates for the stronger attenuation of the solar UV radiation by the urban aerosols.”

Answer:

Suggested changes were made.

Introduction

P2, L5: add appropriate references

P2, L6: add appropriate references

Answer:

The references were added on P2, L9:

Greinert, R., de Vries, E., Erdmann, F., Espina, C., Auvinen, A., Kesminiene, A., and Schuz, J.: European Code against Cancer 4th edition: Ultraviolet radiation and cancer, *Cancer Epidemiol. Biomarkers Prev.*, 39, S75-S83, 2015,

Marionnet C., Pierrard, C., Golebiewski, C., Bernerd, F. et al.: Diversity of Biological Effects Induced by Longwave UVA Rays (UVA1) in Reconstructed Skin, *PLoS ONE* 9(8): e105263, doi:10.1371/journal.pone.0105263, 2014.

P2, L8: replace “depended” with “dependent”

P2, L11: replace “surface UV attenuation” with “attenuation of the solar UV radiation”

Answer:

Suggested changes were made.

P2, L10-12: add references to support your statement that “The absorption by SO₂ (in the UV-B range) and NO₂ (mostly in the UV-A range) is important for the surface UV attenuation only in extreme concentrations of such gases.”

Answer:

The sentence was changed to: “In the spectral range up to ~330 nm, absorption by ozone is usually much stronger than absorption by other main trace gases (SO₂, NO₂) (Cede et al., 2006)” (P2, L11-12). The reference is:

Cede, A., Herman, J., Richter, A., Krotkov, N., and Burrows, J.: Measurements of nitrogen dioxide total column amount using Brewer double spectrophotometer in direct Sun mode, *J. Geophys. Res.*, 111, D05304, doi:10.1029/2005JD006584, 2006.

P2, L12: Replace “surface intensity of UV” with “intensity of the solar UV radiation at the earth surface”. Furthermore, in addition to the properties, the amount of aerosols and clouds also affect UV radiation.

P2, L12-14: Change the phrase: “The negative trends in these variables, found over many of the northern hemisphere mid-latitudinal sites in the 1989s and 1990s, lead to increases of both the UV-B and UV-A irradiance”

With

“Increases of both the UV-B and UV-A irradiance have been reported over several mid-latitudinal sites of the northern hemisphere since the beginning of the 1990s, which have been mainly attributed to decreasing attenuation by aerosols and clouds.”

P2, L15: Replace “An” with “The”.

P2, L15: Replace “the large urban agglomeration” with “large urban agglomerations”.

P2, L17: Replace “UV cloudless sky irradiances” with cloudless-sky UV irradiances”. Also replace “its suburbs” with “a sub-urban area near Athens”.

P2, L19: Replace “The erythemal irradiance at the centre of Athens was 30% lower than at the suburbs with similar values of total ozone (TO₃) for days with increased pollution in the air.”

With

“The erythemal irradiance at the centre of Athens was up to 30% lower than at the outskirts site during days with increased air pollution over Athens basin and similar values of total ozone (TO₃) over the two sites.”

Answer:

Suggested changes were made.

P2, L20-21: What you write here is not clear. Please be more specific. Do you mean differences from the measurements or differences by corresponding simulations over the Athens basin?

Answer:

The sentence was clarified (P2, L22-23): “A similar difference was noticed in the modelled UV-B irradiance with input from measurements of the total ozone (TO₃) and aerosols optical depth (AOD) by the Brewer spectrophotometer (BS) at the outskirts of Athens.

P2, L23: Delete “the” before “winter” and “summer”. Replace “Mexico” with “Mexico City”.

P2, L23-24: Are 9% and 21% the differences between the annual mean levels for winter and summer? Please be more precise.

P2, L24-25: “Corr et al. (2009) ... 0.7 – 0.85”. This sentence is not clear. Please rephrase.

P2, L26: Delete “atmospheric”

P2, L30: Delete “of aerosols”

Answer:

Suggested changes were made.

P3, L3-8: Notice that for a typical Angström parameter of ~1.5, the differences in AOD becomes ~2 times larger for UV-B wavelengths. I suggest that you should provide quantitative estimations of the changes in UV irradiance due to the reported differences in AOD (e.g. for a low SSA = 0.85) at this point, to prove that the reported differences in AOD do not induce large differences in UV irradiance. That can be easily achieved by performing modeling simulations. Furthermore, it should be mentioned that for organic particles, the absorption in the UV range may be even larger than that predicted by interpolating using the Angström parameter for the visible range of the spectrum (e.g. see Bais et al. (2015)* and references therein).

* Bais, A. F., R. L. McKenzie, G. Bernhard, P. J. Aucamp, M. Ilyas, S. Madronich, and K. Tourpali (2015), Ozone depletion and climate change: impacts on UV radiation, *Photochemical & Photobiological Sciences*, 14(1), 19-52, doi:10.1039/c4pp90032d.

Answer:

We performed numerical simulations of the ratio with the use of measured AOD for both sites in the revised manuscript (section 3.3). The reference was added on P3, L8-10:

“However, for organic particles, the absorption in the UV range may be larger than predicted using Angström parameters for the visible range of the spectrum (Bais et al., 2015).”.

P3, L4: Remove “,” after “stated”.

P3, L6: “the difference” instead of “it”

P3, L11: Delete “a specific”

P3, L14: Delete “at”

Methodology

P3,L23: Replace “at Belsk (51.8°N, 20.8°E, 190 m amsl), which is located in a rural region” with “Belsk (51.8°N, 20.8°E, 190 m amsl). The latter is a rural region”

P3, L25: Replace “the area” with “an area”

P3, L28: Replace “spectra accuracy” with “spectral accuracy”

Answer:

Suggested changes were made.

P4, L3: Please provide reference(s) regarding the fact that the estimated uncertainty in the erythemal irradiance is 5%.

Answer:

The reference is:

Gröbner, J., and Schreder, J.: Protocol of the intercomparison at the Polish Geophysical Institute, Warsaw, Poland, May, 20-22 2004 with the travelling standard spectroradiometer B5503 from ECUV within the project QASUME, http://www.pmodwrc.ch/wcc_uv/qasume_audit/reports/2004_05_poland_warsaw_PGI1.pdf, 2004.

P4, L8: In this section you describe how each Brewer (207 and 64) is calibrated. Was the calibration procedure for the two Brewers the same before and after BS207 was moved to Warsaw? Is BS207 also calibrated against BS017? Please add some more information to convince the reader that the changes in the ratio are not due to a change in the calibration procedure or due to change of the BS207 characteristics during transportation from the one site to the other.

Answer:

We added information about the BS207 calibrations on P4, L11-13:

“BS207 was calibrated against BS017 in 2012 and 2013. After the calibration in 2013, it was moved to Warsaw. Furthermore, it has been calibrated 3 to 4 times per year since 2010 with a set of standard lamps that allows elimination of instrument ageing (loss of its sensitivity to UVR).”

P4, L11: Replace “The erythmal action spectrum follows CIE (1987)” with “The erythmal action spectrum is that suggested by the Commission internationale de l'éclairage (CIE) (CIE, 1987)”

Answer:

The suggested change was made.

P4, L13: Were there any criteria for the selection of the partly daily doses used for the comparison between Belsk and Warsaw? Is there a minimum amount of measurements (or measurements per 1 or 2 hours) below which the data are rejected? Calculation of the integral from only a small number of measurements and/or large gaps in the 3- or 6-hour period may lead to large differences between the integrals for the two sites. If not already done, I would suggest using a filter (e.g. use only time intervals with at least one measurement per 1 - 2 hours).

Answer:

We used a filter to 10 measurements per day. BS064 takes measurements 3 times per hour, while BS207 takes measurements 2 times per hour. It changes only if one of the BSs does not work properly, and then the previous filter should be enough. We added the filter for specific time intervals and a few points were removed. Although, that did not have any impact on the ratios.

P4, L15: How confident are you for your cloud detection method? Are there cloudy cases that cannot be detected? Can you estimate if, and in what extent, they affect your results?

Answer:

The answer to this question was incorporated in the text on P4, L23-24:

“There is no strict mathematical criterion applied here, but rather an intuitive inspection of the time series shape.”.

P4, L23-30: The specific paragraph is carelessly written. Please try to re-write it more carefully.

Results

Figure 1: Add some more information in the manuscript for LOWESS filter and/or proper references.

P5, L8-9: I think that the phrase “The most of the differences lie within $\pm 5\%$ range” is not necessary here since in the previous paragraph the 1σ uncertainty (which by the way is 7%) is given.

Answer:

Suggested changes were made. We added information and reference to LOWESS method on P4, L29-30: “The LOWESS (Locally Weighted Scatterplot Smoothing) filter (Cleveland, 1979) was used for smoothing of the curves.”.

The reference is:

Cleveland W.S.: Robust Locally Weighted Regression and Smoothing Scatterplots, Journal of the American Statistical Association 74(368): 829-836, 1979.

Figure 2a: In the specific figure there are two data points near 0.9. Is there any explanation for this large difference between the results from the two instruments for the particular days?

Answer:

Yes, they were probably affected by clouds and were removed.

P5, L22: Please specify that the higher latitude of the site at Warsaw means that for the same time, the solar zenith angle over Warsaw is always lower by $\sim 0.5^\circ$ compared to Belsk.

P5, L23: replace “surface albedo” with “different surface albedo”

P5, L25: replace “in” with “to perform”

P5, L27: Replace the phrase “prescribed values of surface albedo equal to 0.03 at Belsk and a set {0.03, 0.06, 0.12} in Warsaw” with “standard values of surface albedo equal to 0.03 for Belsk and 0.03, 0.06 and 0.12 for Warsaw”

P5, L28: Replace “of” before TO3 with “between” and delete “in”

P5, L30: Do you mean “coincidence” instead of “correspondence”? In this case it is for the entire range of the TO3 variability of both sites and not only of Belsk.

P6, L1: “assuming that” instead of “assuming”

Answer:

Suggested changes were made.

Figure 4: There is an obvious annual cycle of the ratio. I suppose that this is due to the stronger effect of the 0.5° difference in SZAs in winter, when the SZAs are larger. Thus, the effect of larger albedo compensates for the effect of different latitude only for a specific period of the year. The same annual cycle is obvious in Figure 5, possibly due to the remaining effect of the difference in SZA. Given that in Figure 4 there are no results for December and January, when the effect of SZA is expected to be even larger, while in Figure 5 there are results for the particular months, I believe that the deviation of the mean ratio from unity is partially due to the effect of different SZAs. I suggest that you should either quantify the remaining effect due to different SZAs and take it into account in the discussion of the results presented in Figures 5 and 6, or alternatively compare the irradiances for specific SZAs (thus slightly different time).

Answer:

Yes, we do agree with the Referee #1. We restructured the modelling section (now section 3.3) and performed a set of numerical simulations with LibRadtran. The dependence of the ratio on SZA was shown in Figure 6.

P6, L11: remove “of”

P6, L11: remove “in”

P6, L15: replace

“in the periods symmetrical around local noon for 6h for all-sky and 3h before noon for cloudless sky conditions”

with

“for 6h symmetrical periods around local noon for all-skies, and 3h periods before local noon for cloudless skies”

P6, L17: Delete “previously”

P6,L19: Replace “The” with “A”

Answer:

Suggested changes were made.

P6,L23: It seems to me that the ratio oscillates around ~1.05 and not 1. As already commented, I believe that part of the spread in the calculated ratios is due to the remaining effect of the difference in the latitude of the two sites.

Answer:

Yes, we do agree with the Referee #1. It was changed on P6, L15: “The ratio oscillates around 1.05 within the range between 0.9 and 1.2.”.

Figure 5: I suppose that the slightly different pattern of the temporal evolution of the ratios for the erythemal doses and the UV-A1 doses are again because of the effect of the different SZA. The effect of different SZAs is stronger for lower wavelengths, being partially responsible for the larger ratios of the erythemal doses compared to those of the UV-A1 doses.

We do agree with the Referee #1.

P7, L6: “attenuates” instead of “attenuate”

P7, L10: “emissions” instead of “emission”

Answer:

Suggested changes were made.

P7, L11: add references to support your statement that “causing numerous cases over the EU air quality threshold”

Answer:

The reference was added on P7, L27-28: “(...) causing numerous cases over the EU air quality threshold (Monitoring System of Air Quality in Mazowieckie Region, <http://sojp.wios.warszawa.pl/>).”.

P7, L12: what do you mean with the phrase “makes specific boundary layer”? Please explain (e.g. is the upper limit of the boundary layer higher compared to the nearby rural areas?).

P7, L12-13: Add references.

Answer:

The explanation and references were added on P7, L29-31:

“(…) i.e. in the boundary layer factors like wind, temperature, moisture, turbulence and energy budget fields differ from nearby rural sites (e.g. Fortuniak et al., 2005, Miao et al., 2009, Haberlie et al., 2015).”.

P7, L15: “higher AOD at 500 nm over Warsaw” instead of “higher Warsaw AOD values at 500 nm”

P7, L16: “similar differences” instead of “similar values”

P7, L21: “~2% more attenuation” is more accurate than “~2% attenuation”.

Answer:

Suggested changes were added.

P7, L21-24: Again, these numbers may change after re-evaluation of the effect of the SZA.

Answer:

Re-evaluated numbers were added on P8, L2-7:

“(…) the Belsk/Warsaw ratio between the erythemal and UV-A (324 nm) doses is ~1.06 and ~1.04, whereas the ratio is ~1.08 and ~1.06 for all-sky conditions, respectively. The aerosol effects are responsible for ~2% larger erythemal and UV-A near-noon doses at Belsk. The cloud effects add 2%, enlarging the Belsk-Warsaw difference. The SZA effects due to the longitudinal/latitudinal difference between the sites lead to 3% (or 2%) greater erythemal (or UV-A) doses at Belsk. The difference is even larger in the cold period of the year (for higher SZAs). The unexplained 1% higher doses at the rural site for the erythemal doses ratio could be attributable to instrument issues.”.

P7, L30-31: “An Indirect method for BS has been proposed by Bais et al. (2005)” instead of “Indirect method for BS has been proposed (Bais et al., 2005)”

Answer:

The suggested change was made.

P8, L2: add appropriate reference to support that the typical SSA for rural sites is 0.92.

Answer:

The explanation was added on P5, L7: “(...) SSA=0.92, which is a mean value measured by the CIMEL sunphotometer at Belsk (...)”.

P8, L2-4: Since the overall difference is 6% and AOD difference is responsible for 2% (according to what is written in the previous paragraph) then SSA differences should compensate for 4% - and not 5% - of the difference. However, this might be different if you take into account the effect of the SZA.

Answer:

The discussion about SSA and AOD effect was changed, after taking into account the effect of SZA. It is in section 4 (P8, L8-18):

“It seems possible that urban aerosols lead to higher absorption of the UV irradiance, i.e. small SSA values (<0.9) could characterise such aerosols. On the other hand, the albedo of urban surfaces is higher in the snowless period, that may compensate the effects of lower urban aerosols’ SSA. Analysing the UV radiation in the Mexico City metropolitan area, Castro et al. (2001) found the urban albedo of 0.12 over asphalt and grey surface cement sites. This is four times larger than the commonly used albedo of 0.03 over grass. Parisi et al. (2004) found that over some non-shaded parts of the city with high albedo (e.g. concrete surface) there is an amplification of the human exposure of up to 7% for people in the upright position. We performed RTM simulations with the observed TO3 and AOD values over Warsaw to fully compensate (by absorbing aerosols) the UV increase due to changes in albedo from 0.03 to 0.12. SSA=0.86 and 0.85, for SZA=60° and 30°, respectively, are found for the city site, i.e., 0.06 and 0.07 less than the value previously used in our RTM simulations for rural aerosols. Such estimate looks probable, as the Warsaw observing site is among the most polluted parts of the city because of abnormal vehicle emissions in the nearby main city roads.”.

P8, L5-7: I suggest moving the sentence “Kazadzis et al. (2009b) ... quality there” to P7, L27, after “... to SSA changes.”

P8, L6: “in Thessaloniki” instead of “in the Thessaloniki”

Answer:

Suggested changes were made.

P8, L20 – 31: As already commented, the effect of difference in SZA is more pronounced during the cold period (larger SZAs) and less pronounced during the warm period (smaller SZAs). The effect of different SZAs has to be removed – or taken into account - properly, so that you can get more accurate conclusions.

Answer:

It was reconsidered.

Figure 8: I recommend adding a paragraph (e.g. 3.4) and expand the discussion relative to Figure 8. Furthermore, the discussion regarding the agreement of your results with the results of other recent studies (e.g. Zerefos et al. (2012), de Bock et al. (2014), Fountoulakis et al. (2016)), as well as the reasons for this agreement could be expanded.

Answer:

We have decided to remove this figure and the part of the discussion as it is not connected with the Belsk-Warsaw comparison.

P8, L24 - 26: “Thus, it seems possible that increased cloudiness over urban areas does not necessarily mean increased attenuation of solar radiation, since modification of the cloud structure and properties by the urban aerosols may lead to the formation of clouds which attenuate the solar radiation less effectively” instead of

“Thus it seems possible that even higher cloudiness over urban areas does not mean higher attenuation of solar radiation, because the urban aerosols modify the cloud structure compensating the effect of increased cloud cover there”

P8, L27: “the” instead of “an”

P8, L32: “level” instead of “levels” and “higher than in the past” instead of “high”

Answer:

Suggested changes were made.

P8, L33: Zerefos et al. (2012) discuss the trends of the UVR after the mid-1990s. Thus, in the particular study they do not discuss the increase of the UVR due to the decrease of ozone until the mid-1990s.

P9, L2-3: Add proper reference to support your statement that that the environmental pollution was enormous in the mid-1970 and early-1980. Furthermore, the UV increases because aerosol and clouds decrease relative to the past – not because they were high in the past.

P9, L6: I do not think that 5-8% is “slightly” lower. I suggest removing the phrase “only slightly”.

Answer:

This part of the manuscript was removed. The long-term trend is out of main scope of the revised manuscript.

P9, L8: “parts” instead of “part”

Answer:

Suggested change was made.