

Response to Editor's (Stelios Kazadzis) comments

I would like to include here some comments on the revised manuscript.

Page 2, line 14, clouding – clouds

P 2, L31: trends – positive trends

P3 L16: "We will strive to support (or disprove) the hypothesis by comparing the erythemal and UV-A (324 nm) radiation measurements by the BSs in Warsaw and Belsk for the period May 2013-December 2015."

I think this can be eliminated as at this point as you can clearly say if this hypothesis is correct or not.

Answer:

The suggested changes were made.

Page 4 , L2 stray light needs a reference.

Answer:

The following reference was added to the manuscript:

Bais, A. F., Zerefos, C. S., McElroy, C. T.: Solar UVB measurements with the double- and single-monochromator Brewer ozone spectrophotometers, *Geophys. Res. Lett.*, 238, 833–836, doi: 10.1029/96GL00842, 1996.

P4, L28 the same ratio – Which one (wavelengths/erythemal) ?

Answer:

This issue was clarified in the paper, P4, L24-29: "Ratios between erythemal and UV-A (324 nm) doses (...) The same ratios are measured for the period of the Warsaw observations (May 2013 to December 2015) by BS207 and BS064 at Belsk to assess the impact of the urban agglomeration on the erythemal and UV-A radiation."

P5 line 8-13

Since there are other publication that are showing significant differences of UV ssa compared with the visible one especially at urban areas I would suggest to change the paragraph (and remove non used references after that) :

"We used SSA at 440 nm as a constant for the whole ultraviolet spectrum, as it was found that monthly averages estimated from BS at Uccle were in close agreement with the CIMEL measurements at 440 nm, especially for 320 nm (Nikitidou et al., 2013). Furthermore, Liu et al. (1991) performed Mie calculations for the rural aerosol model (Shettle and Fenn, 1979) and suggested that for this type of aerosol, SSA is approximately independent of wavelength. There are no measurements performed for SSA at the UV wavelength range."

To:

Since there are no AERONET related measurements of SSA at UV wavelengths , we used SSA at 440 nm as a constant for the whole ultraviolet spectrum, as it was found that monthly averages estimated from BS at Uccle were in close agreement with the CIMEL measurements at 440 nm, especially for 320 nm (Nikitidou et al., 2013).

Also, because the suggestion here that SSA is independent of wavelength is in contradiction with your discussion hypothesis of SSA can be lower in the UV.

Answer:

The suggested change was made.

P5 line 20 The mean ratio of which wavelength range (erythemal)?

Answer:

We meant erythemal doses. It was changed to "The mean value of the ratio between erythemal doses (...)" (P5, L19-20).

P6 line 8 : (local noon - 3h, local noon-0.5h) is not 3 hours.

Answer:

It should be “(local noon -3.5h, local noon -0.5h)” and was corrected.

Figure 8a: AOD ratios are misleading in this case. In addition, absolute AOD differences are related with changes in solar radiation and not their ratio. I would suggest to put AOD differences instead and change the text accordingly.

Answer:

The suggested change was made. Figure 8a and according text was corrected.

I would suggest to include a table in the end of section 3 including all mean cloudless sky ratios and standard deviations for all factors analyzed (intercomparison, solar angle, ozone, AOD, actual ratios). In order to summarize the quantification of all effects.

Answer:

All cloudless sky ratios and their standard deviations were included in Table 1.

I still think that the latitude difference of the two stations (solar zenith angle effect) as also pointed out from the reviewers can be eliminated. This is because including it to the factors affecting the differences among the sites introduces an uncertainty as it is changes from day to day and in the end in terms of percentage is the most important difference.

This can be done by either normalizing the irradiance of one of the stations using the solar zenith angel functions and compare them again. Or, as suggested, use ratios of measurements (and not 3 or 6 hour averages) for certain solar zenith angle windows e.g. X- 1 degrees where X can be e.g. 45 – 60 – 75 degrees. (75 degrees will capture the whole year). Then even if the measurements correspond to different time for the two stations, they are only slightly affected by the solar zenith angle issue.

Answer:

We calculated ratios according to Editor’s comment. The results are in Table 1 and on P7, L8-12:

“To eliminate the SZA’s effect on the ratios, we calculated also mean irradiances ratios for specified SZA windows for cloudless conditions. Calculations were done for SZA windows: $45^{\circ}\pm 1^{\circ}$, $60^{\circ}\pm 1^{\circ}$ and $75^{\circ}\pm 1^{\circ}$. For erythemal irradiances, the ratios were 1.02 ± 0.05 (1σ), 1.03 ± 0.04 (1σ) and 1.02 ± 0.05 (1σ), respectively. For UV-A (324 nm) irradiances, the ratios were 1.02 ± 0.05 (1σ), 1.02 ± 0.04 (1σ) and 1.01 ± 0.04 (1σ).”

We also added this result into discussion on P8, L5-8:

“The aerosol effects are responsible for ~2% larger erythemal and UV-A near-noon doses at Belsk, which stays in agreement with calculations of irradiances ratios between the sites for specified SZA windows ($45^{\circ}\pm 1^{\circ}$, $60^{\circ}\pm 1^{\circ}$ and $75^{\circ}\pm 1^{\circ}$). After eliminating the SZA’s effect for cloudless-sky conditions, both erythemal and UV-A (324 nm) irradiances at Belsk were ~2% higher than in Warsaw.”

conclusions

As you write (e.g. for the erythemal) you have (roughly) a 6% difference that can be attributed 3-4% on the different solar angles, 1% on the instrument differences and 2% to aerosol difference. So more or less everything is explained. Thus in the paragraph describing albedo and SSA you are mentioning two hypothetical (there are

no measurements) suggestions (a: albedo might be higher in Warsaw site and b. SSA might be lower). I would suggest rewriting this paragraph mostly suggesting that these two parameters (albedo and SSA); a. has been just assumed, b. they can be different and c. there is a possibility that (based on the modeling calculations) the effect of the one is masking the effect of the other. All the above, having in mind that this is a discussion that is not based in actual measurements.

Answer:

The paragraph was re-written to: “(...) We performed RTM simulations to show that the effect of higher surface albedo in Warsaw (the UV irradiances increase) can be compensated by lower values of SSA. We did not measure surface albedo and SSA values. Thus, we assume that the surface albedo in Warsaw can be in the range of 0.03 to 0.12 and 0.03 at Belsk. We also assume, that SSA at Belsk is 0.92, which is a mean value measured by CIMEL photometer at 440 nm. For calculations, we used observed TO_3 and AOD values over Warsaw. SSA=0.86 and 0.85, for SZA=60° and 30°, respectively, were 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 located in the most polluted part of the city because of high vehicle emissions from the nearby main city road.”

“Our study proves that the UV level in Warsaw is slightly lower than that found in cleaner suburbs of the city. Thus urban aerosols and clouds over Warsaw do not provide an effective shield against excessive UVR”

I would change that to “Our study proves that the UV level in Warsaw is slightly lower than that found in cleaner suburbs of the city. The differences that were attributed due to AOD differences are in the order of the accuracy of the instruments used. Based on the Brewer measurements, urban aerosols and clouds over Warsaw only partially act as an effective shield against excessive UVR. In addition, it would be interesting to try to justify this conclusion.

Answer:

The last paragraph was rephrased following the Editor’s suggestion. In addition, we added a justification of our conclusion on P9, L24-27: “For example, for UV index 5, time needed to get 1 MED (minimum erythema dose) for the person with phototype II is 33 minutes and for phototype III is 40 minutes (Fitzpatrick, 1988). Taking into consideration the attenuation of erythemal irradiances by 4%, which is the summarised effect of aerosols and clouds in Warsaw, this time for both phototypes changes only by 2 minutes. This small difference is not significant for planning and executing routine daily activities.”

Effects of urban agglomeration on surface UV doses: a comparison of Brewer measurements in Warsaw and Belsk, Poland, for the period 2013-2015

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Abstract. Specific aerosols and cloud properties over large urban regions seem to generate an island, similar to the well-known city heat island, leading to lower UV radiation intensity compared to the surrounding less polluted areas, thus creating a shield against excessive human exposure to UV radiation. The present study focuses on differences between erythemal and UV-A (324 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 located about 60 km south-west of the city. Ratios between erythemal and UV-A partly daily doses, obtained during all-sky and cloudless-sky conditions for the period May 2013-December 2015, were analysed to infer a specific cloud and aerosol forcing on the surface UV doses over Warsaw. Radiative model simulations were carried out to find sources of the observed differences between the sites. It was found that Warsaw urban agglomeration induced 8% and 6% attenuation of the erythemal and UV-A doses, respectively, mostly due to the lower Sun elevation in Warsaw during the near-noon measurements, and the larger optical depth of the city aerosols and increased cloudiness. It could be hypothesised that the expected stronger absorption of the solar UV radiation by urban aerosols is compensated here by a higher surface reflectivity over the city.

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1 Introduction

Excessive exposure to the ultraviolet radiation (UVR) reaching the Earth's surface has a detrimental impact on human health. The overexposure to UV-B radiation (290-315 nm) can cause erythema (redness of the skin), DNA and cellular damage (due to the generation of the reactive oxygen species), and immunosuppression. Longer UV wavelengths, UV-A (315-400 nm), can be cancerogenic but also responsible for photoaging and various eye diseases, including cataracts. Overexposure to both UV-B and UV-A could lead to increased risks of cutaneous melanoma, non-melanoma skin cancers, and various health problems (e.g. Marionnet et al., 2014; Greinert et al., 2015). While UV-B is strongly dependent on the latitude and thickness of the ozone layer, UV-A, especially UV-A1, the so-called long-wave UV-A (340-400 nm), is ozone independent, more intense, and less variable with latitude (Sabziparvar et al., 1999). 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).

The intensity of the solar UV radiation at the earth's surface depends significantly on properties and amount of ~~clouding~~ clouds and aerosols. Upward UV-B and UV-A trends have been reported over several mid-latitude sites of the northern hemisphere since the beginning of the 1990s, which have been mainly attributed to decreasing attenuation by aerosols and clouds (e.g. Krzyścin et al., 2011; Zerefos et al., 2012; De Bock et al., 2014).

Attenuation of the incoming solar radiation seems to be higher over large urban agglomerations relative to surrounding rural areas due to the excessive light scattering and absorption by anthropogenic aerosols. Papayannis et al. (1998) found differences between cloudless-sky UV irradiances measured over Athens and a suburban area near Athens. In Athens, the concentration of atmospheric aerosols was higher than at the suburban site. The erythemal irradiance at the centre of Athens was up to 30% lower than at the suburban site during days with increased air pollution over the Athens basin. 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. Acosta and Evans (2000) measured erythemal irradiances in the centre and suburbs of Mexico City in the period 1994-1995. During this period in winter, the erythemal irradiance was 9% greater in the suburbs than in the centre of Mexico City, while during summer, the recorded values were up to 43% greater (the mean value was 21%). Corr et al. (2009) found strong absorption of UVR by urban aerosols over Mexico City with a single scattering albedo (SSA) in the range 0.7-0.85. Even larger attenuation (~60%) of the UVR due to aerosols was reported in Guangzhou, China, in the dry season from October to January (Deng et al., 2012). Kazadzis et al. (2009a) found that for some cloudless days, differences in AOD among three sites (an urban, rural, and industrial area) located in Thessaloniki and at the outskirts of the city can account for up to 20% differences in the UV irradiance. Fountoulakis et al. (2016) noticed that positive trends in the amount of absorbing urban aerosols over Thessaloniki might counteract the expected UVR increase due to the long-term decrease of AOD there.

The atmosphere over Poland is one of the most particulate matter (PM) polluted in Europe. PM10 and PM2.5 levels measured in Warsaw, as well as in most other larger cities in Poland, exceed the tolerable PM limit many times during a year (Polish Ministry of Environment, 2014). However, Zawadzka et al. (2013) analyzed measurements taken by the Microtops II and CIMEL sunphotometer and stated that a small positive bias for AOD at 500 nm between Warsaw and a rural site (Belsk) which is ~60 km south-west of the city was not larger than 0.02, whereas for lower values of wind velocity the difference reached 0.04. The bias calculated from satellite measurements with MODIS (Moderate Resolution Imaging Spectroradiometer) was ~0.05. The authors did not find any significant differences in the Angström parameters between the sites for the visible range, so it could be hypothesised that AOD values in the UV range also differ only slightly. 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). Similar differences between Warsaw and Belsk were reported by Chubarova et al. (2011), who analysed results of aerosol measurements by the CIMEL sunphotometers located in Moscow (megacity with a population over 10 million) and Zvenigorod (population of approx. 16 thousand).

It seems possible that a large urban agglomeration could generate specific cloud properties (due to the heat island effect and creation of specific cloud condensation nuclei consisting of urban aerosols), higher loading of aerosols, and higher albedo than that in urban sites. The working hypothesis is that the Warsaw agglomeration produces a kind of shield against the incoming UV radiation. ~~We will strive to support (or disprove) the hypothesis by comparing the erythemal and UV-A (324 nm) radiation measurements by the BSs in Warsaw and Belsk for the period May 2013-December 2015.~~

2 Methodology

Monitoring of the UV spectra by BS is carried out by the Institute of Geophysics, Polish Academy of Sciences (IGF PAS), at the Central Geophysical Observatory Belsk since 1992 by the single monochromator BS, serial number 64 (BS064), and in Warsaw since 2013 by the double monochromator BS, serial number 207 (BS207) installed on the roof (elevation ~25 m above street level) of the IGF PAS main building. Previously, BS207 was working at Belsk (2010-2013). Comparison of BS064 and BS207 for that period will allow us to assess the differences between the measured UV doses due to instrumental differences. BS207 was moved to Warsaw in the middle of 2013.

The present study focuses on differences in the erythemal and UV-A (324 nm) doses measured by the BSs in Warsaw (52.3°N, 21.0°E, 130 m amsl) and Belsk (51.8°N, 20.8°E, 190 m amsl). The wavelength 324 nm was chosen because it is one of the longest wavelengths measured directly by both BSs and the gaseous absorption by the main trace gases (O₃, NO₂, and SO₂) is weak at this wavelength. The Belsk observatory is located in a rural region (the largest orchard region in Poland) far from urban and industrial developments. Surroundings of the city measuring site consist of grass, trees, concrete constructions (buildings, pedestrian footpaths), and asphalt roads.

BS064 is an older generation instrument - Mark II type, which is equipped with a single monochromator. Its spectral range is 290-325 nm in 0.5 nm steps and a spectral resolution of 0.6 nm (FWHM). The spectral accuracy decreases

for higher values of AOD and for larger solar zenith angles, i.e. for cases with an enlarged contribution of the diffuse component in the total UV radiation that increases the stray-light effect on the instrument- (Bais et al., 1996). Furthermore, it does not have a ventilation system. The quality control of its performance has been assessed by almost yearly calibration against the travelling world standard BS, serial number 17 (BS017). BS017 itself is regularly compared with a set of three Brewer instruments, so-called “Brewer reference triad” (Fioletov et al., 2005). BS064 was also compared with Bentham DM-150 during the project Quality Assurance of Spectral Ultraviolet Measurements (QUASUME) in May 2004 (Gröbner et al., 2005, 2006). The estimated 1σ uncertainty of the erythemal irradiance is about 5% (Gröbner and Schreder, 2004).

BS207 is the newest type instrument - Mark III, which is equipped with a double monochromator reducing significantly the stray-light effect. It is also equipped with a ventilation system which prevents overheating of the instrument during hotter days. Its spectral characteristics are the same as BS064, however the spectral range is wider extends to 363 nm. 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). For both instruments, the SHICRivm software was used to extend the spectra up to 400 nm and to eliminate erroneous spectra (Slaper et al., 1995).

The erythemal irradiance is calculated as the integral over the wavelength range 290-400 nm BS spectra after the SHICRivm standardisation, which is weighted by the erythemal action spectrum. The UV-A (324 nm) irradiance is taken without any weighting. The erythemal action spectrum is that suggested by the Commission Internationale de l'éclairage (CIE) (CIE, 1987). Further, the partly daily erythemal and UV-A (324 nm) doses are calculated as a time integral of the pertaining irradiance for the 6h period for all-sky conditions (local noon-3h, local noon +3h) and the 3h period for cloudless-sky conditions (local noon-3.5h, local noon-0.5h). Cloudless-sky conditions are identified using a two step algorithm. The first step is a preliminary search for such days using the criterion: the solar UV irradiance derivative with solar zenith angle is negative. In the next step, the smoothness of the time series for the day which fulfilled the first criterion, is examined, i.e. the bell-shape of the UV time series must be identified. There is no strict mathematical criterion applied here, but rather an intuitive inspection of the time series shape.

Ratios between erythemal and UV-A (324 nm) doses measured by BS064 and BS207, based on collocated observations at Belsk for the period October 2010 – April 2013, allow us to estimate the uncertainty range of the ratio related to differences in BS instrumental characteristics and in time of observations. The BS measurements are not synchronised, as the spectrum ranges are different. The same ~~ratio is~~ ratios are measured for the period of the Warsaw observations (May 2013 to December 2015) by BS207 and BS064 at Belsk to assess the impact of the urban agglomeration on the erythemal and UV-A radiation. The LOWESS (Locally Weighted Scatterplot Smoothing) filter (Cleveland, 1979) was used for smoothing of the curves.

Numerical simulations for the cloudless-sky conditions of the ratio between Warsaw and Belsk were performed to calculate differences caused by various factors, such as the geographical location (Belsk is slightly to the south), TO_3 , surface albedo, and aerosols properties (AOD, SSA). Simulations were performed with the radiation transfer model (RTM)

libRadtran (Mayer and Kylling, 2005). The following model input parameters, which are from the simultaneous measurements at both sites, are used in calculations: daily mean total ozone by the BSs standard measurements, AOD at 550 nm measured by the MODIS for the period 2013-2015. MODIS Aerosol Product values are available globally and include AOD at 550 nm over land and ocean. Remote sensing of aerosol properties using MODIS is presented by Ichoku et al. (2004). In this study, we used arrays of Level 2 (MOD 04, Collection 6) data produced daily at the spatial resolution of 10×10 km pixelation. From satellite data, we selected daily mean values of AOD from the nearest pixel to the measurement sites. Other input parameters are constants representing typical values used in the UV modelling, e.g. albedo of 0.03 for rural surfaces and SSA=0.92, which is a mean value measured by the CIMEL sunphotometer at Belsk (level 1.5 from AERONET – Aerosol Robotic Network) at 440 nm (<http://aeronet.gsfc.nasa.gov>). ~~We Since there are no AERONET related measurements of SSA at UV wavelengths, we~~ used SSA at 440 nm as a constant for the whole ultraviolet spectrum, as it was found that monthly averages estimated from BS at Uccle were in close agreement with the CIMEL measurements at 440 nm, especially for 320 nm (Nikitidou et al., 2013). ~~Furthermore, Liu et al. (1991) performed Mie calculations for the rural aerosol model (Shettle and Fenn, 1979) and suggested that for this type of aerosol, SSA is approximately independent of wavelength. There are no measurements performed for SSA at the UV wavelength range. 2013).~~ To identify the impact of the selected parameter on the ratio between the sites' doses we use the RTM model, allowing variability only for this parameter and keeping constant other RTM input parameters. For example, to quantify the dependence of the ratio on the geographical location of the sites the RTM simulations were performed using fixed TO₃, AOD, and time (10:40 GMT), but the simulations were for consecutive days throughout the whole year.

3 Results

3.1 Comparison between measurements at Belsk

In the period from October 2010 to April 2013 both BSs were working simultaneously at Belsk. Figure 1a shows the time series of the measured ratio (BS064/BS207) between the 6h erythemal all-sky doses. The mean value of the ratio between erythemal doses is $1.02 \pm 0.07(1\sigma)$. Figure 1b illustrates that the 1-1 relation between the doses is appropriate for the whole range of measured irradiances. The coefficient of determination based on this data set is 0.99. The mean ratio for UV-A (324 nm) is $1.02 \pm 0.07(1\sigma)$ for all-sky conditions.

Outliers greater than 10% sometimes appear, as the measurements were not synchronous. It is difficult to have synchronised measurements by our BSs, as the scanning time is different because of the various spectral ranges, i.e. 290-325 nm for BS064 and 290-363 nm for BS207. BS064 measures UV spectrum three times per hour, BS207 only two times per hour. Thus local cloudiness may be a source of large standard deviations of the mean ratios calculated during all-sky conditions. To remove the effect of cloudiness, we analysed the ratios derived from 3h cloudless-sky measurements before solar noon. The cloudless-sky doses were calculated for a shorter period compared to those for the all-sky conditions, as cloudless-sky conditions in Poland usually prevail before noon.

Figure 2a shows the time series of the measured BS064/BS207 ratio for the cloudless-sky conditions and the corresponding scatter plot (Fig.2b). The mean value of the ratio is 1.01 ± 0.03 (1σ) and there is almost a 1-1 relation between the erythemal doses by both BSs. That is also supported by high value (0.998) of the coefficient of determination. For UV-A (324 nm) doses, the ratio is 1.00 ± 0.04 (1σ). Thus, the performance of BS064 and BS207 was practically the same during the Belsk's intercomparison. The agreement between the output of both BSs was almost perfect, suggesting that the instrumental differences did not have much influence on the ratio between the doses.

3.2 Comparison between measurements at Belsk and Warsaw

BSs were working simultaneously in Warsaw and Belsk in the period from May 2013 to December 2015. The erythemal and UV-A (324 nm) doses calculated for these sites for 6h periods (local noon - 3h, local noon + 3h) for all-skies, and 3h periods (local noon - ~~3h~~3.5h, local noon-0.5h) for cloudless skies, were analyzed to find the Belsk-Warsaw ratio between the measured doses (BS064/BS207). If the ratio obtained during the cloudless-sky conditions differs significantly from that obtained during the cloudless-sky conditions during the Belsk's BSs intercomparison, it will allow us to estimate the effect of urban aerosols on the surface UVR. A similar approach with the use of all-sky data will also provide an estimate of the effect of urban cloud on the surface UVR.

Figure 3 shows the time series of BS064/BS207 measured ratio for erythemal doses (Fig. 3a) and the UV-A (324 nm) doses (Fig. 3b) for cloudless-sky conditions simultaneously appearing both in Warsaw and Belsk during 3h measurements. The ratio oscillates around 1.05 within the range between 0.9 and 1.2. The main reason for this scatter is using the interpolated erythemal (or UV-A) irradiance values at the beginning (local noon-3.5h) and at the end (local noon-0.5h) of the calculated period. BS observations rarely coincided with these moments. Thus linear interpolated values were used, taken from observations closest to the beginning or to the end of the period, i.e. the irradiance values just outside the observing period were also taken into account. The mean value of the Belsk-Warsaw ratio is 1.06 ± 0.04 (1σ) and 1.04 ± 0.04 (1σ) for the erythemal and UV-A (324 nm) dose, respectively. The corresponding values calculated for 6h doses during all-sky conditions (Fig.4) are 1.08 ± 0.19 (1σ) and 1.06 ± 0.18 (1σ), respectively. Much larger uncertainty ranges of the estimates for all-sky conditions are due to the cloudiness effects, but the mean values of the ratio are only slightly larger than those found during the Belsk intercomparison of the instruments. In spite of possible different cloud properties over Belsk and Warsaw during 6h measurements, the determination coefficient values are still high, i.e. equal to 0.96 for erythemal and UV-A (324 nm) doses. The 1-1 correspondence between doses is maintained for the whole range of the data (Fig.5).

The standard statistical test for a difference in the mean values taken from two large samples of an unknown distribution (Daniel and Cross, 2013) was used to find out if the BS064/BS207 mean ratio obtained during Belsk-Warsaw comparison of BSs is significantly larger than the ratio found during the Belsk intercomparison. The hypothesis that the mean value of BS064/BS207 ratio is higher during the Belsk-Warsaw comparison is supported by the test at the significance level better than 0.01 both for cloudless-sky and all-sky conditions.

3.3 Sources of the Belsk-Warsaw differences in the erythemal and UV-A doses

The more northern location of the Warsaw site results in lower SZA of $\sim 0.5^\circ$ at the same time for BSs observations. Other factors affecting the ratio between the measured doses at the rural and urban site during cloudless conditions are differences in TO_3 , surface albedo, and aerosol properties (AOD or SSA). In this sub-section, the modelled cloudless-sky irradiances are analysed for Warsaw and Belsk to discuss sources of the BS064/BS207 ratio variability.

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 GMT (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. To eliminate the SZA's effect on the ratios, we calculated also mean irradiances ratios for specified SZA windows for cloudless conditions. Calculations were done for SZA windows: $45^\circ \pm 1^\circ$, $60^\circ \pm 1^\circ$ and $75^\circ \pm 1^\circ$. For erythemal irradiances, the ratios were 1.02 ± 0.05 (1σ), 1.03 ± 0.04 (1σ) and 1.02 ± 0.05 (1σ), respectively. For UV-A (324 nm) irradiances, the ratios were 1.02 ± 0.05 (1σ), 1.02 ± 0.04 (1σ) and 1.01 ± 0.04 (1σ).

The total ozone difference between the sites is quite small (Fig.7). The mean TO_3 ratio (BS064/BS207) taken from all coinciding daily TO_3 values is 1.00 ± 0.01 (1σ). All data points are in close proximity to the diagonal line representing the 1-1 relationship between the variables (Fig. 7b). The mean modelled ratios between the erythemal and UV-A (324 nm) irradiances calculated for the selected fixed SZA and the site measured TO_3 values are 1.00 ± 0.01 (1σ) and 1.00 ± 0.002 (1σ), respectively, for all considered SZAs (30° , 40° , 50° and 60°). Thus, TO_3 is not a factor responsible for the UV difference between the sites.

The AOD effect on the Brewers' ratio is inferred from the RTM simulations based on the measured AOD at 550 nm by MODIS for the period 2013-2015 on days when the data were available for both sites. Daily AOD means are taken into consideration. The calculation was performed separately for various SZAs (40° , 60° and 70°) and fixed $\text{SSA}=0.92$. Fig.8 shows that AOD at 550 nm is slightly higher over the city. The mean AOD is equal to 0.26 and 0.20 over the urban and rural site, respectively. RTM simulations performed using the observed AOD values for various fixed SZAs (40° , 60° and 70°) yield the BS064/BS207 ~~ratio~~difference is almost the same ~~1.02 ± 0.02~~ 0.9206 ± 0.07 (1σ) for all considered SZAs, for the erythemal and UV-A (324 nm) irradiances. All calculated ratios are shown in Table 1.

4 Discussion and conclusions

Warsaw agglomeration has over 3.5 million population, with high pollution due to heavy vehicle emissions and industry (mainly electric power), causing numerous cases over the EU air quality threshold (Monitoring System of Air Quality in Mazowieckie Region, <http://sojp.wios.warszawa.pl/>). Like other large cities, it is expected that Warsaw produces

the well-known heat island that makes specific boundary layer, 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), allowing anthropogenic aerosols to reach higher atmospheric layers that may enhance AOD and affect cloud properties (e.g. level of cloudiness, droplet size, liquid water content).

5 The Warsaw agglomeration attenuates only slightly the erythemal and UV-A (324nm) radiation. Under cloudless conditions, 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, which stays in agreement with calculations of irradiances ratios between the sites for specified SZA windows ($45^{\circ}\pm 1^{\circ}$, $60^{\circ}\pm 1^{\circ}$ and $75^{\circ}\pm 1^{\circ}$). After eliminating the SZA's effect for cloudless-sky conditions, both erythemal and UV-A (324 nm) irradiances at Belsk were ~2% higher than in Warsaw. 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.

15 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~~ to show that the effect of higher surface albedo in Warsaw (the UV irradiances increase) can be compensated by lower values of SSA. We did not measure surface albedo and SSA values. Thus, we assume that the surface albedo in Warsaw can be in the range of 0.03 to 0.12 and 0.03 at Belsk. We also assume, that SSA at Belsk is 0.92, which is a mean value measured by CIMEL photometer at 440 nm. For calculations, we used 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~~ were 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 ~~located in~~ the most polluted part ~~part~~ of the city because of abnormal ~~high~~ vehicle emissions in ~~from~~ the nearby main city roads ~~road~~.

30 Fountoulakis et al. (2016) discussed factors important for the UV spectral variability in Thessaloniki. They pointed out that the cloudless-sky UV-A irradiances could be sensitive not only to AOD changes but also to SSA changes. Kazadzis et al. (2009b) found that UV-A irradiance increase in Thessaloniki for the period 1998- 2006 cannot be explained only by the AOD changes, but also by the changes of SSA over the area, due to the improvement of the air quality there. Chubarova et al. (2011) analysing results by CIMEL sun photometers located in Moscow and in Zvenigorod (less polluted site) found that the uncertainty range of SSA is too high, precluding discussion of the SSA urban effects. However, they found that SSA in

Moscow for the visible range of solar radiation was 0.02-0.03 smaller than that obtained from the clean site. It is worth mentioning that there is a lack of the direct retrieval to obtain SSA from UV spectral measurements. An indirect method for BS was proposed by Bais et al. (2005) depending on the assumed values of the asymmetry parameter, surface albedo, aerosol vertical profile, and the extraterrestrial solar spectrum.

5 The cloud effects should be more pronounced during the warm period of the year, where the city heat island may generate stronger convection than that in the cold period of the year. Romanov (1999) analysing NOAA satellite images retrieved higher cloud cover in summer over central Moscow compared to its suburbs. Inoue and Kimura (2004) found that in Tokyo there were more low-level clouds in the summer period (July-August) compared to rural sites in Kanto region. Moreover, urban heat islands lead to more thunderstorm initiation episodes (e.g. Shepherd, 2005; Haberie et al., 2015).

10 The classical theory (Twomey, 1977) states that when there are more aerosols high above the surface due to stronger updraft generated by the city warm island, aerosols serve as cloud condensation nuclei, reduce the size of cloud effective radius and increase the number of droplets, causing larger cloud optical thickness (COT) and finally higher attenuation of radiation reaching the Earth's surface. Thus, additional cloudiness generated over large cities may act as an umbrella against excessive UV radiation. We calculate BS064/BS207 ratio during Belsk-Warsaw comparison campaign
15 taking into account measurements in the warm period, 15 May – 14 September. We expected to find a higher ratio for that period according to the classical theory stated above. However, the ratio is only slightly lower, i.e. $1.06 \pm 0.17(1\sigma)$, for the erythemal doses, and $1.06 \pm 0.16(1\sigma)$ for UV-A (324 nm) doses, and part of this difference is the effect of different SZAs between the sites. This may suggest that contrary to expectation, COT is smaller over urban areas. Jin et al. (2005) discussed aerosol-cloud relationship over New York and Houston. They found that thick urban aerosols correspond to low COT there.
20 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.

Our study proves that the UV level in Warsaw is slightly lower than that found in cleaner suburbs of the city. ~~Thus urban aerosols and clouds over Warsaw do not provide an effective shield against excessive UVR.~~ The differences that were
25 attributed due to AOD differences are in the order of the accuracy of the instruments used. Based on the Brewer measurements, urban aerosols and clouds over Warsaw only partially act as an effective shield against excessive UVR. For example, for UV index 5, time needed to get 1 MED (minimum erythema dose) for the person with phototype II is 33 minutes and for phototype III is 40 minutes (Fitzpatrick, 1988). Taking into consideration the attenuation of erythemal irradiances by 4%, which is the summarised effect of aerosols and clouds in Warsaw, this time for both phototypes changes
30 only by 2 minutes. This small difference is not significant for planning and executing routine daily activities.

Acknowledgements. This work was partially supported by statutory activities No. 3841/E-41/S/2016 of the Ministry of Science and Higher Education of Poland. We appreciate support from early-carrier scientist grant (A. Czerwińska) No. 500-10-18 by the Institute of Geophysics, Polish Academy of Sciences.

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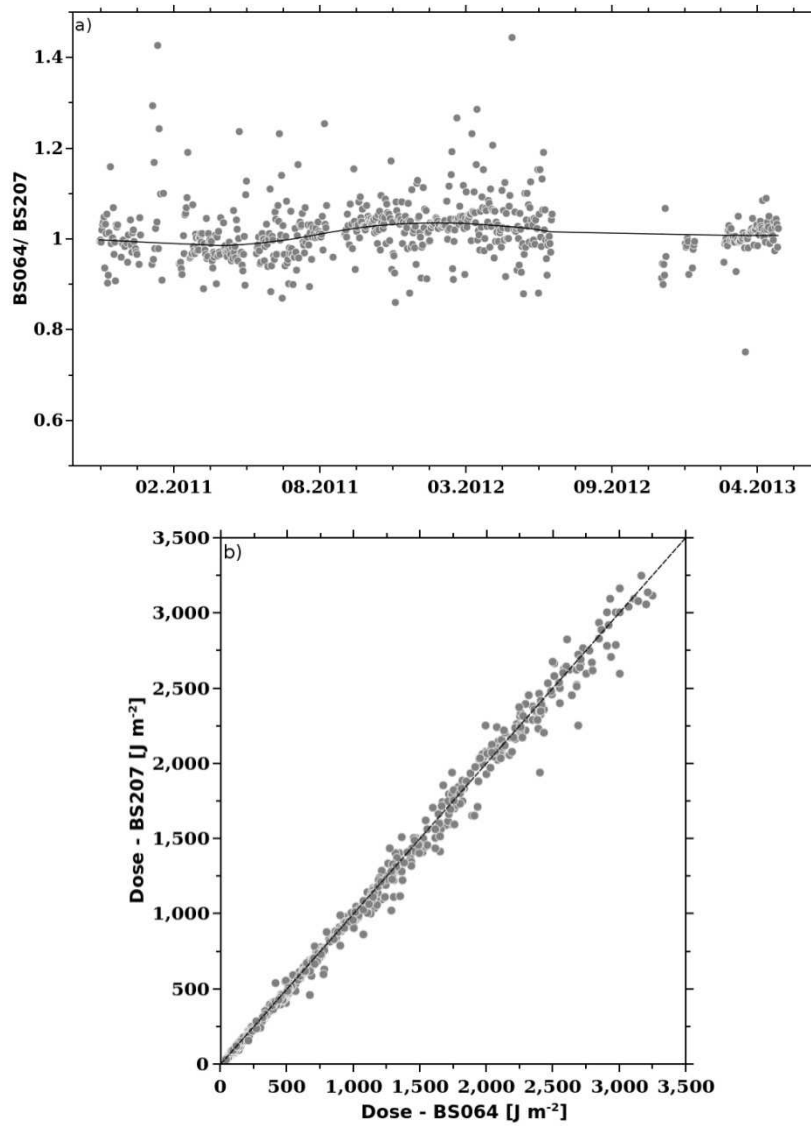
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	<u>Ratio</u> <u>(erythema)</u>	<u>Ratio</u> <u>(324 nm)</u>
<u>B/W, SZA 45°±1° (meas.)</u>	<u>1.02±0.05</u>	<u>1.02±0.05</u>
<u>B/W, SZA 60°±1° (meas.)</u>	<u>1.03±0.04</u>	<u>1.02±0.04</u>
<u>B/W, SZA 75°±1° (meas.)</u>	<u>1.02±0.05</u>	<u>1.01±0.04</u>
<u>B/B (meas.)</u>	<u>1.01±0.03</u>	<u>1.00±0.03</u>
<u>B/W SZA (mod.)</u>	<u>1.03±0.02</u>	<u>1.02±0.01</u>
<u>B/W TO₃ (mod.)</u>	<u>1.00±0.01</u>	<u>1.00±0.01</u>
<u>B/W AOD (mod.)</u>	<u>1.02±0.02</u>	<u>1.02±0.02</u>
<u>B/W (meas.)</u>	<u>1.06±0.04</u>	<u>1.04±0.04</u>

5 **Table 1: Mean ratios and corresponding standard deviations for comparison between UV doses and irradiances (erythema, UV-A – 324 nm) modelled (mod.) by LibRadtran and measured (meas.) by BSs in Warsaw (W) and Belsk (B) for cloudless-sky conditions. B/B is a ratio between doses for BSs intercomparison (BS064/BS207) at Belsk.**



5 **Figure 1a:** The ratio between erythemal 6h (noon \pm 3 hr) doses measured by the Brewer Spectrophotometer No. 64 and No. 207 while working simultaneously at Belsk (all-sky conditions). The solid curve represents the smoothed data by LOWESS filter.

Figure 1b: Scatter plot of doses measured by Brewer spectrophotometers No. 207 and No. 64.

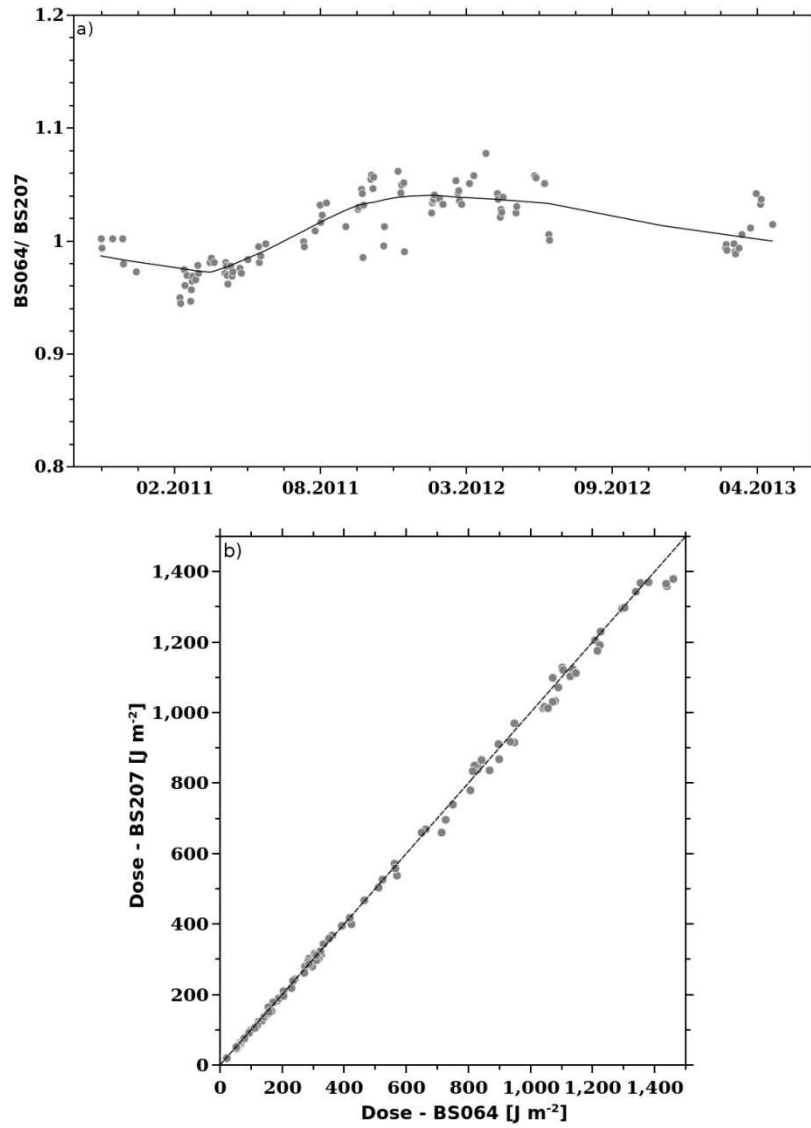
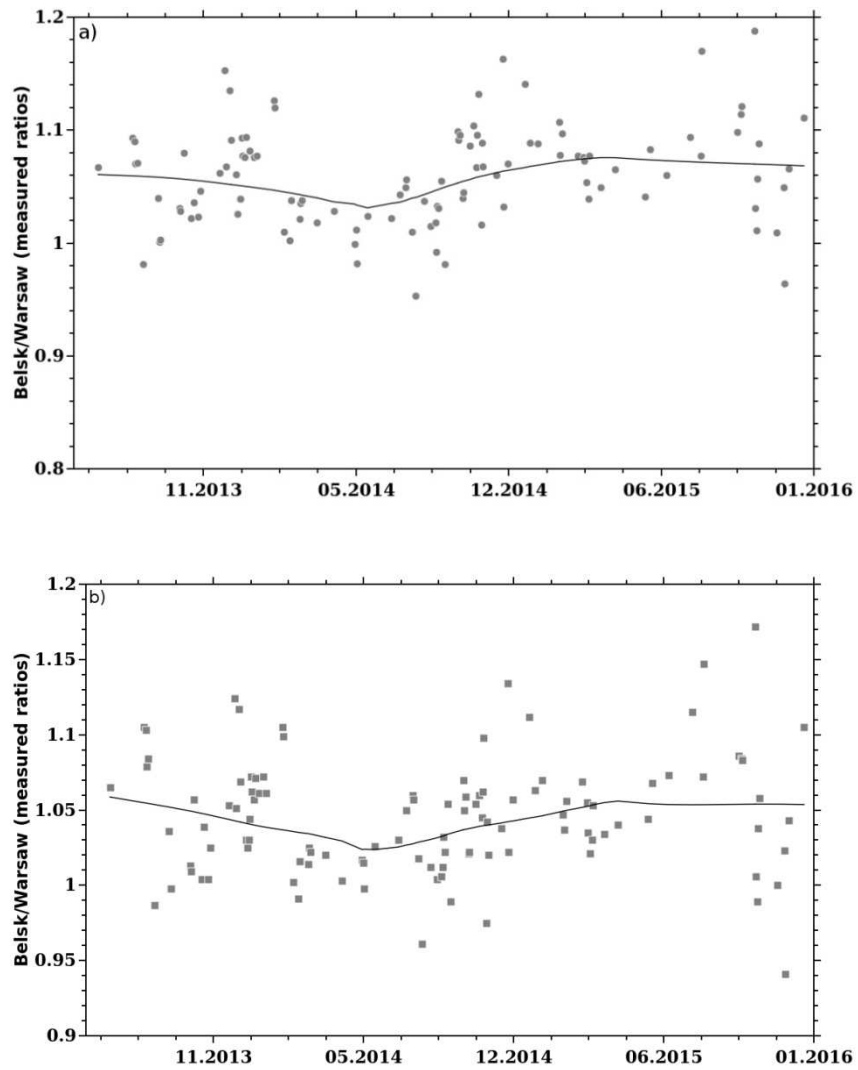


Figure 2a: Same as Fig.1a but for cloudless-sky conditions and 3h doses calculated for the period before noon (noon-3.5h, noon-0.5h).

5 Figure 2b: Same as Fig.1b but for cloudless-sky conditions and 3h doses calculated for the period before noon (noon-3.5h, noon-0.5h).



5 Figure 3: The Belsk/Warsaw ratio between the partial daily 3h doses (noon-3.5h, noon-0.5h) measured during cloudless-sky conditions existing over both sites for erythemal doses (a) and UV-A (324nm) doses (b). Solid curves represent the smoothed data by LOWESS filter.

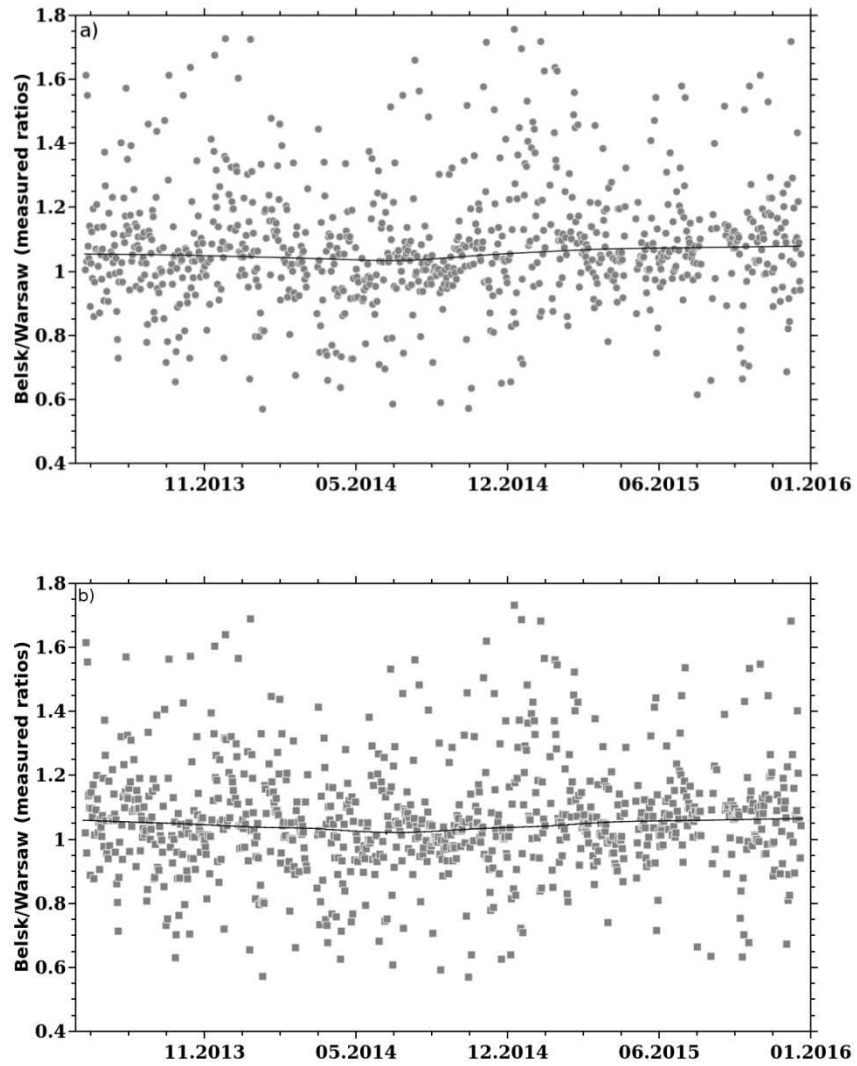


Figure 4: Same as Fig.5 but for the near noon partial daily dose (noon-3h, noon+3h) for all-sky conditions. Solid curves represent the smoothed data by LOWESS filter.

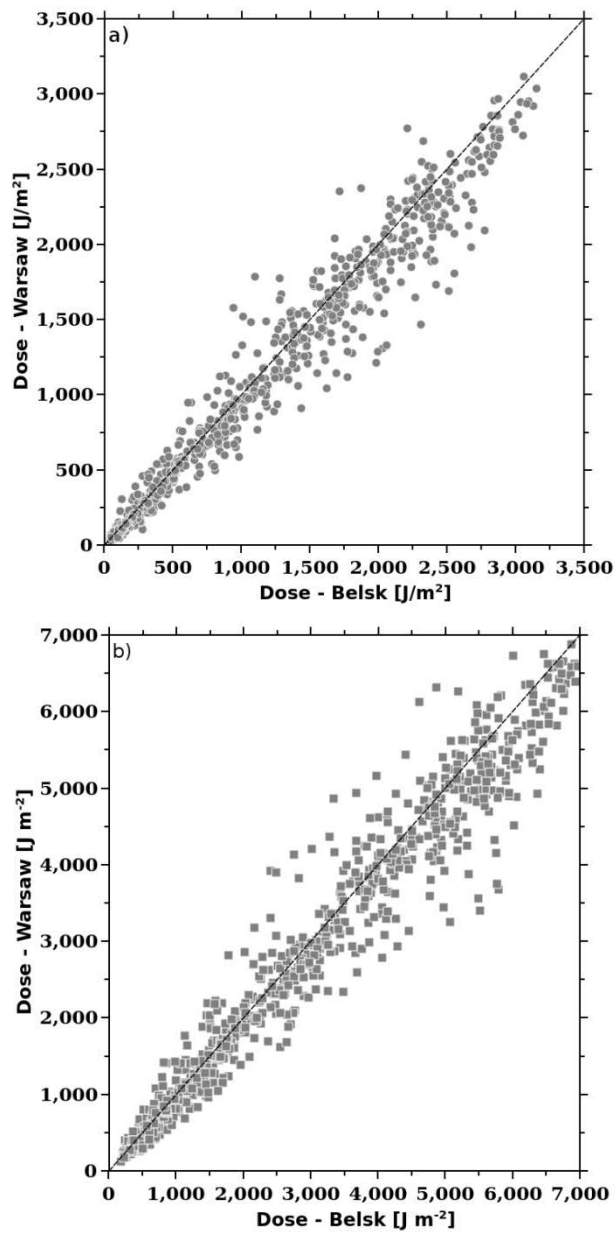
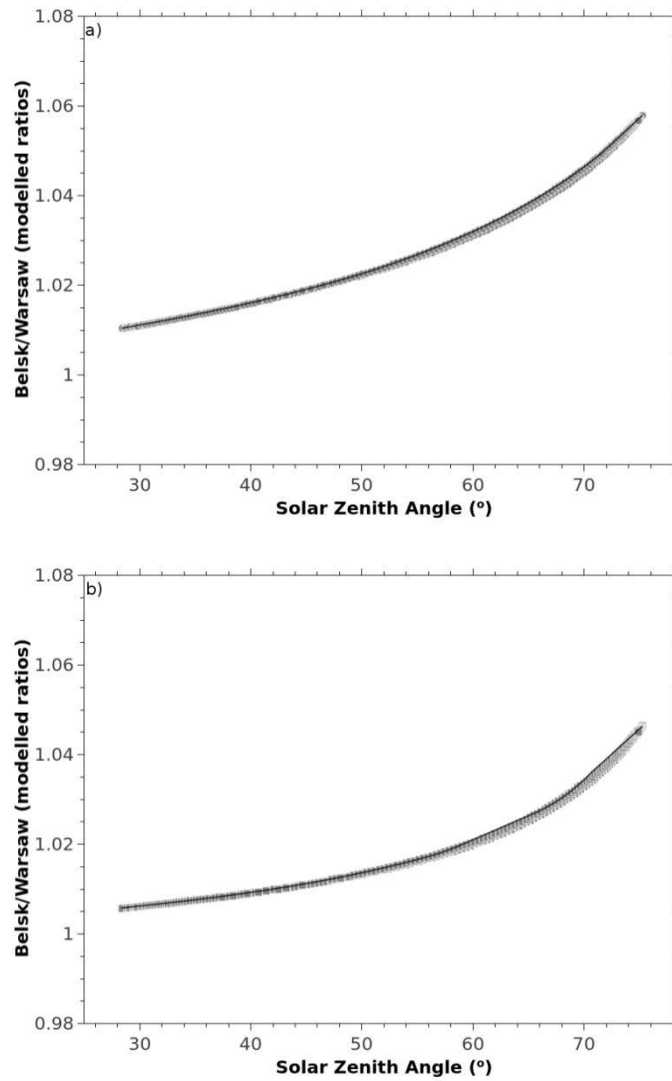


Figure 5: Scatter plot of partial daily doses (local noon-3h, local noon+3h) measured in Warsaw and Belsk: erythemal doses (a) and UV-A (324nm) doses (b).



5 **Figure 6: The Belsk/Warsaw ratio between erythemal (a) and UV-A (324nm) (b) irradiances calculated by the libRadtran model for 2015 versus SZA at Belsk for the 10:40 (GMT). The solid curve represents the smoothed data by LOWESS filter.**

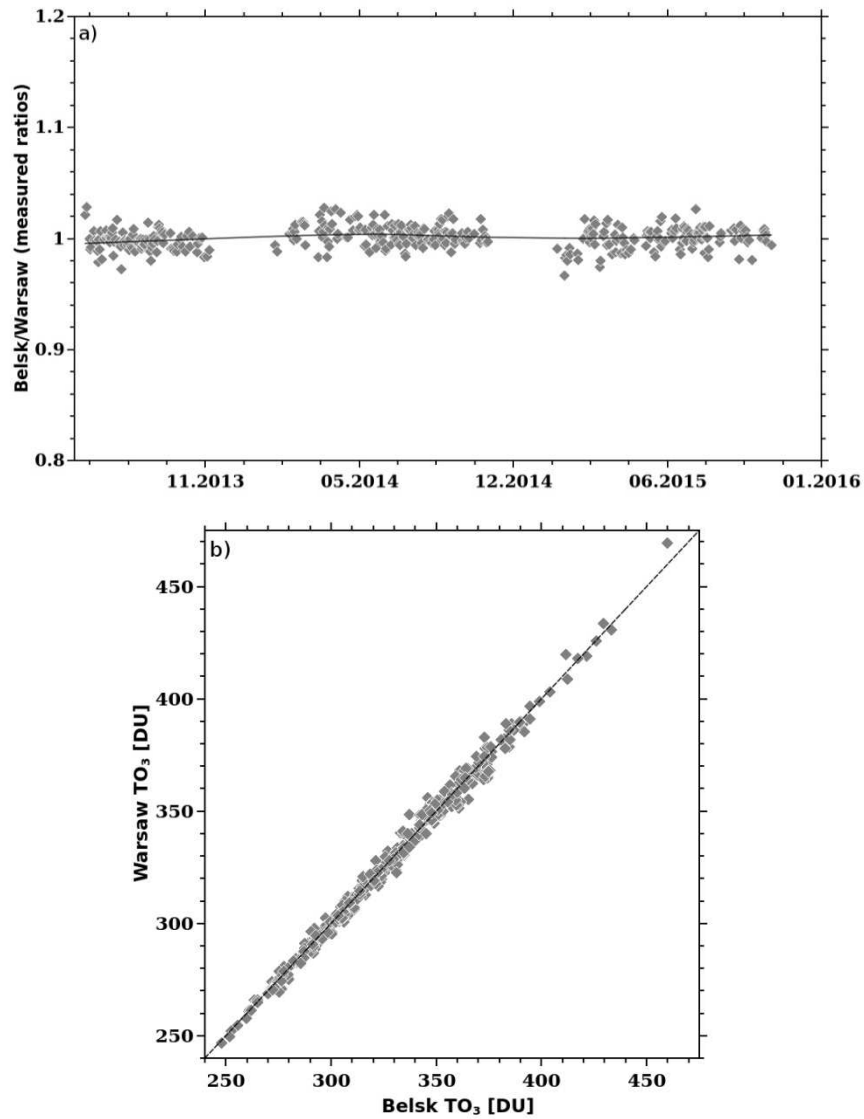
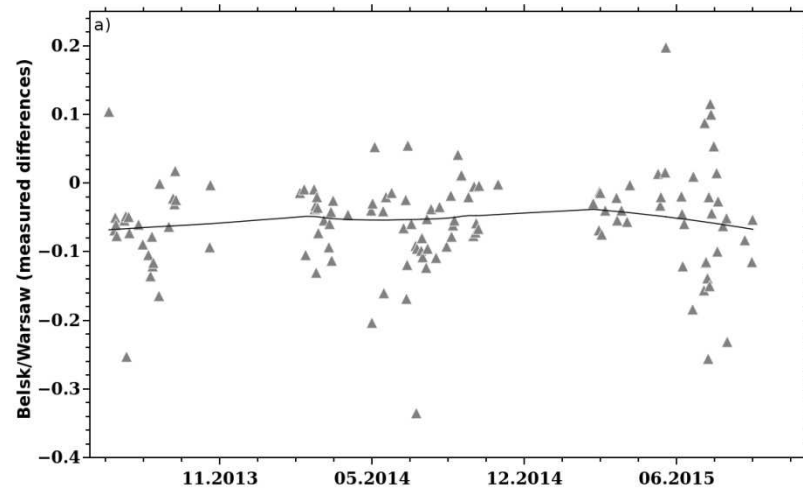
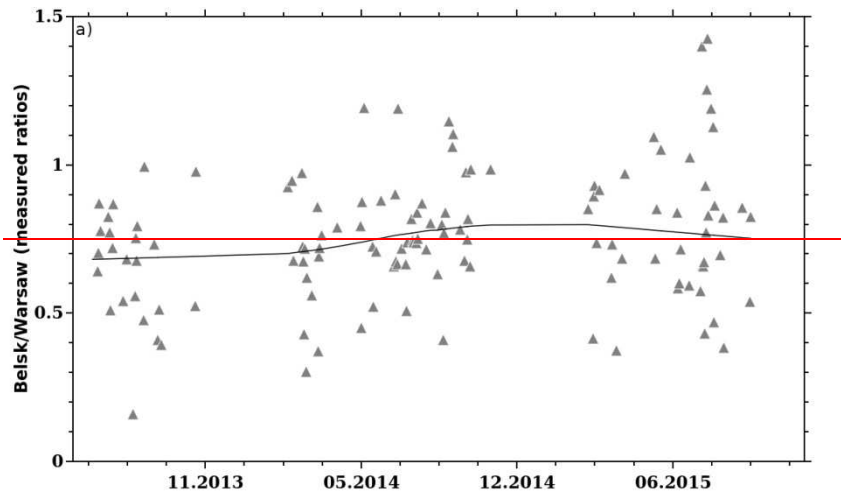


Figure 7a: The ratio between total ozone values measured by the Brewer Spectrophotometer No. 64 and No. 207 while working simultaneously at Belsk and in Warsaw for the period May 2013-December 2015. The solid curve represents the smoothed data by LOWESS filter.

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Figure 7b: Scatter plot of total ozone values measured by the Brewer Spectrophotometers No. 207 and No. 064 while working simultaneously at Belsk and in Warsaw from May 2013 to December 2015.



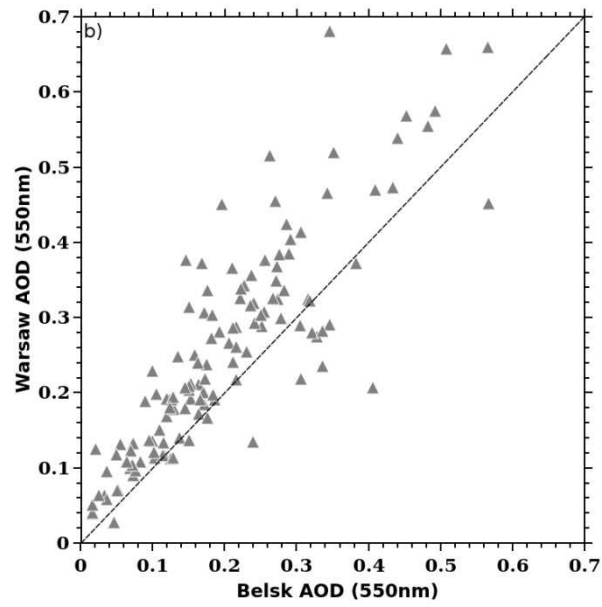


Figure 8a: The ratio difference between AOD at 550nm measured simultaneously by MODIS over Belsk and Warsaw in the period May 2013-December 2015. The solid curve represents the smoothed data by LOWESS filter.

Figure 8b: Scatter plot of AOD at 550nm measured by MODIS over Belsk and Warsaw from May 2013 to December 2015.