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Total atmospheric ozone determined from spectral measurements of direct solar UV irradiance

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Abstract. With a double monochromator, high resolution spectral measurements of direct solar UV-irradiance were performed in Arosa during February and March, 1993. Total atmospheric ozone amount is determined by fitting model calculations to the measured spectra. The results are compared with the operationally performed measurements of a Dobson and a Brewer spectrometer. The total ozone amount determined from spectral measurements differs from the results of the Dobson instrument by -1.1±0.9 % and from those of the Brewer instrument by -0.4±0.7 %.

Introduction

Stratospheric ozone protects the earth's surface from harmful solar radiation in the UVB-region. Due to the global decrease of stratospheric ozone during the last decade [Stolarski et al., 1992] there is an increasing concern in public and science about harmful biological effects of solar UVB-radiation. Generally, the global flux from the upper hemisphere through a horizontal surface is accepted as a biologically relevant parameter. Measurements with high spectral resolution permit the weighting of global irradiance with action spectra for different biological processes, for example erythemal reaction of the skin [McKinlay and Diffey, 1987] or DNA damage [Setlow, 1974].

The dependence of solar UV-irradiance on several atmospheric parameters, such as total ozone amount, aerosol content, and cloud cover, is relevant. Hence, the simultaneous determination of these parameters is very valuable for the interpretation of solar global spectra. Stamnes et al. [1991] describe a method to determine total atmospheric ozone amount from spectral measurements of global solar irradiance. However, one can expect more accurate results from spectral measurements of direct solar irradiance as modelling of direct solar irradiance introduces less uncertainties. Therefore we adapted a high resolution double monochromator for measurements of both global and direct solar UV irradiance and developed an algorithm for calculating the total ozone amount and the aerosol optical depth from direct solar UV irradiance. In this paper, we present the results of an intercomparison with Dobson and Brewer spectrometers in Arosa (Switzerland) in February and March 1993. As entirely different calibration procedures are used for the double monochromator and for the Dobson and Brewer instruments, this comparison demonstrates the accuracy of the total ozone determination from spectral measurements of direct solar irradiance and from commonly used ground based total ozone instruments.

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Instruments and site description

The field measurements took place at the Light Climatic Observatory (LKO) in Arosa (Switzerland, 1840 m a.s.l.), an ozone measuring station run by the Swiss Meteorological Institute. Total ozone observations with Dobson [Dobson, 1957] and Brewer [Kerr et al., 1985] spectrometers are based on relative measurements of direct solar irradiance at certain wavelengths. Both instruments are routinely in operation at the LKO. The Dobson instrument D101 has been calibrated against the Primary World Dobson instrument at the Dobson intercomparison 1986 in Arosa. A further Dobson intercomparison in summer 1990 in Arosa demonstrated the stability of the calibration. The Dobson instrument is the standard instrument for ground based total ozone observations in the Global Ozone Observing System (GO_3OS) of the World Meteorological Organisation [1992]. The calibration constants of the Brewer Br40 instrument were updated by a comparison with the Brewer standard instrument in summer 1993. Both Dobson and Brewer standard instruments had originally been calibrated at the Mauna Loa observatory in Hawaii using a procedure based on the Langley plot method [Komhyr et al., 1989]. If weather conditions allow it, 4-7 Dobson measurements are performed on working days at the LKO, whereas the Brewer instrument operates automatically and performs total ozone measurements every 10 min throughout the whole day.

For spectral measurements, a high resolution double monochromator with a full bandwidth of 0.86 nm at 50 % of the maximum was used. The spectra were taken from 290 nm to 350 nm with steps of 0.5 nm. One spectral scan takes about one minute during which stable atmospheric conditions are required. A teflon diffuser was used for measuring global irradiance from the upper hemisphere. For measurements of direct solar irradiance, a lens optics with a field of view of ±0.6° was mounted on a sun tracking device with an angular accuracy of ±0.1°. A rotating mechanism alternates the diffuser and the lens optics positioned in front of a quartz fibre which is connected to the monochromator. The instrument's wavelength accuracy is improved by an algorithm [Huber et al., 1993] to determine small wavelength shifts of a measured spectrum (usually <0.1 nm) from a comparison of its Fraunhofer structure between 340 nm and 350 nm with a high resolution extraterrestrial spectrum [VanHoosier et al., 1988].

The instrument is calibrated against a 1000 W standard lamp which has been ultimately calibrated at the Physikalisch-Technische Bundesanstalt in Braunschweig (Germany). Due to uncertainties in the lamp certificate and to the limited reproducibility in the position of the instrument relative to the lamp, the absolute accuracy of the calibration is limited to about ±5 %. Furthermore, slight inaccuracies in the angular position of the lens optics pointing towards the sun may cause a movement of the sun out of the direction of maximum sensitivity during the scan. This can result in a relative error <2 % over the spectral range from 290 nm to 350 nm.

Calculation of total ozone from direct solar UV irradiance

The total ozone amount and the aerosol optical depth were calculated by fitting model calculations to spectral measurements of direct solar irradiance. The model uses an extraterrestrial spectrum with a full bandwidth of 0.15 nm at 50 % of the maximum [VanHoosier et al., 1988]. It calculates the attenuation of the direct beam, which is due to ozone absorption, Rayleigh scattering, and aerosol absorption and scattering. The curved atmosphere is described by 23 layers with a thickness of 2 km up to an altitude of 26 km and more (3-20 km) up to an altitude of 100 km. Temperature, air pressure, ozone and aerosol concentrations can be selected independently for each layer, while constant conditions are assumed within the layer. For Rayleigh scattering, the cross sections given by Nicolet [1984] are used. Ozone absorption in each layer is determined by using the temperature-dependent cross sections published by Paur and Bass [1985], the temperature profile of the US standard atmosphere [Liljequist and Cehak, 1990] and an average ozone profile for midlatitudes [Iqbal, 1983].

Aerosol optical depth $\tau(\lambda)$ is assumed to have a linear wavelength dependency between 290 nm and 350 nm. Therefore, the attenuation by aerosols is described by the factor $\exp(-\mu\cdot\tau(\lambda))$ with

$$\tau(\lambda) = \tau_0 + \eta\ (\lambda\text{-}320\ \text{nm}). \qquad \text{(Eq. 1)}$$

μ is the relative airmass for aerosol, τ_0 is the aerosol optical depth at 320 nm and η describes the wavelength dependency of the aerosol optical depth. The aerosol profile is assumed to decrease linearly from sea level to an altitude of 10 km where it reaches zero.

For all wavelengths of the high resolution extraterrestrial spectrum, the extinction of direct solar irradiance by the atmosphere is calculated. For comparison with measured spectra, the calculated high resolution spectra at the measurement site are convoluted with the instrument's slit function. The calculation of total ozone amount ω and the parameters for aerosol optical depth (τ_0 and η) is performed by minimising the sum of squares of the relative deviations between calculated spectra $I_c(\lambda;\ \omega,\ \tau_0, \eta)$ and measured spectral direct solar irradiance $I_m(\lambda)$:

$$\sum_{\lambda}\left[\frac{I_m(\lambda)-I_c(\lambda;\omega,\tau_0,\eta)}{I_m(\lambda)}\right]^2 \Rightarrow \min. \qquad \text{(Eq. 2)}$$

Minimising Eq. 2 results in equal weighting of the percentile deviations, independent from the absolute intensity. This optimisation problem with three free parameters ω, τ_0 and η is numerically solved by using the Marquardt algorithm [Marquardt, 1963] which is a recursive method to resolve non-linear least squares problems: The free parameters are varied systematically until the optimisation condition (Eq. 2) is fulfilled to a certain predefined accuracy. The algorithm guarantees numerical stability through a flexible step-size control, thus preventing oscillations in the iteration process.

The parameters ω, τ_0, and η fulfilling the optimisation condition (Eq. 2) give the best model description (in a least square sense) for each measured spectrum. Fig. 1 shows the suitability of this three-parametric model to describe the direct solar irradiance from 295 nm to 350 nm. The spectra obtained from this model differ from the measurements by less than 5% with no systematic wavelength dependency. Most of the difference is due to small wavelength shifts between the extraterrestrial spectrum and the measurements and to uncertainties in the slit function and in the structure of the extraterrestrial spectrum.

The calculated ozone amount ω is hardly influenced by inaccuracies of the measured spectra of direct solar irradiance, which

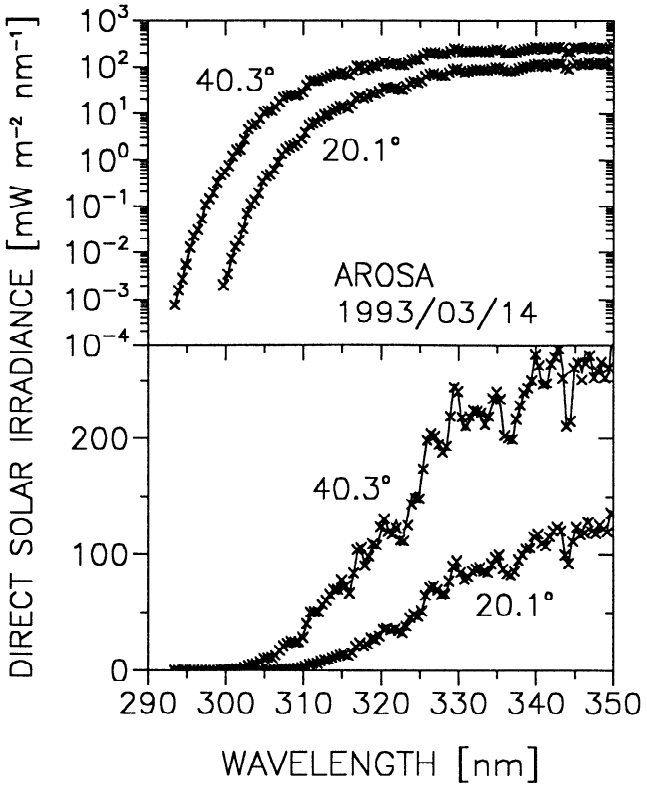

Figure 1. Comparison of UV-spectra obtained from model calculations with spectral measurements of direct solar UV-irradiance at 20.1° and 40.3° solar elevation in Arosa (Switzerland, 1840 m a.s.l.) on March 14, 1993. The crosses (x) represent the measured spectra, the solid line shows the results of the model calculation after total ozone amount ω and the parameters for description of aerosol optical depth τ_0 and η (see Eq. 2) have been optimised (20.1° solar elevation: ω = 322.2 DU, τ_0 = 0.155, η = $-3.9\cdot10^{-4}$ nm^{-1}; 40.3° solar elevation: ω = 329.8 DU, τ_0 = 0.239, η = $1.9\cdot10^{-4}$ nm^{-1}).

is clear from the description of aerosol optical depth (Eq. 1): An error by a constant factor for the whole spectrum causes solely an additive error in τ_0, an error by a factor with linear dependency on wavelength causes solely an additive error in η. Thus, ω is only influenced by errors with quadratic or even higher wavelength dependency. However, the absolute uncertainty in the instrument's calibration of ±5% for the whole spectral range causes an absolute uncertainty in τ_0 of ±0.018 at 20° solar elevation and of ±0.033 at 40° solar elevation. Similarly, an uncertainty in η of $\pm2.9\cdot10^{-4}$ nm^{-1} at 20° solar elevation and of $\pm5.5\cdot10^{-4}$ nm^{-1} at 40° solar elevation may occur due to an error changing linearly by 2% from 290 nm to 350 nm. Such an error can be caused by a slightly wrong angular position of the lens optics. At low aerosol optical depths these uncertainties may prevent a physical interpretation of η. This happens in Arosa on March 14, 1993 at 40.3° solar elevation (Fig. 1), when the calculated value of η equals to a positive value ($1.9\cdot10^{-4}$ nm^{-1}) which is not significant.

Furthermore, wrong optical pathlengths may be calculated by the model as the assumed aerosol and ozone profiles may differ from reality. Since extinction by ozone and aerosol is determined by the product of optical pathlength and ozone amount respectively aerosol optical depth, a relative error in the optical pathlength results in the same relative error (with opposite sign) in the calculated ozone amount respectively in the calculated aerosol optical depth. Therefore, the sensitivity of the results to

changes in the profile assumptions has been analysed: The assumed linear aerosol profile was altered to two extremes (all the aerosol in the lowest layer; exponential vertical aerosol distribution corresponding to air pressure). For both extreme profiles, the calculated values for the parameters τ_0 and η changed by less than 1.5% at solar elevations above 10°. The assumed ozone profile was altered to the monthly mean ozone profiles at Payerne for April and for October [Staehelin and Duetsch, 1989]. In both cases, the calculated ozone amount ω changed by less than 0.6% at solar elevations above 10°.

Aerosol optical depth at 320 nm has been compared with the broadband aerosol optical depth determined with the quartz window and the OG1 filter of a Linke-Feussner-actinometer [Kuhn, 1972]. The actinometric evaluation provides a mean value for aerosol optical depth from the short wavelength end of the solar spectrum up to 525 nm with its optical centre at 427 nm. Due to the wavelength dependency of aerosol optical depth, the spectrometer and the actinometer results are not directly comparable. Nevertheless, diurnal changes in the total amount of atmospheric aerosol are observed with both instruments as can be seen in Arosa on March 8, 1993 (Fig. 2b). A more pronounced variation took place on August 28 and August 29, 1992 in Panorama near Thessaloniki (Greece, 385 m a.s.l.), which is also well documented in both the spectrometric and the actinometric results [Blumthaler et al., 1993]. During these two days aerosol optical depth at 320 nm varied from 0.38 to 1.39, whereas the broadband optical depth, centred at 427 nm, varied simultaneously from 0.31 to 1.10. Further measurements performed in high northern latitudes (Tromsø, Norway, 150 m a.s.l.) show much lower aerosol optical depth, for example 0.14 (320 nm) and 0.11 (broadband, centred at 427 nm) for June 25, 1993. Extremely low aerosol optical depths (0.06 at 320 nm; 0.04 for broadband, centred at 427 nm) are observed at the High Alpine Research Station Jungfraujoch (Switzerland, 3576 m a.s.l., October 28, 1993). Differences in the ratio between the aerosol optical depth at 320 nm and the broadband aerosol optical depth, centred at 427 nm, indicate different types of aerosol. However, a detailed comparison between aerosol optical depth determined from spectral measurements and from filter instruments is the target of further investigations.

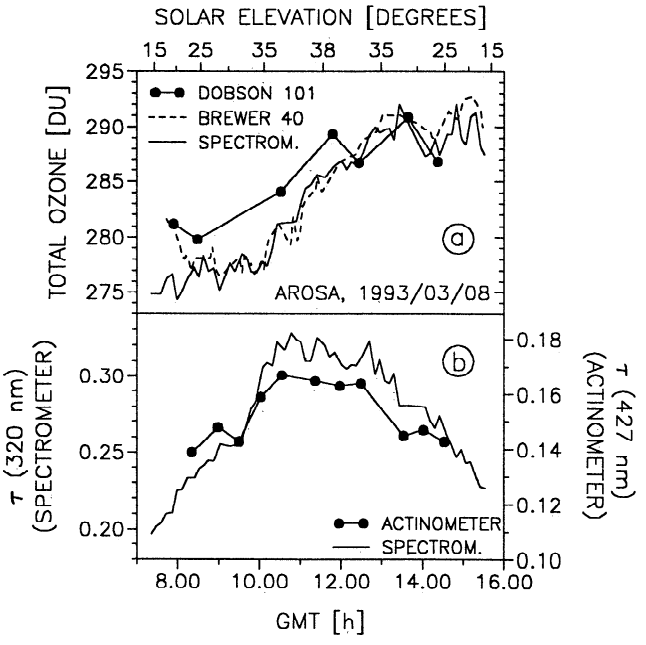

Figure 2. Diurnal variation of total ozone and aerosol optical depth on March 8, 1993 in Arosa (Switzerland, 1840 m a.s.l.).

a. total ozone determined from spectral measurements of direct solar UV-irradiance and from Dobson and Brewer measurements.

b. aerosol optical depth at 320 nm (τ_{320}) determined from spectral measurements of direct solar UV-irradiance and aerosol optical depth for a broadband filter with its optical centre at 427 nm (τ_{427}) determined from measurements of a Linke-Feussner-actinometer. A factor of 1.8 is chosen between the axes for τ_{320} and for τ_{427} in order to show the similar diurnal variation.

Comparison of ozone values

In February and March 1993, an intensive intercomparison between total ozone values determined from high resolution spectral measurements of direct solar irradiance and from the Dobson 101 (AD wavelength pairs) and the Brewer 40 instruments took place in Arosa. On 21 days, 657 spectra of direct solar irradiance were measured with completely uncovered sun and 231 spectra were taken with the solar disk uniformly covered with clouds but still visible. The latter situation occurs very often with cirrostratus clouds. Generally, cloud cover results in an extinction of direct irradiance which is approximately constant between 290 nm and 350 nm. As a consequence of the model description of aerosol extinction (Eq. 1), this constant factor influences only the parameter τ_0, whereas the calculated total ozone amount ω remains unchanged. As far as the comparison of total ozone values determined from spectral measurements of direct solar irradiance with those from Dobson and Brewer instruments is concerned, no significant differences can be seen between measurements with uncovered and with uniformly covered sun (not shown).

A typical diurnal change of total ozone measured by all three instruments is shown for March 8, 1993 (Fig. 2a). Total ozone amount determined from the spectral measurements agrees with that from the Brewer instrument within 2 DU, whereas the total ozone amount obtained from the Dobson instrument is about 5 DU high in the morning. Whereas the same temporal changes can be found for spectrometer and Brewer results, the relatively small number of Dobson measurements prevent a detailed comparison of the diurnal course.

As the total ozone amount changes throughout the day and the three instruments did not measure simultaneously, it was necessary to interpolate the spectrometer measurements linearly with time in order to get ozone values for the same time as for the Dobson and the Brewer instruments. Because of the high measurement frequency of the spectrometer (every 10-15 min), this does not introduce any significant error. Due to the less frequent sampling schedule of the Dobson observations, only 105 Dobson measurements are available for comparison, whereas there are 890 quasi-simultaneous measurements (spectrometer results interpolated) of the Brewer instrument and the spectrometer.

For the entire period of February and March 1993, the total ozone amount determined from spectral measurements of direct solar irradiance is low by 0.4±0.7% compared with the results from the Brewer instrument and by 1.1±0.9% compared with the results from the Dobson instrument. The difference to the Dobson instrument does not show any significant correlation with the total ozone amount and the solar elevation (Fig. 3). On the contrary, the difference to the Brewer instrument shows a small but significant ozone dependency of 0.81% per 100 DU and a dependency on solar elevation of 0.53% per 10°. This dependency on solar elevation cannot be caused by the different procedures for calculation of the relative airmass as they agree better than 0.3% for solar elevations >15°. Nevertheless, these systematic differences between the spectrometer and the Brewer instrument are within the statistical errors for each instantaneous measurement (spectrometer: typically ±0.5 DU; Brewer instrument: typically ±1 DU).

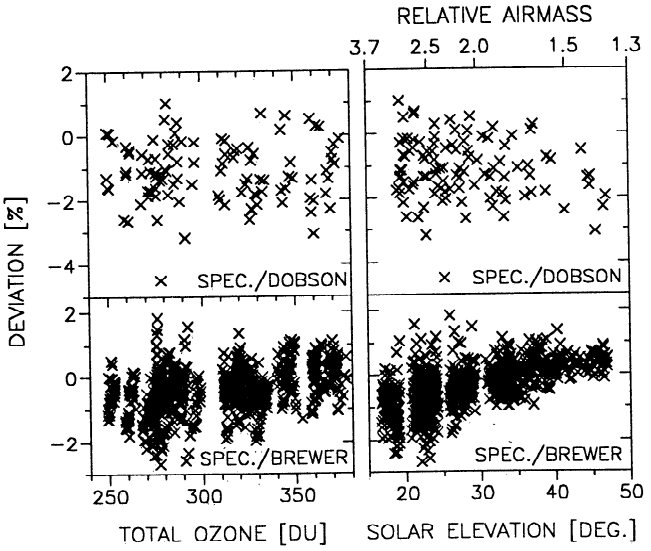

Figure 3. Deviation of instantaneous total ozone amount determined from spectral measurements of direct solar UV irradiance from Brewer and Dobson measurements as a function of total ozone and of solar elevation for all 21 days of the intercomparison period.

Whereas the spread of the total ozone from the spectrometer relative to the Dobson instrument is almost invariant with solar elevation, the spread relative to the Brewer instrument decreases strongly with increasing solar elevation. This indicates that the spectrometer and the Brewer instrument tend to show higher statistical variation at low solar elevations. However, both instruments show less statistical variation than the Dobson instrument at all solar elevations. After the intercomparison, the relatively high variation of the Dobson instrument was improved by a change in its data acquisition system in summer 1993.

Conclusion

Spectral measurements of direct solar UV irradiance permit the determination of total ozone amount and aerosol optical depth quite satisfactory. As these two parameters are influencing global UV irradiance and are therefore relevant for biologically effective doses, they should be determined simultaneously when measuring solar UV-radiation.

The results of the total ozone comparison is encouraging because total ozone amount has been calculated both based on the Langley plot method as well as based on high resolution spectral measurements, an extraterrestrial spectrum and a transparency model of the atmosphere. The results cover a wide range of total ozone amount and airmass. The very satisfactory agreement confirms that properly calibrated and maintained instruments are able to measure total ozone with an accuracy of 1 % in routine operation.

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