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Ozone variability in the upper stratosphere during the declining phase of the solar cycle 22

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Abstract. Recent studies of the solar cycle variation of ozone have shown that the response of ozone in the upper stratosphere to solar UV variation, as inferred from the SBUV (Solar Backscatter Ultraviolet) type measurements, is about a factor of two greater than estimated from 2-D photochemical models. Because of potential errors in accounting for the long term instrument drift in the SBUV type of measurements, the significance of this discrepancy is difficult to quantify. In this paper, ozone measurements from the Microwave Limb Sounder (MLS) and the solar irradiance measurements from the Solar Stellar Irradiance Comparison Experiment (SOLSTICE) and the Solar Ultraviolet Irradiance Monitor (SUSIM) onboard the Upper Atmosphere Research Satellite (UARS) are analyzed to estimate the upper stratosphere ozone response to changes in the solar UV irradiance. During the three year period of UARS measurements, analyzed here for the declining phase of the solar cycle 22, the solar irradiance in the 200-205 nm range decreased by about 5 % from a near solar maximum to a near solar minimum level. During the same period, ozone mixing ratio measured from the MLS instrument decreased by about 2-4 % in the 0.7-3 hPa region. In the upper stratosphere, the general characteristics of the MLS time series are similar to those inferred from the NOAA-11 SBUV/2 measurements. The SBUV/2 trends above 1.5 hPa, however, are significantly greater than those derived from the MLS data. The UARS data suggest that the long term solar UV response of ozone in the upper stratosphere is underestimated by 2-D photochemical models as in previous studies based on the SBUV type measurements.

Introduction

Recent studies of the solar cycle variation of ozone [*Hood et al.*, 1993; *Chandra and McPeters*, 1994, to be referred as CM 94] have shown that the response of ozone in the upper stratosphere to solar UV variation, as inferred from the SBUV type measurements, is significantly greater than estimated from 2-D photochemical models [*Garcia et al.*, 1984; *Brasseur, 1993* and the references therein; *Huang and Brasseur*, 1993; *Fleming et al.*, 1995]. For example, in CM94, the variation in ozone mixing ratio, based on the 15 years of the combined version 6 data from the Nimbus-7 SBUV and the NOAA-11 SBUV/2 instruments, is estimated to be about 6-7 % at 2 hPa over an 11 year solar cycle. This is a factor of 2 to 3 greater than the values estimated from 2-D models. Similar discrepancies between model calculations and observations are also observed in the ozone-UV sensitivity S_{uv}, defined as the percentage change in ozone for 1 % change in the solar UV flux in the 200-205 nm band. This wavelength region primarily contributes to the production of ozone in the stratosphere through photodissociation of O_2 and subsequent recombination of O and O_2 in the presence of a third body. The value of S_{uv} at 2 hPa, estimated from the combined Nimbus-7 SBUV and NOAA-11 SBUV/2 data, is close to unity and is a factor of 2-3 greater than the model estimates [*Brasseur, 1993*; *Fleming et al.*, 1995]. It is also a factor of 2-3 greater than the S_{uv} values estimated from the solar ozone relationship on time scales of a solar rotation [CM 94]. In the upper stratosphere, where photochemical time constants are of the order of a day, S_{uv} should not change significantly on time scales ranging from the 27 day solar rotation period to the 11 year solar cycle.

The discrepancies between the observed and model response of ozone to the 11 year solar cycle modulation of UV may be attributed to one or more of the following: (1) the deficiencies in the version 6 algorithm that estimates the instrument drift in the SBUV and SBUV/2 spectrometers, (2) incorrect characterization of the solar cycle component of the UV irradiance based on the MgII index and the scale factor SF (defined as the % change in Solar UV irradiance for 1 % change in MgII index), and (3) deficiencies in 2-D models related to ozone photochemistry and dynamics in the upper stratosphere. For example, the model may be deficient in simulating the dynamics of the upper stratosphere [*Hood et al.*, 1993] which could affect the water vapor concentration and indirectly ozone. The long term calibration of both the SBUV and the SBUV/2 instruments is affected by the degradation of the diffuser plates exposed to the solar UV irradiance. For the version 6 SBUV data, the long term calibration for the total ozone wavelengths was established using a 'pair justification' technique while the calibration of the profiling wavelengths was established using the Langley plot technique [*Bhartia et al.*, 1995]. For the NOAA11 SBUV/2 instrument, the long term calibration was established using an on-board calibration lamp system. The post-launch calibration after January 1993 was extrapolated on the assumption that the rate of diffuser degradation had not changed [*Hollandsworth et al.*, 1995].

The calibration of the solar instruments is also affected by the exposure to solar radiation. The use of the MgII index as a proxy for long term measurements of the solar UV irradiance has an advantage that it is based on the measurement of the ratio of intensities in the center and wings of the unresolved MgII h and k solar absorption lines. Being a ratio, it is relatively less sensitive to the instrument degradation. However, to estimate relative changes in different wavelength bands of the solar UV irradiance with respect to the MgII index, it is necessary to specify the values of SF. In CM94, SF for the 200-205 nm band was assumed to be unity. This value is based on the estimate provided by *DeLand and Cebula* [1993] from their study of relative changes in the UV irradiance and the MgII index on time scales of a solar rotation.

The simultaneous measurements of the solar UV irradiance, ozone and temperature on the UARS satellite provide an opportunity to address these issues and estimate the solar cycle variation of ozone in the upper stratosphere by well calibrated UARS instruments. The UV irradiance in the spectral range 200-205 nm is measured by the SOLSTICE and SUSIM instruments with an estimated long term precision of about 1% [*Rottman et al.*, 1993; *Brueckner et. al.*, 1993]. The ozone and temperature are measured by the UARS MLS instrument on a near continuous and a near global basis and do not have known degradation sources. Of relevance to this paper, excellent tracking between MLS ozone and other data sets (ozonesondes, Lidar, TOMS), is demonstrated by *Froidevaux et al.* [1994, 1996]. For the upper stratosphere in particular, average comparison between MLS and SAGE II from

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September 1991 through 1994 also give very small differences in trends [see *Cunnold et al.*, 1996]. These data support upper stratosphere trend differences between the two data sets of less than 0.5 % per year. The MLS data now covers about a 4 year period since the launch of the UARS satellite on September 12, 1991. Though this is a relatively short period for studying solar cycle variation of ozone, it encompasses a period from a near solar maximum to a near solar minimum condition. The purpose of this paper is to study the upper stratospheric ozone response to changes in the solar UV irradiance based on the UARS data in the context of the solar cycle related changes in these parameters discussed in CM 94.

Ozone, Temperature and Solar Irradiance Data

The ozone from the MLS instrument is measured in two radiometric bands, near 183-GHz and 205-GHz respectively. The MLS retrieval technique uses a sequential estimation approach to obtain tangent pressure and temperature from the 63-GHz O_2 band, followed by mixing ratio retrievals in other bands. We present here the data from the 205-GHz channel, which provides the most complete coverage of ozone data since the launch of the UARS MLS. Both the ozone and temperature time series were generated from the MLS Version 3/level 3AL data files on the Central Data Handling Facility at the Goddard Space Flight Center, also stored on the Goddard Distributed Active Archive Center (DAAC). These data are zonally averaged into 4° latitude bands. The solar irradiance data from the SOLSTICE and SUSIM instruments, used in this study, are also taken from the Goddard DAAC. They were generated from the SOLSTICE version 8 and SUSIM version 16 data. Some of the important characteristics of these data, related to the MgII index and the 27 day solar rotation and the 11 year solar cycle, are discussed in *Chandra et al.*[1995a].

Figure 1 shows relative changes in the daily values of the integrated solar UV irradiance between 200-205 nm range (F200-205), derived from the SOLSTICE and SUSIM data. The time periods correspond to October 3, 1991 to December 31, 1994 for the SOLSTICE data and October 20, 1991 to July 20, 1994 for the SUSIM data. Each time series is expressed in terms of percent change with respect to the mean (dotted lines) and is smoothed twice with a 35 day running average (solid lines) to suppress short term fluctuations related to 27 day solar rotations. A comparison of the F(200-205) time series from the two instruments suggests similar changes in the solar irradiance in this wavelength range. The data from both instruments show a clear indication of the declining phase of the solar cycle 22 from a near solar maximum to a near solar minimum condition. The downward trends in F(200-205) from both the SOLSTICE and SUSIM data are about 1.8 to 2 % per year and are strongly modulated by the 27 day solar rotational cycles.

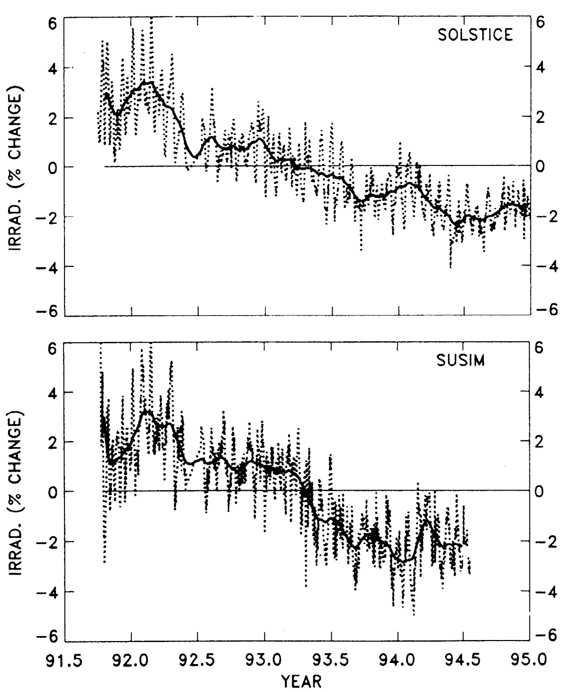

Figure 1. Relative changes in the daily values of the integrated solar UV irradiance between 200-205 nm range, (F200-205), derived from the SOLSTICE (upper panel) and SUSIM data (lower panel). Each time series is expressed as % change with respect to the mean of the time series (dotted lines) and is smoothed twice with a 35 day running average (solid lines).

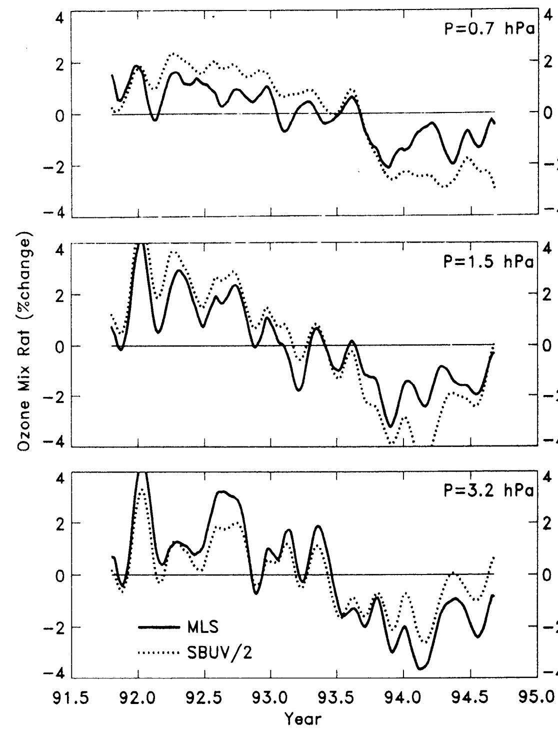

Figure 2. Relative changes in ozone inferred from the MLS measurements at 0.7, 1.5 and 3.2 hPa. Shown in this figure are the corresponding changes in ozone from the SBUV/2 data at Umkher layers 10 (top), 9 (middle) and 8 (bottom). The time series are averages of the time series between ± 35° after removing the seasonal harmonics from each latitude band.

Figure 2 shows relative changes in ozone at 0.7. 1.5, and 3.2 hPa inferred from the MLS data. The time series are averages of the time series between ± 35° latitude after removing the seasonal harmonics consisting of annual, and semi-annual components. Shown in this figure are also the ozone time series derived from the SBUV/2 data in Umkher layers 10, 9 and 8. These layers correspond approximately to the MLS pressure range shown in the upper, middle and the lower panels of Figure 2. The SBUV/2 time series are also averaged between ±35° latitude after removing the seasonal cycles as in the MLS time series. All the time series are smoothed with a 35 day running average (averaged twice) to minimize the effects of diurnal, solar rotation, and short term (4-6 weeks) dynamical perturbations. Figure 2 shows remarkable similarities between ozone time series inferred from the MLS and the SBUV/2 instruments both with respect to fluctuations of 4-6 month periods and the downward trends from 1991 to 1994. The SBUV/2 time series at 0.7 and 1.5 hPa, however, appear to have larger slopes than those in MLS at these altitudes. For example, the linear trends inferred from the MLS data are respectively- .99% and -1.5% per year at these altitudes. The corresponding trends inferred from the SBUV/2 time series are respectively 1.8% and -2.4% per year, all with an error bar of about 0.1 % per year at the 2σ level. Some of these difference may be related to the diffuser plate degradation of the SBUV/2 instrument which may have been underestimated, particularly after the middle of 1993. The agreement between the MLS and the SBUV/2 time series is less apparent below 4 hPa (not shown) because the SBUV/2 measurements are affected by volcanic aerosols at tropical latitudes [*Torres and Bhartia*, 1995].

The short term fluctuations of about 4 to 6 months seen in both the SBUV/2 and MLS data are of dynamical nature and are inversely related to dynamically-induced temperature fluctuations shown in Figure 3 [CM 94]. The ozone and temperature phase relation is a manifestation of temperature dependent ozone photochemistry in the upper stratosphere and is extensively discussed in the literature [e.g., *Chandra*. 1990 and the references therein]. The combined effects of solar cycle variation of ozone and dynamically-induced temperature fluctuations of 4-6 months indicated in figures 1, 2 an 3 can be estimated using a linear regression model as follows:

$$\delta O3 = S_0 + S_T * \delta T + S_{uv} * \delta F \qquad (1)$$

where δO3, δT are daily values of ozone and temperature shown in Figure 3 and δF the corresponding value of F(200-205) inferred from the smoothed time series in Figure 1. The regression coefficients S_T and S_{uv} may be interpreted as ozone sensitivity to temperature and UV irradiance (% change for 1° K change in temperature or for 1 % change in F(200-205).

Figure 4 compares the MLS ozone time series with the regression model using equation (1) at 0.7, 1.5 and 3.2 hPa. The modelled time series are highly correlated with the observed ozone time series in this altitude range and capture most of their important features including the UV related changes associated with the declining phase of the solar activity. Figure 4 clearly shows the significance of including the effects of dynamically-induced temperature oscillations in the regression model. For example, while the F(200-205) time series in Figure 1 continues to decrease or reaches a near minimum level after the middle of 1993, the ozone time series in Figure 2 seems to show a slight increase. This increase is a manifestation of dynamically-induced temperature oscillations as seen in the modelled ozone in Figure 4.

The values of S_{uv} estimated from the regression model are respectively 0.54 , 0.91 and 0.92 with an error bar of ± .04 at the 2σ level. The corresponding values of S_T are respectively -0.27, -1.03, and -1.34 with an error bar of ± 0.1 at the 2σ level. These values of S_T are in the same range as the values derived from the Nimbus-7 SBUV ozone and the NMC temperature time series from 1979 to 1986 [*Chandra*, 1990]. The values of S_{uv} in the 1.5 to 3.2 hPa range are comparable to 0.98 estimated from the 15 years of the combined SBUV and SBUV/2 data at these altitudes as given in CM94.

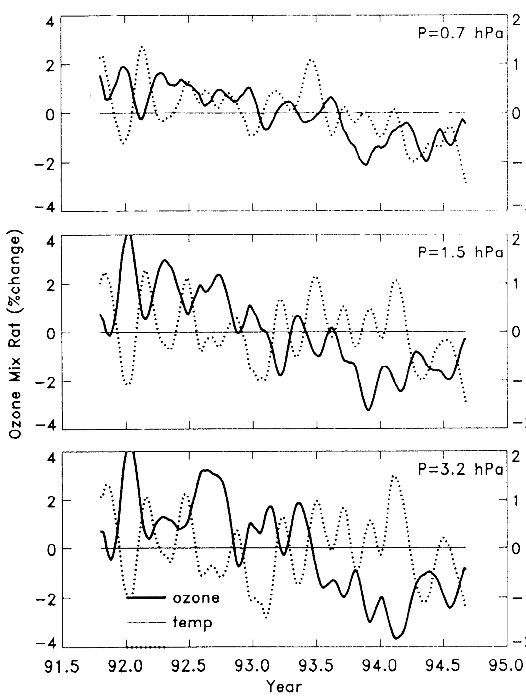

Figure 3. Same as Figure 2 except that MLS ozone time series are compared with temperature time series.

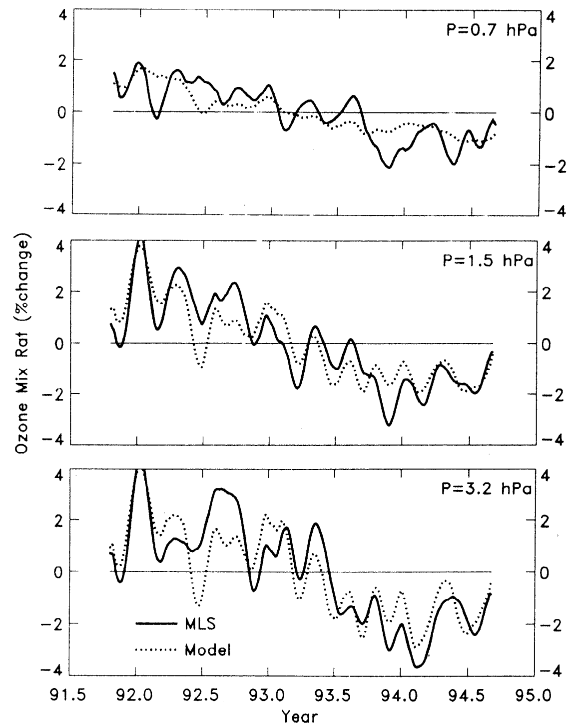

Figure 4. Same as Figure 3 except that the MLS ozone time series are compared with ozone time series derived from a linear regression model (dotted lines) based on the MLS temperature and the SOLSTICE UV measurements as discussed in the text.

In using the regression model we implicitly assumed that the decrease in ozone during the declining phase of solar cycle 22 is entirely due to changes in the solar UV irradiance. In reality, some of the observed decrease may be due to anthropogenic perturbations of ozone as discussed in *Chandra et al.*[1995b] and *Hollandsworth et al.* [1995]. Unfortunately, based on the three years of UARS data, it is not possible to distinguish between the solar cycle and the anthropogenic components. The problem is less ambiguous if one uses a larger data set covering a time period of about a solar cycle. *Chandra et al.* [1995b] and *Hollandsworth et al.* [1995] have shown that the low latitude ozone trends in the upper stratosphere are about 2- 4% per decade but are statistically not significant. Their analyses were based on the 15 years of the combined Nimbus-7 and NOAA-11 data which extended over more than a solar cycle. If one allows for a trend of 2-4% per decade as a real atmospheric change due to anthropogenic perturbations, the value of S_{uv} estimated from equation (1) will decrease by 10-20%. The estimated values of S_{uv}, after accounting for possible anthropogenic effects, are in the range of 0.4 to 0.8 in the upper stratosphere. These values suggest that for a change of 5% in F(200-205) over a solar cycle, ozone may vary from about 2% at 0.7 hPa to about 4% in the 1.5 to 3.2 hPa range.

It may be noted that the values of S_{uv} in the upper stratosphere at low latitudes, derived from the SAGE (Stratospheric Aerosol and Gas Experiment) I and SAGE II measurements, are in the range of 0.2 to 0.3 and are statistically not significant [*Wang et al.*, 1996]. The SAGE values of S_{uv} appear to be significantly less than the values derived from the SBUV or MLS measurements. The SAGE values of S_{uv} are however derived from ozone number density at fixed altitude and are not directly comparable to the SBUV or the MLS values which are derived from the ozone mixing ratio on pressure surfaces. When SAGE ozone values are converted to mixing ratios on pressure surfaces, they show somewhat larger value of S_{uv} than SBUV in the tropics (over the period 1979-1991) but similar values at other latitudes. The tropical differences in the same frame of reference may be associated with the NMC derived tropical temperature changes used in converting the SAGE ozone values to mixing ratio at pressure surfaces [Cunnold, personal communication, 1996]. This possibility was also raised by McPeters et al.[1994] in their comparison of SBUV and SAGE II ozone profiles.

Summary and Conclusions

In this paper, we studied the ozone response to long term changes in the solar UV irradiance in the upper stratosphere based on well calibrated ozone, temperature and solar irradiance measurements from a number of instruments on the UARS satellite. Even though the UARS data base is limited to 3-4 year period after September 1991, it covers an important phase of solar cycle when the solar activity changed from a near solar maximum to a near solar minimum level. A comparison of the MLS and the NOAA11 SBUV/2 ozone time series during this period suggests a continuous decrease in ozone in the upper stratosphere in phase with the solar UV flux in the 200-205 nm band measured from both the SOLSTICE and SUSIM instruments. The rate of decrease is relatively larger in the SBUV/2 than in MLS data above 1.5 hPa. This difference may be attributed to the degradation of the diffuser plate of the SBUV/2 instrument which may not have been fully corrected at shorter wavelengths. Both the MLS and the SBUV/2 data show short term oscillations of 4-5 months which are forced by dynamically-induced temperature oscillations.

The ozone sensitivity to F(200-205), related to the solar cycle variation, is in the range of 0.7 to 0.8 between 1.5 and 3.2 hPa. These values are about 25 to 30 % larger than the values derived from 2-D photochemical models which do not include the possible effect of the solar cycle variation of temperature [*Fleming et al.*, 1995]. The differences between the calculated and the observed sensitivity are even larger when photochemical models allow for the solar cycle variation of temperature [*Huang and Brasseur*, 1993; *Brasseur*, 1993; see also *Fleming et al.* 1995]. The 2-D models tend to underestimate both the absolute values as well as the seasonal and latitudinal variations of ozone in the upper stratosphere [*Chandra et al.*, 1993]. The UARS measurements suggest that these models also underestimate the long term solar response of ozone in the upper stratosphere, in general agreement with previous studies. A clear explanation of the observed solar cycle variation of ozone in the upper stratosphere will require improved understanding of the coupled chemical, radiative, and dynamical processes in that region.

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