Atmos. Chem. Phys., 12, 2469–2477, 2012 www.atmos-chem-phys.net/12/2469/2012/ doi:10.5194/acp-12-2469-2012 © Author(s) 2012. CC Attribution 3.0 License.





Evidence of a possible turning point in solar UV-B over Canada, Europe and Japan

C. S. Zerefos^{1,2,4}, K. Tourpali³, K. Eleftheratos^{1,4}, S. Kazadzis⁵, C. Meleti³, U. Feister⁶, T. Koskela⁷, and A. Heikkilä⁷

¹Biomedical Research Foundation of the Academy of Athens, Athens, Greece

²Navarino Environmental Observatory (N.E.O.), Greece

³Department of Physics, University of Thessaloniki, Greece

⁴Faculty of Geology and Geoenvironment, University of Athens, Greece

⁵Institute for Environmental Research and Sustainable Development, National Observatory of Athens, Greece

⁶Meteorological Observatory Lindenberg, German Meteorological Service, Germany

⁷Climate Change Unit, Finnish Meteorological Institute, Helsinki, Finland

Correspondence to: C. S. Zerefos (zerefos@geol.uoa.gr)

Received: 14 June 2011 – Published in Atmos. Chem. Phys. Discuss.: 24 October 2011 Revised: 29 December 2011 – Accepted: 22 January 2012 – Published: 5 March 2012

Abstract. This study examines the long-term variability of UV solar irradiances at 305 nm and 325 nm over selected sites in Canada, Europe and Japan. Site selection was restricted to the availability of the most complete UV spectroradiometric datasets during the period 1990-2011. The analysis includes the long-term variability of total ozone, aerosol optical depth and cloud fraction at the sites studied. The results, based on observations and modeling, suggest that over Canada, Europe and Japan the period under study can be divided into three sub-periods of scientific merit: the first period (1991–1994) is the period perturbed by the Pinatubo volcanic eruption, during which excess volcanic aerosol has enhanced the "conventional" amplification factor of UV-B at ground level by an additional factor that depends on solar elevation. The increase of the UV-B amplification factor is the result of enhanced scattering processes caused by the injection of huge amounts of volcanic aerosols during the perturbed period. The second period (1995-2006) is characterized by a 0.14 % yr⁻¹ increase in total ozone and an increasing trend in spectral irradiance by $0.94 \% \text{ yr}^{-1}$ at 305 nm and 0.88 \% yr^{-1} at 325 nm. That paradox was caused by the significant decline of the aerosol optical depth by more than $1 \% \text{ yr}^{-1}$ (the "brightening" effect) and the absence of any statistically significant trend in the cloud fraction. The third period (2007–2011) shows statistically significant evidence of a slowdown or even a turning point in the previously reported upward UV-B trends over Canada, Europe and Japan.

1 Introduction

The world "avoided" following the success of the Montreal Protocol, and evidence for recovery of the ozone layer over the Northern Hemisphere (Zanis et al., 2006; Chipperfield et al., 2007; Harris et al., 2008; Newman et al., 2009; Zerefos et al., 2009a; WMO, 2011) suggest that, other factors remaining constant, the observed increases of the harmful solar UV-B doses (Zerefos, 2002) are expected to have levelled off or even decreased in the last decade. The 2011 WMO/UNEP Ozone Assessment and a recent paper by den Outer et al. (2010) reported the continuation through 2005 of upward trends in UV-B over Europe. They found these upward trends to range between $+0.3 \% \text{ yr}^{-1}$ and $+0.6 \% \text{ yr}^{-1}$, of which 2/3 they attributed to the combined decrease of cloudiness and aerosol optical depth and 1/3 to the past long-term ozone change through 2005.

Trends in cloudiness and aerosols are different in different places on Earth and their long-term effects vary in the different wavelengths of the solar spectrum (Bais et al., 1993; Wild, 2009; Wild et al., 2009; Zerefos et al., 2009b; Cermak et al., 2010). Using the longest available time series of spectral UV measurements (Fioletov et al., 2001, 2004) and updating them through 2011, we provide here tentative evidence that we may have passed maximum UV-B exposure levels over various sites located at northern latitudes ranging from 25° N to 60° N. The analysis is based on 305 nm and 325 nm solar irradiances at stations with spectroradiometric data during the period 1990–2011. Our analysis is supported

			_	_
Latitude	Longitude	Data used	Instruments	Data source
58.75	-94.07	1992-2011	Brewer MKII	WOUDC
53.55	-114.1	1992-2011	Brewer MKII	WOUDC
48.78	-123.13	1990–2011	Brewer MKII, MKIV	WOUDC
43.78	-79.47	1990–2011	Brewer MKII	WOUDC
60.81	23.49	1996–2011	Brewer MKIII	FMI
52.21	14.12	1995-2011	Brewer MKIV, Spectro 320D	DWD
51.44	-0.94	1992-2011	Optronics, Bentham	WOUDC
50.18	15.83	1994–2010	Brewer MKIV	EUVDB
40.63	22.95	1992–2011	Brewer MKII, MKIII	AUTH
43.05	141.33	1991-2011	Brewer MKII, MKIII	WOUDC
36.05	140.13	1990-2011	Brewer MKII, MKIII	WOUDC
26.2	127.67	1991–2011	Brewer MKII, MKIII	WOUDC
	53.55 48.78 43.78 60.81 52.21 51.44 50.18 40.63 43.05 36.05	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	58.75 -94.07 1992–2011 Brewer MKII 53.55 -114.1 1992–2011 Brewer MKII 48.78 -123.13 1990–2011 Brewer MKII, MKIV 43.78 -79.47 1990–2011 Brewer MKII 60.81 23.49 1996–2011 Brewer MKIV 52.21 14.12 1995–2011 Brewer MKIV, Spectro 320D 51.44 -0.94 1992–2011 Optronics, Bentham 50.18 15.83 1994–2010 Brewer MKIV 40.63 22.95 1992–2011 Brewer MKII, MKIII 43.05 141.33 1991–2011 Brewer MKII, MKIII 36.05 140.13 1990–2011 Brewer MKII, MKIII

 Table 1. Mid-latitude stations with long series of accessible spectral UV data analysed in this study.

by observations of other atmospheric variables affecting the transmittance of UV solar irradiance at the Earth's surface, namely, clouds, ozone and aerosols.

The basic idea to perform the present work came from the observation of different trends (positive for columnar ozone, negative for the aerosols and no trends in cloudiness) at the sites studied during 1995–2011. In the absence of any significant trends in aerosols and clouds, the recovery of columnar ozone would result in a decrease in UV-B solar irradiances. However, as is shown in the analysis that follows, UV-B solar irradiances continued their positive trends through about 2005, after which they have levelled-off and then seem to have reversed their upward trend. This can only be explained if we consider the opposite effects the aerosols and ozone trends have on UV-B change, since the observed trend in cloudiness is insignificant. Indeed, we show here evidence that the aerosol decline (brightening effect) has offset the effect of the ozone upward trend on UV-B from 1995 to about 2006. Since 2007, the continued upward trend of ozone dominated and overwhelmed the opposing trend imposed by the aerosol decline.

2 Data sources and methodology

The longest and most complete available time series of UV spectroradiometric dataset in the 20-yr period of study (1990–2011) were used throughout this work at 12 sites listed in Table 1. Daily UV solar irradiances at 305 nm and 325 nm and total column ozone data for stations in Canada and Japan were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) operated by Environment Canada (www.woudc.org). Daily UV solar irradi-

ance data for Europe were obtained from the European UV Database (http://www.ozone.fmi.fi/uvdb). Monthly means from daily UV solar irradiances were calculated at each station only if 14 daily summaries were available for each month.

In addition to spectral UV solar irradiances and total ozone, this study includes the analysis of time series of aerosol optical depth (AOD) at 550 nm and the cloud fraction from satellite data for the period 1990–2011. In the case of AOD, two overlapping datasets have been used: (1) for the period 1990–2006, the NASA/GACP, Global Aerosol Climatology Project (Mishchenko et al., 2007a); and (2) for the period 2000–2011, the Terra AOD experiment (MODIS) from the MODerate-resolution Imaging Spectroradiometer (Levy et al., 2007). The MODIS/Terra AOD data were taken at a spatial resolution of $1^{\circ} \times 1^{\circ}$ around each of the ground based monitoring stations, listed in Table 1.

The relation between AOD monthly values at 550 nm from MODIS and at 320 nm from ground-based measurements was next checked for correlation in their long-term variability. Unfortunately we had common AOD ground-based data at 320 nm and from MODIS at 550 nm, with sufficient detail, only at the stations of Thessaloniki 40° N and Lindenberg 52° N. These datasets have been correlated on different time scales. Both on short and on longer time scales the ground-based AOD at 320 nm were highly correlated $(r \approx +0.8)$ with the satellite AOD at 550 nm (see Supplement Fig. S1). Another issue that should be noted here is that NASA/GACP AOD data at 550 nm are provided over areas near large water bodies such as the oceans, seas and the lakes, where the surface reflectance is often low. In this work we have used $2.5^{\circ} \times 2.5^{\circ}$ square degree resolution at the eight out of the twelve spectroradiometric stations

C. S. Zerefos et al.: Evidence of a possible turning point

located near water bodies. A $5^{\circ} \times 5^{\circ}$ sq. deg. resolution was used for the remaining four stations. Statistical comparisons of NASA/GACP AOD at 550 nm with ship-borne sun-photometer AOD data at 550 nm have shown significant correlations. It has been found that the ensemble-averaged NASA/GACP AOD at 550 nm overestimates the ensembleaveraged sun-photometer data only by about 3.6% with a random error of about 0.04 (Mishchenko et al., 2007b and references therein). In order to test the accuracy of the time series created at each station from the NASA/GACP data, the corresponding MODIS/Terra data for the overlapping period (2000–2006) have been used. The calculated correlation coefficients between the common-period datasets are significant at the 99% confidence level (+0.56 for stations in Canada, +0.60 for stations in Europe, +0.59 for stations in Japan, and +0.76 for the ensemble of all stations).

Similarly, cloud fraction data for the period 1990–2006 were taken from the NASA International Satellite Cloud Climatology Project (ISCCP), (Rossow and Schiffer, 1999) and for the period 2000–2011 the cloud fraction was taken from the MODIS/Terra dataset. Both datasets are consistent with each other, since the deseasonalized monthly time series of cloud fraction from ISCCP and from MODIS during their common period 2000–2006, are highly correlated ($r \approx +0.6$, confidence level >99.9%). The correlation coefficients calculated separately for the different geographical regions of the study were found to be also highly significant (+0.6 for Canada, +0.7 for Europe, and +0.7 for Japan).

All parameters used in this study have been deseasonalized and averaged as follows: first, we deseasonalized the monthly mean data at each station by subtracting the longterm monthly mean pertaining to the same calendar month. Next, we calculated the averages over each geographical region separately (Canada, Europe and Japan) by averaging the deseasonalized data of the stations belonging to each region. Finally, the overall deseasonalized averages of all stations were obtained by averaging the three regional deseasonalized averages over Canada, Europe and Japan.

The procedure described by Reinsel (2002) and Newchurch et al. (2003), commonly used to investigate trends in ozone time series, was used in this work. We have applied here a statistical linear trend model, the generalized form of which can be described as the sum of a monthly trend, the QBO effect, the solar cycle effect, the cloud effect and a noise term as shown in Eq. (1):

$$Y_t = \alpha \operatorname{Tr}_t + \beta_1 Z_{1;t} + \beta_2 Z_{2;t} + \beta_3 Z_{3;t} + N_t$$
(1)

where, Y_t stands for the monthly deseasonalized UV data, Tr_t the monthly linear trend. $Z_{1;t}$ a proxy time series used to describe the QBO effect. Here we use 2 terms, namely the averaged equatorial zonal winds at 30 and 50 hPa. The term $Z_{2;t}$ accounts for the effect of the 11-year solar cycle, using the time series of the F10.7cm solar radio flux density as proxy, and the term $Z_{3;t}$. is a proxy time series to describe the cloud fraction effect. N_t is the unexplained noise term. The statistical model is autoregressive AR(1), and the term N_t satisfies Eq. (2):

$$N_t = \phi N_{t-1} + \varepsilon_t. \tag{2}$$

in which the ε_t are the residuals used to compute the cumulative sums of residuals (plotted in Fig. 3), after removing the autoregressive component ϕN_{t-1} .

In order to derive the proxy time series for the cloud fraction, we have used the deseasonalised cloud fraction time series from the NASA/ISCCP and from the MODIS/Terra datasets described above. The time series were tested for homogeneity during their common period of records (2000-2006). No systematic discontinuities were found, and the correlation between NASA/ISCCP and MODIS/Terra deseasonalized time series was highly significant ($r \approx +0.6$). The NASA/ISCCP time series was adjusted to the MODIS/Terra, taking into account their correlation and the standard deviations. From 2000 on, the MODIS/Terra series was used until the end of September 2011, so that a unique time series for the cloud fraction over the period 1995-2011 was constructed and used in Eq. (1) as a proxy time series for the cloud effect. This was done separately for every region and finally to the average of all three regions, so in total we got four cloud fraction proxy time series (Europe, Canada, Japan as well as their average).

The statistical analysis using Eqs. (1) and (2) was applied to the time series of 305 nm and 325 nm, using the deseasonalised UV time series for the regions of Europe, Canada, Japan and their average. We have used in this statistical model as cloud fraction proxy time series the appropriate regional and average time series. The aerosol optical depth has not been used in this statistical model.

3 Results and discussion

Figure 1 shows the deseasonalized departures (in %) of total ozone, 305 nm and 325 nm UV solar irradiances, the AOD at 550 nm and the cloud fraction, averaged over all stations. On the top of Fig. 1, monthly mean zonal winds at 30 hPa and 50 hPa, were averaged and plotted to represent the QBO. The gray vertical zone sweeping the composite time series was drawn to indicate the period when the Pinatubo eruption had significant effects to both the total ozone and the AOD values (Granier and Brasseur, 1992; Zerefos et al., 1994). Noteworthy here is the perturbation introduced by the Pinatubo aerosol in the well-known anti-correlation between UV-B and total ozone (Zerefos, 2002).

During the period of study (January 1990 through September 2011), the long-term variability of total ozone and AOD underwent important interannual fluctuations, while the corresponding variability of cloud fraction had no significant trend. It is well known that these three parameters and their long-term changes are the decisive factors in any attempt to

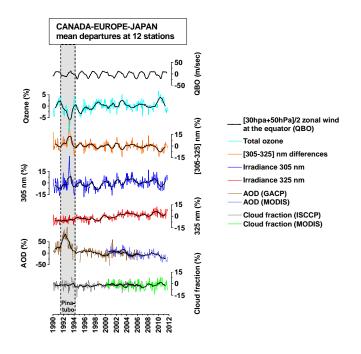


Fig. 1. Changes in (%) in total ozone, UV irradiances at 305 and 325 nm, AOD at 550 nm and cloud fraction averaged over 12 stations located between 25° N and 60° N. The third time series shows the long-term variability of the difference (in %) between 305 nm and 325 nm solar spectral irradiance. Solid black curves are 12-month running means.

understand the interannual variability of solar ultraviolet radiation. However, notable statistically significant interannual changes in total ozone and AOD can be seen, during the volcanically perturbed period from the Pinatubo eruption and the post-volcanic period to present (1995–2011). It is interesting to note the extreme values of AOD at 550 nm during the volcanically perturbed period and its decreasing trend in the post Pinatubo period. This long term decreasing trend of AOD at 550 nm (brightening effect) is characteristic at all stations under study.

The post-Pinatubo opposite long-term variability of total ozone and AOD, provided us with the unique opportunity to search for any possible leveling of UV-B reaching ground level, as would be expected from the observed positive trend of total ozone as seen in Fig. 1. If AOD and cloud fraction had no trend, UV-B should have started its leveling off and its decrease in time due to the total ozone increase. However, although the cloud fraction remained statistically unchanged, AOD with its monotonous decreasing trend has had the effect of delaying the leveling or even decreasing UV-B due to the observed ozone upward trend, up to the last few years of the record. The above discussion is supported by the third

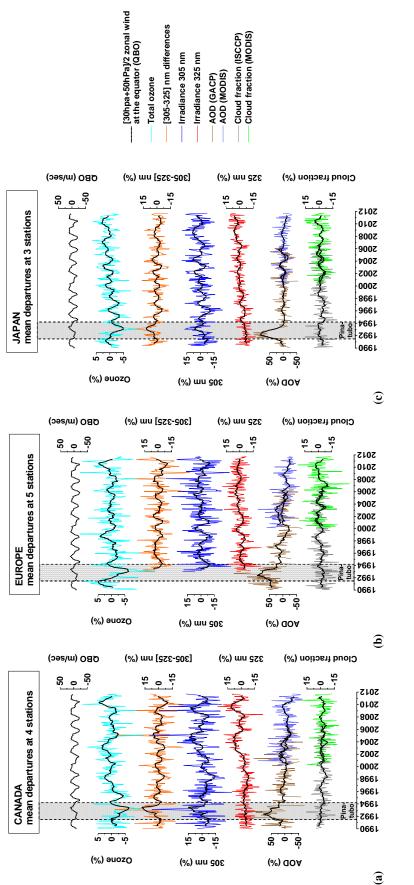
C. S. Zerefos et al.: Evidence of a possible turning point

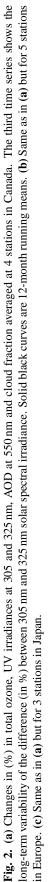
time series in Fig. 1, which shows the long-term variability of the difference (in per cent) between the 305 nm and the 325 nm spectral solar irradiances which is dependent mostly on ozone. The anticorrelation between total ozone and the (305 nm–325 nm) irradiance difference is highly significant, exceeding 0.8 both on short and on longer time scales. This high anticorrelation emphasizes the role that AOD trends have played in delaying the long-term UV-B "recovery" over about half of the Northern Hemisphere. We emphasize that the (305 nm–325 nm) difference is about free from the combined effects of changes in aerosol and cloud fraction.

Trying to quantify these arguments, we have calculated trend estimates and tested them for significance, based on Eq. (1) for the period from January 1995 through September 2011. We note here that the time series for cloud fraction and QBO do not have any significant trends throughout the past 20-yr period. If these terms are used in Eq. (1), the results of 305 nm and 325 nm UV irradiances averaged over all stations, display increasing rates of $(0.37 \% \pm 0.05 \%)$ yr⁻¹ and $(0.55 \% \pm 0.03 \%)$ yr⁻¹, respectively, while the average ozone levels increased at the rate of $(0.1 \% \pm 0.02 \%)$ yr⁻¹. During the same period, AOD levels appear to decline at rates exceeding 1 % yr⁻¹ (see also Table 2).

To examine whether the changes seen in Fig. 1 are representative for the stations in all examined geographical regions and not an artifact of the data processing, we have separated the stations in the different regions under study and calculated the averages individually for Canada, Europe and Japan. The results for the individual regions are presented in Fig. 2, respectively. As can be seen, the trends in the examined parameters are similar in all geographical regions under study. Table 3 summarizes the observed changes for the regions of Canada, Europe and Japan, separately, which are found to be in agreement with the trends shown in Table 2.

Because of the importance of the observed UV changes in the last years of the record, we have further examined the rate of change in UV irradiances following the statistical trend analysis described in Sect. 2 (Eqs. 1 and 2). The period from Jan 1995 through Dec 2005 has been used as the base period for the estimation of linear trends and the coefficients that correspond to each of the terms of the statistical model described before. Using these coefficients (estimated from the 1/1995–12/2005 fit), we have calculated from Eq. (2) the residual series N_t and subsequently the ε_t , extending the series beyond 2005 for the full period until the end of the record (9/2011). These residual time series (Fig. 3, left panels) are filtered from QBO, solar cycle and cloud fraction effects, based on their linear relationship with the deseasonalised UV time series.





(a) 305nm

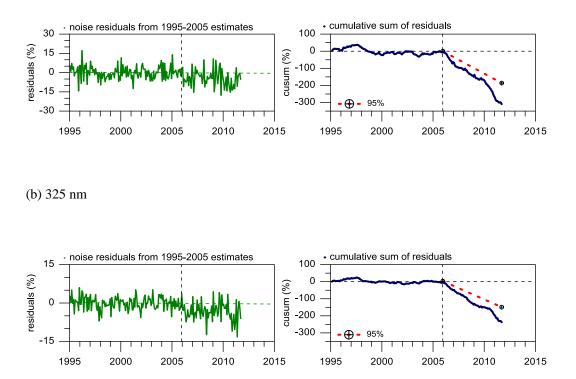


Fig. 3. White noise residuals (left panels) and their CUSUMs (ε_t , right panels) for all stations average. The red dotted line indicates the negative part of the 95% confidence envelope of departures from natural variability and model uncertainty.

Table 2. Amplitude (i.e., $(\max-\min)/2$) of atmospheric parameters during 1991–1994 (volcanic period) and during 1995–2011 (post-volcanic period). The right column shows the trends in per cent per year averaged at the 12 stations. Values in brackets refer to statistical significance of each trend.

12 stations average	Amplitude (%)		Trend per year (%)	
	1991–1994	1995–2011	1995–2011	
Ozone	7.9	5.7	$+0.10 \pm 0.02$ (99 %)	
305 nm	25.7	17.5	$+0.37 \pm 0.05$ (99 %)	
325 nm	6.8	11.7	$+0.55\pm0.03~(99~\%)$	Period
AOD (GACP)	71.4	56.2	-1.00 ± 0.39 (95 %)	1991-2006
AOD (MODIS)	-	37.8	-2.52 ± 0.32 (99%)	2000-2011
Clouds (ISCCP)	13.1	9.1	+0.10 ± 0.08 (-)	1991–2006
Clouds (MODIS)	-	10.9	$+0.01\pm0.11$ (-)	2000-2011

The 95 % confidence limits due to the statistical model uncertainty and the unexplained noise (due to unresolved fluctuations) have been calculated according to Newchurch et al. (2003, their appendix B). The cumulative sum of residuals (CUSUM) procedure can assess the systematic departure of UV (at either wavelength) computed from the trend line calculated for 1995–2005 and extended after 2005 (i.e. linear trend forecast). A negative CUSUM indicates a smaller change in the UV increasing rate in comparison to the base period. Statistical significance can be assessed by comparison to the 95 % confidence limits as presented in Fig. 3.

For the UV 305 nm departures (upper panels in Fig. 3), the station average shows large negative CUSUM of residuals, which exceeds the 95 % confidence limit, thus indicating that the positive trend seen in 305 nm UV records until the end of the 2005 (0.94 % \pm 0.07 % yr⁻¹) began reversing since about

	Post-pinatubo trends in (%) yr ⁻¹				
	Canada (4 stations)	Europe (5 stations)	Japan (3 stations)		
Ozone	$+0.09 \pm 0.03$ (99 %)	$+0.09 \pm 0.03 \ (95 \ \%)$	$+0.19\pm0.03~(99~\%)$		
305 nm	$+0.55\pm0.10$ (99%)	+0.11 ± 0.08 (-)	$+0.59\pm0.07~(99~\%)$		
325 nm	$+0.72\pm0.06~(99~\%)$	$+0.34 \pm 0.04$ (99%)	$+0.67 \pm 0.03 \ (99 \ \%)$		
AOD (GACP) AOD (MODIS)	$\begin{array}{c} -1.70 \pm 0.69 \ (95 \ \%) \\ -2.55 \pm 0.71 \ (99 \ \%) \end{array}$	$\begin{array}{c} -1.66 \pm 0.60 \ (99 \ \%) \\ -4.28 \pm 0.57 \ (99 \ \%) \end{array}$	$\begin{array}{c} -0.09 \pm 0.61 \ (-) \\ -0.89 \pm 0.42 \ (95 \ \%) \end{array}$		
Clouds (ISCCP) Clouds (MODIS)	$+0.02 \pm 0.12$ (-) -0.08 ± 0.18 (-)	-0.03 ± 0.17 (-) -0.06 ± 0.24 (-)	$+0.24 \pm 0.14$ (-) $+0.18 \pm 0.17$ (-)		

Table 3. Trends (in % per year) over Canada, Europe and Japan. Values in brackets refer to statistical significance of each trend.

2007. Similar results were found if the regions of Canada and Europe are examined separately, while for Japan we found barely significant results at the 95 % confidence limit. In the case of 325 nm departures (bottom panels in Fig. 3), the station average shows also large negative CUSUM of residuals, exceeding the 95 % cl., indicating a slowdown of the trend of the previous period (0.88 % \pm 0.04 % yr⁻¹ for 1995–2005). The same applies for the stations in the region of Canada and Europe, while in Japan there is no indication of a significant change in the trend of 325 nm irradiances.

To quantify further the hypothesis of the synergy of solar brightening and ozone upward trends to the overall UV-B variation, we have applied radiative transfer model calculations using the LibRadtran package (Mayer and Kylling, 2005). The radiative transfer model was fed with the groundbased observed ozone and MODIS-AOD at 550 nm measurements to calculate irradiances at 305 nm for all stations during the period (2000–2011). The model results together with the observations are displayed in Fig. 4. On top of that figure, the significant upward trend in total ozone and the downward trend in AOD are dominant. Keeping constant the AOD to its mean value, the calculated effect of ozone on UV-B is shown by curve A. Curve C shows the effect of "brightening" at 305 nm as a result of the reduced AOD and curve B shows the calculated effect on 305 nm from the synergy of both ozone and AOD trends. The corresponding best fit modelled curve is also shown for comparison. Curves A and C are respectively best fit to their respective model calculations. All calculations were based on deseasonalized monthly means assuming cloudless conditions. The observed 305 nm monthly mean deseasonalized irradiances averaged over Canada, Europe and Japan are shown by the open circles describing the UV-B variability due to the changes in ozone, aerosols and clouds. The centre of the circle with the two arrows shown in Fig. 4 is the interception of curves A and C which probably marks the starting point of the levelling off, in the upward UV-B trend, which is followed by a decrease in the subsequent few years through September 2011. The model esti-

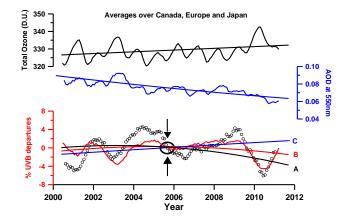


Fig. 4. Upper part: total ozone (12 month running mean) from ground-based measurements at 12 stations, Middle: average AOD from MODIS/Terra (12 month running mean) over the 12 stations. Lower part: 305 nm irradiance departures (red) calculated with the RT model, and open circles are observed 305 nm solar irradiances. Curves A, B, C are 2nd deg. polynomial fit of the calculated contribution of ozone (A), aerosol (C) and ozone and aerosol together (B) to the 305 nm irradiance variation.

mates a -1.5% to -2% UV-B decrease from 2007 to 2011 under cloudless conditions. The overall calculated effect of ozone increase on 305 nm irradiances is estimated to be on the order of -4% during 2007–2011. The corresponding effect of AOD at 305 nm irradiances was calculated to be on the order of +1.8% increase. These model calculations confirm the statistical results derived from the analysis of observations as discussed previously in connection to Fig. 1.

Before closing this paragraph we provide here some modelling results on the UV variability during the Pinatubo period. In the volcanically perturbed period seen in Fig. 1 and Table 2, we note a significant 8 % ozone loss and a disproportionately increase in 305 nm solar irradiance by about 25 %, while the 325 nm solar irradiance remained within its expected range of variability. The well-known anti-correlation between UV-B and total ozone (i.e. the amplification factor) is significantly enhanced at 305 nm (by a factor of 3) compared to any other time period in the series. We have used the LibRadtran package (Mayer and Kylling, 2005) to investigate the role played by the volcanic aerosols in explaining this enhancement. More specifically, we have calculated the UV irradiance for a constant (8%) ozone decrease and for different aerosol scenarios and various solar zenith angles. The results are solar zenith angle dependent, so they can not directly be compared with the results shown in Fig. 1. This is because data presented in Fig. 1 are monthly means of daily doses, thus representing measurements that have been performed at a variety of solar zenith angles. Our calculations show that for constant aerosol optical depth, aerosols have a larger effect on UV when they are redistributed from the troposphere to the stratosphere, combined with the 8% ozone reduction. Calculations for 45, 60 and 75 degrees of solar zenith angle showed (additional to the ozone related reduction) enhancements of 2 %, 5 % and 20 %, respectively, when a scenario of urban/extreme volcanic aerosol below/above 2 km respectively is selected, relative to the non-volcanic case.

4 Conclusions

This study examines the long-term variability of UV solar irradiances in connection to the observed upward trends in total ozone and downward trends in aerosols. The statistical analysis and model calculations have shown that the long-term variability of UV-B irradiances can be divided into three sub-periods which are characterized by different physical processes affecting the interannual variation of UV-B. The first period is the period perturbed by the Pinatubo volcanic eruption (1991-1994), for which it is shown that the excess volcanic aerosol might have enhanced by an additional 6% the "conventional" amplification factor of UV-B at ground level. The second period (1995-2006) is characterized by a UV-B increase caused by the synergy of upward ozone trend and tropospheric aerosol decline (brightening effect), during which the observed cloudiness remained without statistically significant trends. During that second period, the trend in spectral irradiance is +0.94 % yr⁻¹ at 305 nm and $+0.88 \text{ % yr}^{-1}$ at 325 nm. Model results show the synergistic effects on UV-B of the opposite trends observed in total ozone and aerosols to be in agreement with the observations. The third period, which refers to the last 5 yrs, might provide for the first time significant statistical and model-derived evidence, indicating a slowdown of the upward trends observed in the past, over the sites studied where UV-B trends seem to have undergone a turning point most probably after 2007.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/12/2469/2012/ acp-12-2469-2012-supplement.pdf.

Acknowledgements. The study was conducted within the EU-funded projects SCOUT-O3 (505390-GOCE-CT-2004) and QUANTIFY (003893-GOCE). The work was partially supported by the Navarino Environmental Observatory and the Mariolopoulos-Kanaginis Foundation for the Environmental Sciences. S.Kazadzis would like to acknowledge Marie Curie project ACI-UV, PERG05-GA-2009-247492. We acknowledge the assistance of V. Fioletov for providing the ftp link ftp://exp-studies.tor.ec.gc.ca/pub/uvdata/ to preliminary UV data for the stations in Canada after 2007. We acknowledge the MODIS mission scientists and associated NASA personnel for the production of the data used in this research effort. The NASA/GACP AOD data were obtained from the webpage http://gacp.giss.nasa.gov/. The NASA/ISCCP cloud fraction data were obtained from the webpage http://isccp.giss.nasa.gov/. The MODIS/Terra AOD and cloud fraction data were obtained from the webpage http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/ gui.cgi?instance{_}id=MODIS{_}MONTHLY{_}L3. The QBO indexes are from the CDAS Reanalysis data and are the zonally averaged winds at 30 and 50 hPa and taken from over the equator (http://www.cpc.ncep.noaa.gov/data/indices/). The F10.7cm solar radio flux density, used as proxy of the 11-year solar cycle, was acquired from the Solar and Terrestrial Physics Division (STP) of NOAA's National Geophysical Data Center (NGDC) Website (http://www.ngdc.noaa.gov/stp/stp.html).

Edited by: B. Mayer

References

- Bais, A. F., Zerefos, C. S., Meleti, C., Ziomas, I. C., and Tourpali, K.: Spectral measurements of solar UVB radiation and its relations to total ozone, SO₂, and clouds, J. Geophys. Res., 98, 5199–5204, 1993.
- Cermak, J., Wild, M., Knutti, R., Mishchenko, M. I., and Heidinger, A. K.: Consistency of global satellite-derived aerosol and cloud data sets with recent brightening observations, Geophys. Res. Lett., 37, L21704, doi:10.1029/2010GL044632, 2010.
- Chipperfield, M. P., Fioletov, V. E., Bregman, B., Burrows, J., Connor, B. J., Haigh, J. D., Harris, N. R. P., Hauchecorne, A., Hood, L. L., Kawa, S. R., Krzyscin, J. W., Logan, J. A., Muthama, N. J., Polvani, L., Randel, W. J., Sasaki, T., Staehelin, J., Stolarski, R. S., Thomason, L. W., and Zawodny, J. M.: Global ozone: past and present. Chapter 3 in Scientific Assessment of Ozone Depletion: 2006, Global Ozone Research and Monitoring Project-Report No. 50, 572 pp., World Meteorological Organization, Geneva, Switzerland, 2007.
- den Outer, P. N., Slaper, H., Kaurola, J., Lindfors, A., Kazantzidis, A., Bais, A. F., Feister, U., Junk, J., Janouch, M., and Josefsson, W.: Reconstructing of erythemal ultraviolet radiation levels in Europe for the past 4 decades, J. Geophys. Res., 115, D10102, doi:10.1029/2009JD012827, 2010.

C. S. Zerefos et al.: Evidence of a possible turning point

- Fioletov, V. E., McArthur, L. J. B., Kerr, J. B., and Wardle, D. I.: Long-term variations of UV-B irradiance over Canada estimated from Brewer observations and derived from ozone and pyranometer measurements, J. Geophys. Res., 106, 23009–23027, doi:10.1029/2001JD000367, 2001.
- Fioletov, V. E., Kimlin, M. G., Krotkov, N., McArthur, L. J. B., Kerr, J. B., Wardle, D. I., Herman, J. R., Meltzer, R., Mathews, T. W., and Kaurola, J.: UV index climatology over the United States and Canada from ground-based and satellite estimates, J. Geophys. Res., 109, D22308, doi:10.1029/2004JD004820, 2004.
- Harris, N. R. P., Kyrö, E., Staehelin, J., Brunner, D., Andersen, S.-B., Godin-Beekmann, S., Dhomse, S., Hadjinicolaou, P., Hansen, G., Isaksen, I., Jrrar, A., Karpetchko, A., Kivi, R., Knudsen, B., Krizan, P., Lastovicka, J., Maeder, J., Orsolini, Y., Pyle, J. A., Rex, M., Vanicek, K., Weber, M., Wohltmann, I., Zanis, P., and Zerefos, C.: Ozone trends at northern mid- and high latitudes – a European perspective, Ann. Geophys., 26, 1207–1220, doi:10.5194/angeo-26-1207-2008, 2008.
- Granier, C. and Brasseur, G.: Impact of Heterogeneous Chemistry on Model Predictions of Ozone Changes, J. Geophys. Res., 97, 18015–18033, 1992.
- Levy, R. C., Remer, L. A., Mattoo, S., Vermote, E. F., and Kaufman, Y. J.: Second-generation operational algorithm: Retrieval of aerosol properties over land from inversion of Moderate Resolution Imaging Spectroradiometer spectral reflectance, J. Geophys. Res., 112, D13211, doi:10.1029/2006JD007811, 2007.
- Mayer, B. and Kylling, A.: Technical note: The libRadtran software package for radiative transfer calculations – description and examples of use, Atmos. Chem. Phys., 5, 1855–1877, doi:10.5194/acp-5-1855-2005, 2005.
- Mishchenko, M. I., Geogdzhayev, I. V., Rossow, W. B., Cairns, B., Carlson, B. E., Lacis, A. A., Liu, L., and Travis, L. D.: Longterm satellite record reveals likely recent aerosol trend, Science, 315, 1543, doi:10.1126/science.1136709, 2007a.
- Mishchenko, M. I., Geogdzhayev, I. V., Cairns, B., Carlson, B. E., Chowdhary, J., Lacis, A. A., Liu, L., Rossow, W. B., Travis, L. D., Past, present, and future of global aerosol climatologies derived from satellite observations: A perspective, J. Quant. Spectrosc. Ra., 106, 325–347, doi:10.1016/j.jqsrt.2007.01.007, 2007b.
- Newchurch, M. J., Yang, E.-S., Cunnold, D. M., Reinsel, G. C., Zawodny, J. M., and Russell III, J. M.: Evidence for slowdown in stratospheric ozone loss: First stage of ozone recovery, J. Geophys. Res., 108, 4507, doi:10.1029/2003JD003471, 2003.

- Newman, P. A., Oman, L. D., Douglass, A. R., Fleming, E. L., Frith, S. M., Hurwitz, M. M., Kawa, S. R., Jackman, C. H., Krotkov, N. A., Nash, E. R., Nielsen, J. E., Pawson, S., Stolarski, R. S., and Velders, G. J. M.: What would have happened to the ozone layer if chlorofluorocarbons (CFCs) had not been regulated?, Atmos. Chem. Phys., 9, 2113–2128, doi:10.5194/acp-9-2113-2009, 2009.
- Reinsel, G. C.: Trend analysis of upper stratospheric Umkehr ozone data for evidence of turnaround, Geophys. Res. Lett., 29, 1451, doi:10.1029/2002GL014716, 2002.
- Rossow, W. B. and Schiffer, R. A.: Advances in understanding clouds from ISCCP, B. Am. Meteorol. Soc., 80, 2261–2287, 1999.
- WMO: Scientific Assessment of Ozone Depletion: 2010, Global Ozone Research and Monitoring Project – Report No. 52, 516 pp., Geneva, Switzerland, 2011.
- Wild, M.: Global dimming and brightening: A review, J. Geophys. Res., 114, D00D16, doi:10.1029/2008JD011470, 2009.
- Wild, M., Trüssel, B., Ohmura, A., Long, C. N., König-Langlo, G., Dutton, E. G., and Tsvetkov, A.: Global dimming and brightening: An update beyond 2000, J. Geophys. Res., 114, D00D13, doi:10.1029/2008JD011382, 2009.
- Zanis, P., Maillard, E., Staehelin, J., Zerefos, C., Kosmidis, E., Tourpali, K., and Wohltmann, I.: On the turnaround of stratospheric ozone trends deduced from the reevaluated Umkehr record of Arosa, Switzerland, J. Geophys. Res., 111, D22307, doi:10.1029/2005JD006886, 2006.
- Zerefos, C. S.: Long-term ozone and UV variation at Thessaloniki, Greece, Phys. Chem. Earth, 27, 455–460, 2002.
- Zerefos, C. S., Tourpali, K., and Bais, A. F.: Further studies on possible volcanic signal to the ozone layer, J. Geophys. Res., 99, 25741–25746, 1994.
- Zerefos, C. S., Contopoulos, G., and Skalkeas, G.: Twenty years of Ozone Decline. Proceedings of the Symposium for the 20th Anniversary of the Montreal Protocol, 470 pp., Springer Science + Business Media B. V., 2009a.
- Zerefos, C. S., Eleftheratos, K., Meleti, C., Kazadzis, S., Romanou, A., Ichoku, C., Tselioudis, G., and Bais, A.: Solar dimming and brightening over Thessaloniki, Greece, and Beijing, China, Tellus, 61B, 657–665, doi:10.1111/j.1600-0889.2009.00425.x, 2009b.