

In: *Solar Radiation and Human Health*
Espen Bjertness, editor.
Oslo: The Norwegian Academy of Science and Letters, 2008.

Global monitoring of atmospheric ozone and solar UV radiation

Arne Dahlback

University of Oslo, Oslo, Norway

Correspondence: Arne Dahlback, Department of Physics, University of Oslo, Norway
E-mail: Arne.Dahlback@fys.uio.no Telephone: +47 22 85 56 57

Abstract

The global monitoring network of ground-based ozone measurements consists mainly of Dobson and Brewer spectrophotometers. These instruments provide total ozone column amounts derived from measurements of direct solar UV radiation. Analysis of total ozone measurements from the global monitoring network shows a nearly linear downward trend from 1980 to mid 1990s. Data after mid 1990s indicate that global ozone is no longer decreasing. UV monitoring stations report erythemal UV irradiance levels in terms of UV indices. UV index levels up to 20 have been measured at high altitude stations located at low latitudes. Continuous reliable spectral UV measurements started in the late 1980ies, and, therefore, the existing time series for UV radiation are too short for estimations of global trends.

Introduction

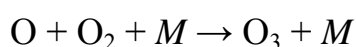
Based on photographic observations of the solar spectrum on top of Teide peak, Tenerife, Spain, Cornu concluded in 1890 that the lack of measurable radiation below 290 nm was due to strong absorption in the atmosphere (1). It was suggested by Hartley in 1880 that the atmosphere contained ozone (2). Fabry and Buisson (3) performed accurate measurements in 1913, and concluded that the amount of ozone in the atmosphere corresponded to a 3-mm-thick layer of pure ozone at standard temperature and air pressure. This corresponds to 300 Dobson Units (DU), which is the commonly used unit for the total ozone amount in the atmosphere, i.e. the amount of ozone in a vertical column from the earth's surface to the 'top' of the atmosphere. The pioneering work of Gordon. M. B. Dobson led to the construction of the well-known Dobson spectrophotometer in the 1920s for accurate measurements of total ozone. After

the discovery of the Antarctic ozone ‘hole’ in the mid 1980s the international interest in measurements of stratospheric ozone and surface UV radiation has increased. Today ozone and surface solar UV radiation is monitored by a large number of ground-based instruments, as well as by satellites.

Atmospheric ozone

Distribution of atmospheric ozone

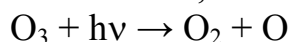
Ozone is formed and destroyed continuously in the atmosphere. Chapman proposed in 1930 a simple photochemical theory involving only oxygen (4). A three-body process forms ozone:



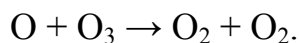
where M is another molecule, usually molecular nitrogen. Atomic oxygen, which is needed in the above process, is produced when molecular oxygen absorbs UV radiation of wavelengths below 242 nm:



Furthermore, ozone is destroyed by absorption of wavelengths below 310 nm:



as well as in a two-body collision:



The short UV wavelengths needed in reaction 1 are available mainly in the upper stratosphere in low latitude regions, and, therefore, the production of ozone is largest over the equator. However, the highest ozone columns are found at high latitudes in winter and spring, and the lowest are normally found in the tropics. This is due to a meridional circulation in the stratosphere termed the Brewer-Dobson circulation. It is characterized by rising motion in the tropical atmosphere and descending motion at high and mid latitudes (5, 6). This transport is strongest towards the winter/spring hemisphere and explains the seasonal behavior of total ozone amount at mid and high latitudes. Figure 1 illustrates the pronounced seasonal and the large day-to-day variations in total ozone abundances at a high latitude site (Oslo, Norway) and the small variations at an equatorial site (Kampala, Uganda). The large day-to-day variations observed at mid and high latitudes, particularly in winter and spring, are caused by a complicated dynamic active stratosphere and by a large number of photochemical reactions involving numerous compounds and reactions not taken into account in Chapman’s oxygen reactions.

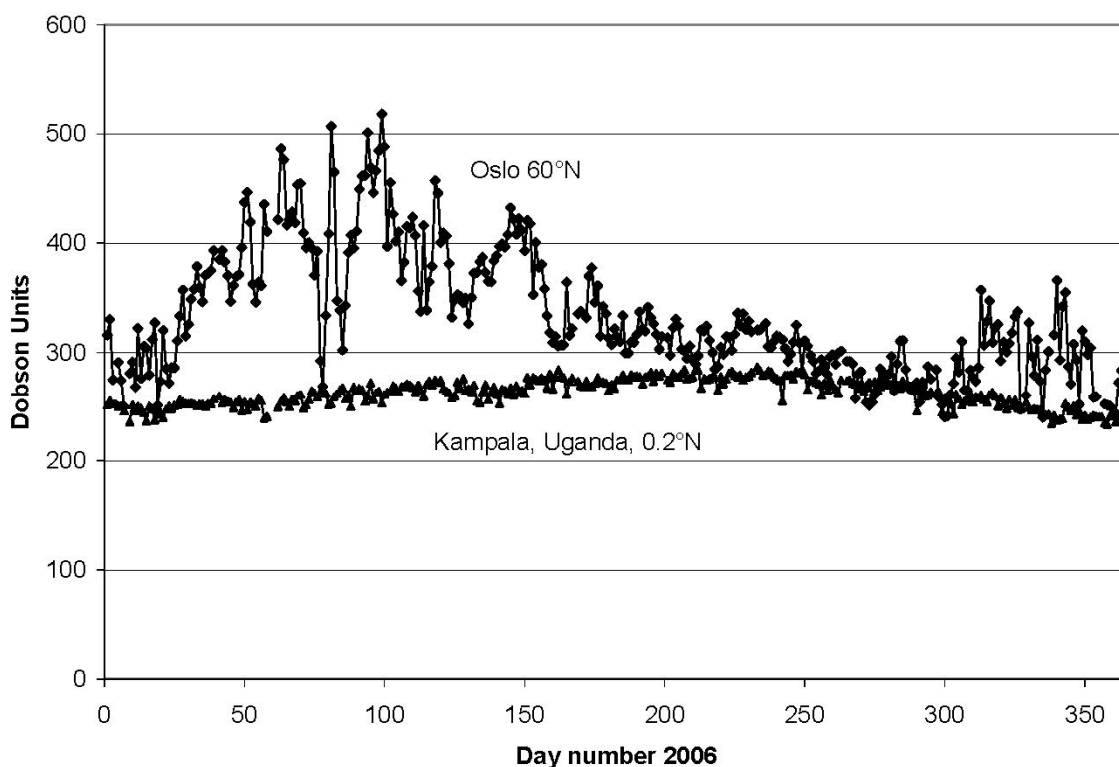


Figure 1. Satellite measurements of total ozone column amounts in Oslo (60° N) and in Kampala, Uganda (0.2° N) in 2006.

Instruments for monitoring total ozone

The Dobson instruments constitute the basis of the ground-based global monitoring network of the Global Atmospheric Watch program of the World Meteorological Organization. The global network was established in 1957, in the International Geophysical Year. The first Dobson instrument was designed in the 1920ies by G. M. B. Dobson. The longest continuous total ozone series started in 1926 in Arosa, Switzerland, and is based on measurements with Dobson instruments. The instrument is designed to measure intensity ratios of solar radiation in narrow wavelength bands in the spectral range 305 nm to 340 nm. In this wavelength range ozone is an important absorber and its absorption is strongly wavelength dependent. The measurements are most accurate when direct sunlight is used. In cloudy sky conditions zenith skylight can be used, however, with less accuracy. The Dobson instruments have proven to have long-term stability, suitable for detecting ozone changes as small as 1 % per decade. However, the instruments have several disadvantages. Calibration and routine tests are very time consuming and measurements have to be performed by a skilled operator. During the 1970s the Brewer spectrometer was developed. It is based on the same measurement principle as the Dobson. The Brewer spectrophotometer makes automated observations of the total ozone column and also provides high wavelength resolution UV spectra. Therefore, this instrument

provide high data quality with much less labor than required with the Dobson instrument. Today more than 150 Brewer instruments are used world-wide.

Total ozone can also be measured by satellite-based instruments. The most well-known satellite instrument for total ozone monitoring is the Total Ozone Mapping Spectrometer (TOMS) installed onboard several satellites since 1978. Today the Ozone Monitoring Instrument (OMI) onboard the Aura satellite launched July 2004 continues the TOMS record. Satellite measurements make it possible to map total ozone on a global basis. The daily satellite overpass over ground-based stations can be valuable for identifying sudden changes in ground-based total ozone measurements. On the other hand, the ground-based measurements are required to assess long-term calibration stability of satellite measurements.

Unfortunately, very few ground-based stations are located at low-latitudes. In recent years multi-channel filter instruments have provided accurate total ozone measurements in clear as well as in cloudy sky conditions (7, 8). These instruments are relatively cheap, have no moving parts, require little attention and are easy to operate. They are therefore suitable for use in remote places, e.g. in the tropical belt where ground-based stations are almost lacking.

Trends

Since the beginning of the 1970s there has been a discussion about atmospheric ozone depletion due to human activities. Theories about ozone depletion caused by exhaust gases from supersonic aircrafts were first introduced. The ozone discussion was further intensified after Molina and Rowland (9) and Stolarski and Cicerone (10) suggested the possibility of ozone destruction based on chlorine and bromine chemistry. This in turn stimulated a number of ozone studies and gave new impetus to the study of global ozone trends. The discovery of the Antarctic 'ozone hole', which confirmed the theories about ozone depletion caused by human release of CFCs, further increased the interest and concern about the ozone layer and the mechanisms responsible for ozone destruction (11, 12).

The amount of atmospheric ozone is determined by a number of chemical and dynamical processes. In a recent report (13) updates of global ozone trends were presented. The EESC (equivalent effective stratospheric chlorine) is an index reflecting the amount of ozone-depleting chlorine and bromine in the stratosphere. Daniel et al. proposed that the global change in total ozone amount can be assumed to be generally proportional to the EESC (14). Analysis of ground-based ozone measurements since 1964 in the latitude band 60° S to 60° N shows that the EESC time series follows the observed ozone changes for 1964 to 2005. There was a near linear downward trend in ozone changes from about

1980 to mid 1990ies. The analyses of the ozone data after the mid 1990ies indicate that the ozone amount is no longer decreasing.

Solar UV radiation

UV spectra and UV controlling factors

The ultraviolet spectrum is usually divided into three regions: UV-A (315 – 400 nm), UV-B (280 – 315 nm) and UV-C (200 – 280 nm). (Note that many authors still use the older UV-A/UV-B separation at 320 nm rather than at 315 nm.) The spectral distribution of solar UV radiation reaching the earth's surface depends on several factors: the radiation emitted by the sun, the earth-sun separation, the optical properties of the atmosphere, the solar elevation and the reflection properties of the surface.

The radiation emitted by the sun exhibits some temporal variations, like the 11-year cycle of sunspot activity and occasional flares. These variations affect mostly the shorter wavelengths of the spectrum, which are completely absorbed by the atmosphere and therefore do not reach the earth's surface.

Due to the elliptical shape of the earth's orbit around the sun, the irradiance of the extraterrestrial solar radiation varies with time of the year. The distance varies by 3.4 % from minimum (early January) to maximum (early July). Since the irradiance depends on the square of the distance it varies by 6.9 % from minimum to maximum.

The absorption properties of the ozone molecule are strongly wavelength dependent. Even a strongly depleted ozone layer will completely absorb UV-C radiation. The absorption in the UV-B region is weaker and a fraction of this radiation reaches the earth's surface. The irradiance of the UV-B radiation at the surface is therefore strongly linked to the total ozone amount. UV-A radiation is almost unaffected by ozone variations.

The irradiance of solar UV radiation at the surface depends on scattering by air molecules (Rayleigh scattering). This scattering varies approximately inversely with fourth power of the wavelength. Rayleigh scattering theory is valid if the wavelength of the radiation is much larger than the size of the scattering particle. Due to the strong wavelength dependence of Rayleigh scattering, the ratio of diffuse to global radiation (diffuse + direct) for clear-sky conditions is larger for UV-B than for UV-A. Furthermore, the ratio also depends on solar elevation because the probability of scattering increases with the path length through the atmosphere. Radiative transfer calculations show that for a cloud-free atmosphere with a sun vertically above and a 350 DU total ozone column amount and bare ground, the fraction of diffuse radiation amounts to 43 % at

310 nm and 21 % at 380 nm. For a 35° solar elevation the corresponding percentages are 67 % at 310 nm and 36 % at 380 nm.

Changes in tropospheric cloud cover can significantly affect UV penetration due to strong scattering. This type of scattering (Mie scattering) is characterized by strong forward scattering. The wavelength dependency is weak and therefore Mie scattering affects UV-B and UV-A radiation similarly.

The fraction of the diffuse radiation received by the surface also depends strongly on the reflection properties of the surface. The reflection property of the surface can be described in terms of a surface albedo which is the ratio of reflected to incoming irradiance at the surface. Figure 2 shows ratios of spectra with three different surface albedos (50, 70 and 90 %) to a spectrum with no surface reflection (bare ground) (This is a way to show how different surface reflectivities (albedo) affect the spectral distribution of the downwelling diffuse + direct radiation).

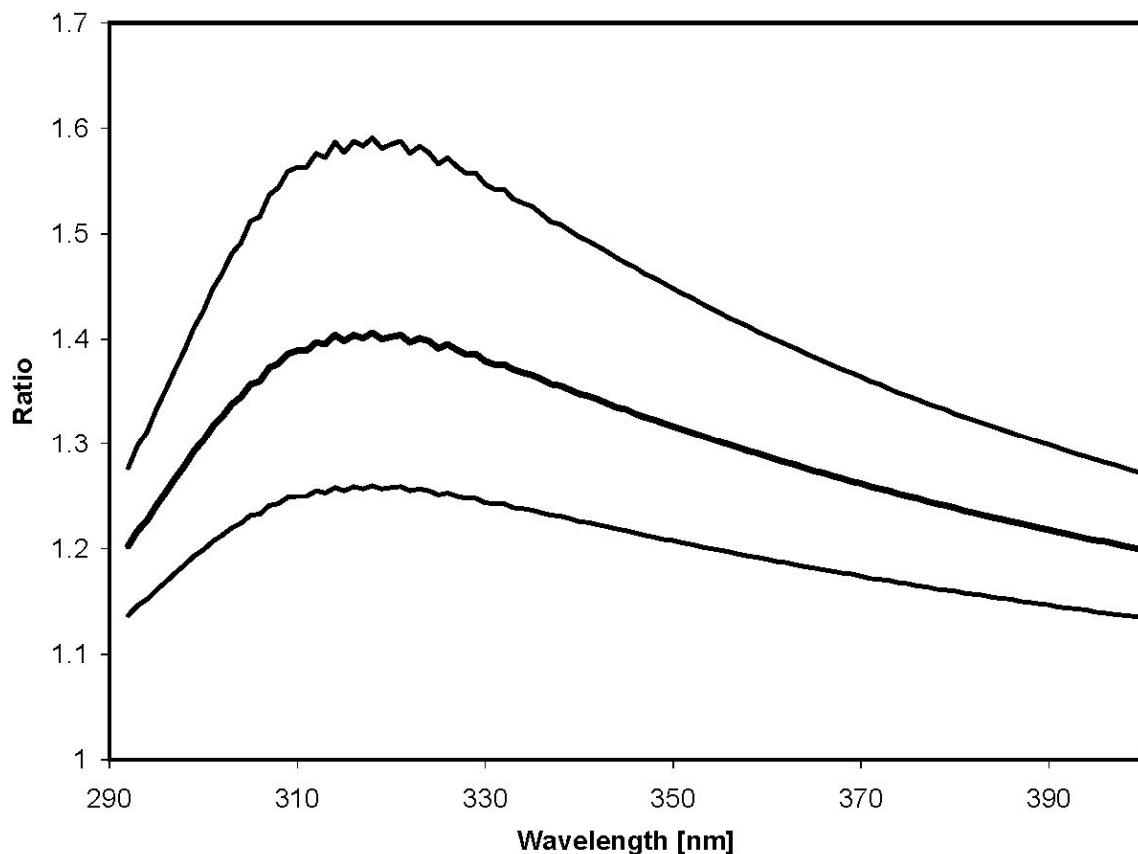


Figure 2. Effect of surface albedo on spectral surface irradiance for clear sky and 35° solar elevation based on radioactive transfer calculations. The upper, mid and low curves represent ratios of spectra with surface albedos of 90%, 70% and 50% to spectra with zero surface albedo.

UV albedos close to 100 % have been reported for new, dry snow. Older snow has generally a lower albedo than new snow (15). Relative to bare ground a 90 % surface albedo will increase the irradiance by up to 60 % around 320 nm as shown in Fig. 2. The wavelength dependence of the ratio seen in Figure 2 is due to the combined effect of wavelength-dependent Rayleigh scattering and ozone absorption.

Aerosols can have a significant effect on the transmission of UV radiation to the surface. The magnitude of the effect depends on the number of scattering and absorbing particles and their physical and chemical makeup (e.g., sulfate haze, soot, dust, sea-salt aerosols). Such particles are frequently found in the lower troposphere and are often associated with pollution. An important consideration is whether the aerosol particles are highly absorbing (e.g., soot) or simply scatter the radiation (e.g., sulfate aerosols). All particles tend to reduce the UV irradiance (the radiation falling on a horizontal surface). However, scattering by non-absorbing aerosols can actually increase the UV irradiance on non-horizontal surfaces due to additional radiation incident from low angles (16).

Instruments

The variability of surface UV irradiance is studied using measurements from ground-based instruments, as well as from estimates derived from a combination from satellite radiation measurements and radiative transfer calculations.

Spectroradiometers are still considered to be the most accurate instruments to measure UV radiation at the earth's surface. The measured quantity is usually global irradiance (direct + diffuse) at a high wavelength resolution (typically around 1 nm). Some spectroradiometers also measure actinic fluxes (radiation received by a point from all directions in space), radiance and direct irradiance. This allows more accurate determination of photolysis rates, spectral aerosol optical depths and column densities of different atmospheric constituents.

Spectroradiometers are expensive, and require extensive labor on operation, maintenance and calibration. Therefore, the global coverage is limited. Broadband radiometers are instruments that measure integrated irradiance over a large wavelength range. They typically have been designed to measure erythemal dose rates. Broadband radiometers are suitable for many applications because of their low cost and ease of operation in remote locations. However, they have limitations in terms of quality assurance and quality control.

Narrowband multi-channel filter radiometers measure UV radiation in typically 4-7 channels with bandwidths ranging from 2 to 10 nm. All channels can be measured at a high frequency, making them suitable for studies of rapid changes in spectral irradiance. Data from such instruments allow calculations of secondary data products, such as reconstructed high wavelength resolution UV

spectra, biologically weighted UV dose rates with various action spectra, total ozone abundance and cloud transmission (7), (17). For solar zenith angles less than 80° total ozone data derived from multi-channel instruments can have an accuracy similar to that of data obtained by Dobson instruments (8). These instruments are relatively inexpensive and easy to operate, which make them suitable for use in remote locations, and they are presently used in UV monitoring networks e.g., in Europe, Africa, South America and U.S.A.

Biologically weighted UV radiation

Generally, the biological effect of UV radiation depends on the wavelength. A UV dose rate D can be defined as

$$D = \int_0 A(\lambda)E(\lambda)d\lambda. \quad (2)$$

Here $A(\lambda)$ is a weighting function that describes a certain biological effect and $E(\lambda)$ is the spectral irradiance at wavelength λ . The UV dose rates are often based on the widely used CIE action spectrum (18) and represent erythemal UV dose rates. By using Eq. (2), we may express the dose rate in W/m^2 . However, the UV dose rate can also be expressed as a UV index. The UV index (UVI) is a dimensionless quantity and is defined as the erythemal UV dose rate in W/m^2 multiplied by $0.04 \text{ m}^2/\text{W}$ (19).

Fig. 3 shows daily one-hour average UVIs around local noon measured with a multi-channel filter radiometer (GUV) at the Department of Physics University of Oslo 1995-2007. The pronounced seasonal variations clearly reveal that the solar elevation is the most important UV controlling factor. Day-to-day changes in the noon UVIs are mainly caused by changing cloud cover. The ozone amount above Oslo is usually quite stable. However, particularly in spring and early summer large day-to-day variations in the ozone column amount appear quite frequently and can be important contributors to the day-to-day changes of the UVI. A summer UVI maximum value of 5-6 is typical for this latitude (60° N). UVI values of 12-14 appear frequently in tropical regions at sea level for clear-sky conditions. Even higher UVIs can occur at high altitudes. Fig. 4 shows UVI measurements from sunrise to sunset with 1 minute time resolution performed with a NILU-UV filter radiometer at Mt. Milha 5060 m above sea level in Tibet (29° N) for a ten day period in June 2003. Cloud cover was highly variable and episodes of low UVI values, even at low solar zenith angles, are due to thick clouds. The maximum measured UVI exceeded 20. This is similar to the highest UV index level reported from Mauna Loa Observatory, Hawaii at 3400 m above sea level (20). Note that the UVI exceeded 12 at solar zenith angles around 35° which is similar solar zenith angles at noon in Oslo in summer. High altitudes, low ozone-column amounts, clean atmospheres, and relatively low latitudes are factors that contribute to the high UV levels on the Tibetan plateau.

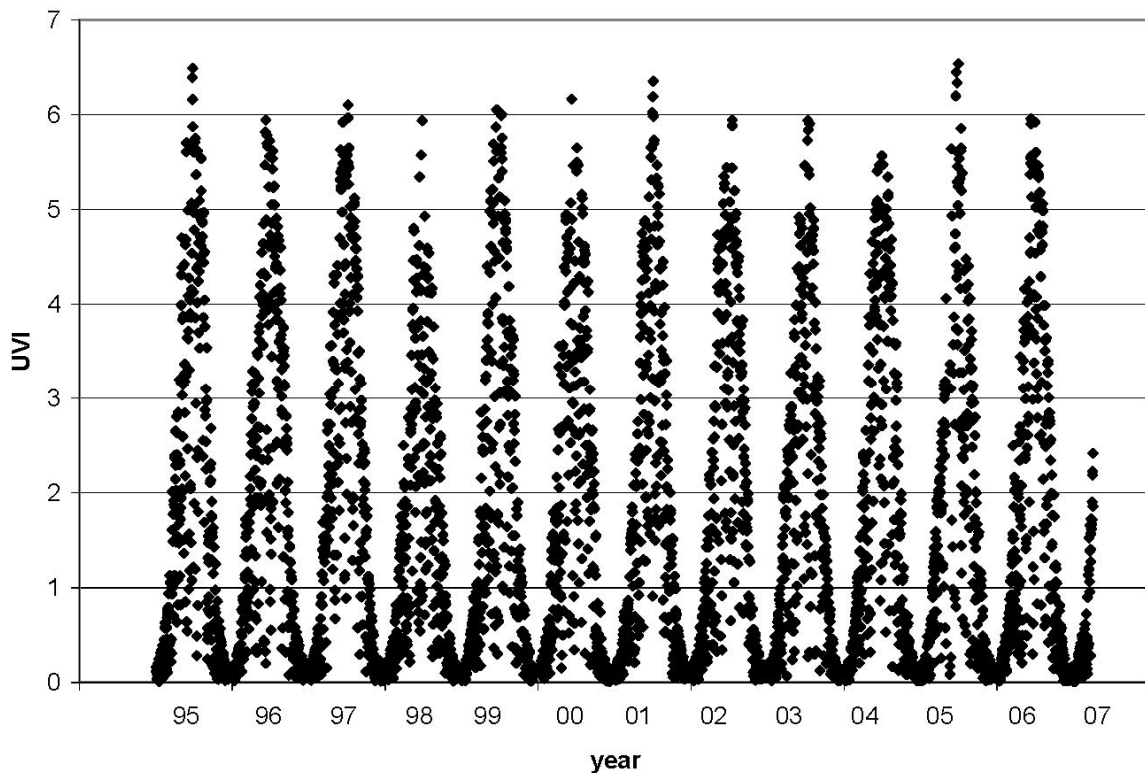


Figure 3. Daily one-hour average UVIs around local noon measured with a multi-channel filter radiometer (GUV) at the Department of Physics University of Oslo 1995-2007.

Long-term changes in surface UV

Routine surface spectral UV measurements started in the late 1980s and very few continuous records of more than 15 years exist. Long-term changes found in a particular UV record may be caused by long-term changes in a single factor or a combination of different factors, like total ozone amount, vertical distribution of ozone, cloud cover, surface albedo and aerosols. Trends in some of the factors (e.g. clouds and aerosols) affecting a UV trend in a record, may generally not be valid elsewhere. Therefore it is presently difficult to determine global trends of surface UV radiation from existing ground-based measurements. Measurements from some stations indicate that UV irradiance levels have been decreasing since the late 1990s, in accordance with observed ozone increases. However, at some stations in the Northern Hemisphere UV irradiance is still increasing, as a consequence of long-term changes in other factors that also affect UV radiation, mainly cloud cover, total ozone amount and aerosols (21).

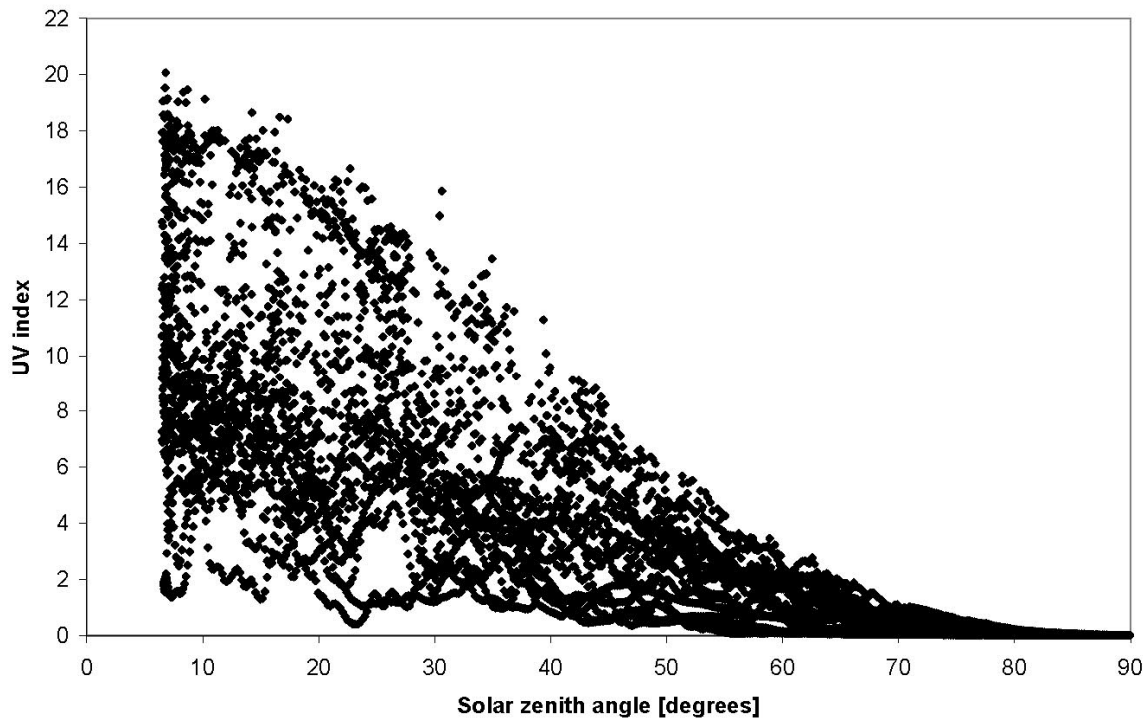


Figure 4. UVIs measured at Mt. Milha, Tibet, China, (5060 m above sea level) in the period day 177-186 2003. One-minute averages every minute from sunrise to sunset are shown.

Acknowledgements

Direct Overpass Earth Probe TOMS data were provided by NASA/GSFC.

References

1. Cornu A. Sur le limite ultra-violette du spectre solaire, d'après des clichés obtenus par M. le Dr. Simony au sommet du pic de Teneriffe. *Compt Rend* 1890; **111**: 941-947.
2. Hartley WN. On the probable absorption of solar radiation by atmospheric ozone. *Chem News* 1880; **42**, 268.
3. Fabry C and Buisson M L'absorption de l'ultraviolet par l'ozone et la limite du spectre solaire. *J Phys* 1913; **3**, 196-206.
4. Chapman S. On ozone and atomic oxygen in the upper atmosphere. *Philos Mag* 1930; **10**: 369-383.

5. Dobson GMB. Observations of the amount of ozone in the Earth's atmosphere and its relation to other geophysical conditions. *Proc R Soc London* 1930; Ser A, 129: 411.
6. Brewer AW. Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere. *Q J R Meteorol Soc* 1949; **75**: 351-363.
7. Høiskar BAK, Haugen R, Danielsen T, Kylling A, Edvardsen K, Dahlback A, Johnsen B, Blumthaler M, Schreder J. Multi-channel moderate-bandwidth filter instrument for measurement of the ozone-column amount, cloud transmittance, and ultraviolet dose rates. *Appl Opt* 2003; **42**: 3472-3479.
8. Dahlback A, Eide H, Høiskar BAK, Olsen RO, Schmidlin Fj, Tsay S-C, Stamnes K. Comparison of data for ozone amount and ultraviolet doses obtained from simultaneous measurements with various standard ultraviolet instruments. *Opt Eng* 2005; **44**(4): 041010-1-041010-9.
9. Molina MJ, Rowland FS. Stratospheric sink for chlorofluoromethanes: Chlorine atom-catalyzed destruction of ozone. *Nature* 1974; **249**, 810-812.
10. Stolarski RS, Cicerone RJ. Stratospheric chlorine: A possible sink for ozone. *Can J Chem* 1974; **52**: 1610-1615.
11. Farman JC, Gardiner BG, Shanklin JD. Large losses of total ozone in Antarctica reveal seasonal ClO_x/NO_x interaction. *Nature* 1985; **315**: 207-210.
12. Solomon S. Stratospheric ozone depletion: A review of concepts and history. *Rev Geophys* 1999; **37**: 275-316.
13. Chipperfield MP, Fioletov VE et al. 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.
14. Daniel JS, Solomon S, Albritton DL. On the evaluation of halocarbon radiative forcing and global warming potentials. *J Geophys Res* 1995; **100** (D1), 1271-1285.
15. Blumthaler M. Solar UV measurements. In: *UV-B radiation Radiation and Ozone Depletion: Effects on Humans, Animals, Plants, Microorganisms, and Materials 1993*; edited by M. Tevini, pp 71-94, Lewis, Boca Raton, Fla.
16. Blumthaler M, Ambach W, Ellinger R. Increase in solar UV radiation with altitude. *J Photochem Photobiol B: Biol* 1997; **39**, 130-134.
17. Dahlback A. Measurements of biologically effective UV doses, total ozone abundances, and cloud effects with multichannel, moderate bandwidth filter instruments. *Appl Opt* 1996; Vol **35**, No 33: 6514-6521.
18. McKinlay AF, Diffey BL. A reference action spectrum for ultraviolet induced erythema in human skin. *CIE J* 1987, **6**, 17-22.

19. WHO, 2002, Global Solar UV Index, A practical guide. Fact Sheet 271, <http://www.who.int/mediacentre/factsheet/who271/en/index.html>
20. Bodhaine BA, Dutton EG, Hofmann DJ, McKenzie RL, Johnston PV. UV measurements at Mauna Loa: July 1995 to July 1996. *J Geophys Res* 1997; **102**, 19265-19273.
21. Bais AF, Lubin D et al. Surface ultraviolet radiation: past, present and future, Chapter 7 in *Scientific Assessment of Ozone Depletion: 2006*, Global Ozone Research and Monitoring Project—Report No. 50, 572 pp., World Meteorological Organization, Geneva, Switzerland, 2007.