

Comparisons between Stratospheric Aerosol and Gas Experiment II and microwave limb sounder ozone measurements and aliasing of SAGE II ozone trends in the lower stratosphere

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Abstract. SAGE II ozone measurements are compared with coincident microwave limb sounder (MLS) measurements over the period September 1991 to December 1993. Between 1.5 and 10 mbar the MLS ozone values are approximately 5% larger than the Stratospheric Aerosol and Gas Experiment (SAGE) II values. These differences are remarkably systematic in space and time. At 1 mbar the mean differences are zero and the mean differences oscillate with level at lower pressures. A month of comparisons against Halogen Occultation Experiment ozone measurements suggests that the differences at pressures less than 1.5 mbar are a feature of the MLS measurements. There are also differences between SAGE II sunrise and sunset measurements at 1 mbar which may be associated with the diurnal tide. At pressures greater than 10 mbar the comparisons indicate that the SAGE II ozone retrievals are being biased by the large aerosol concentrations resulting from the Mount Pinatubo eruption. For a fixed aerosol extinction the SAGE II/MLS difference (ppm) is larger at higher altitudes. It also depends nonlinearly on the aerosol extinction at pressures greater than 20 mbar. These effects are probably caused by the interpolation of the SAGE II aerosol extinction to $0.6 \mu\text{m}$ and by the evolution of the aerosol size distribution. For UARS layer aerosol optical depths less than 2×10^{-3} at $1.02 \mu\text{m}$, the aerosol effect on the SAGE II ozone retrievals is inferred to be $3 \times 10^{10} \text{ cm}^{-3}/10^{-3}$ aerosol layer optical depth at pressures greater than 20 mbar. This is equivalent to approximately 3% of the aerosol extinction at $0.6 \mu\text{m}$ being interpreted as ozone. At low aerosol concentrations and between 10 and 31 mbar, MLS ozone values are found to be approximately 5% larger than SAGE II ozone values (in agreement with the higher-altitude differences). Atmospheric aerosol concentrations prior to the Mount Pinatubo eruption were large enough, particularly in the tropics after Ruiz in 1985, that long-term trends in SAGE II ozone in the lower stratosphere are inferred to be biased downward. As a result, the SAGE II column ozone trends in the tropics over the period 1984–1991 need to be increased by approximately 0.2%/yr. This effect can account for a large fraction of SAGE II total ozone mapping spectrometer (TOMS) column ozone trend differences over this period. Good agreement is found between the TOMS and the SAGE II column ozone trends if the period of comparison is restricted to times of low aerosol concentrations.

1. Introduction

Over the past two decades the solar backscattered ultraviolet (SBUV) instrument on the Nimbus 7 spacecraft and the two Stratospheric Aerosol and Gas Experiment (SAGE) instruments on separate spacecraft have produced excellent long-term records of the behavior of ozone in the middle and upper stratosphere. These data sets have been used to infer long-term changes in upper stratospheric ozone and the measured changes have some features in common [e.g., *McPeters et al.*,

1994] and some similarity to the predicted changes due to the global accumulation of chlorofluorocarbons (CFCs). Nevertheless, the derivation of decadal trends in ozone from these data sets depends on the satellite team's ability to maintain excellent long-term calibration of the instruments and on an understanding of how small retrieval uncertainties can influence the inferred trends. Systematic comparisons of these data sets among themselves and against coincident ozone measurements by other ground-based and satellite sensors can help to illuminate uncertainties associated with each of the measurement systems.

As these satellite systems age (SAGE II) and die (SBUV), it is most desirable that these ozone measurements be intercalibrated against the newer measurement systems in order that

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Paper number 95JD01707.
0148-0227/96/95JD-01707\$05.00

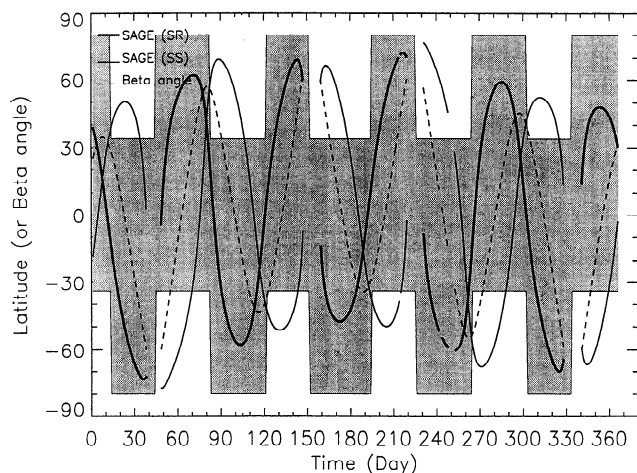


Figure 1. Time series of latitude locations of Stratospheric Aerosol and Gas Experiment (SAGE) II sunrise and sunset and microwave limb sounder (MLS) (the hatched area) observations for 1992. The β angles for the SAGE II instrument are also included.

long-term trends derived from a combination of the ozone sensors be useful. The ozone measurements from the Upper Atmosphere Research Satellite (UARS) provide valuable additions to the global ozone measurement database. SAGE II has continued to operate during the UARS mission, and comparisons against the UARS measurements are useful for both the reasons indicated above. In this paper, comparisons against measurements by the microwave limb sounder (MLS) are emphasized. Short-term comparisons against the Halogen Occultation Experiment (HALOE), the cryogenic limb array etalon spectrometer (CLAES), and the improved stratospheric and mesospheric sounder (ISAMS) measurement system are discussed by *Cunnold et al.*, [this issue].

2. SAGE II Measurements

SAGE II was launched in October 1984 into an orbit inclined at 58° to the ecliptic. It has been taking measurements routinely since that time. Atmospheric trace gas (and aerosol) profiles are measured close to times of spacecraft sunrise and sunset (at ground sunrise and sunset) as the SAGE II telescope views the Sun through the lower part of the atmosphere (i.e., during solar occultation). Two sets of constituent profiles are thus obtained per satellite orbit, a local (ground) sunrise and one sunset. The precession of the SAGE II satellite orbit is such that the locations of ground sunrises and sunsets are nearly identical from year to year. Figure 1 shows the latitudes of sunrise and sunset measurements in 1992. At certain times of the year (e.g., mid-February) the angle between the satellite's orbital plane and the solar viewing direction (the β angle) is large. At that time, the duration of spacecraft sunrise or sunset is unusually long and the trace gas profiles are necessarily obtained over a wide spatial range. These retrievals are regarded as unreliable and profiles with a β angle greater than 60° are not included in the SAGE II data set.

SAGE II measurements are self-calibrating in the sense that the Sun is observed just above the atmosphere and trace gas concentrations are inferred by ratioing the measured solar radiation through the atmosphere to that measured outside the atmosphere. The error bars (precision) on retrieved profiles

are also estimated separately for each profile based on individual profiles being made up of approximately six scans of the Sun for each altitude on the profile. The precision of an individual SAGE II ozone profile between 24 and 48 km, at 1-km resolution, is better than 7% and if the resolution is reduced to 5 km, this improves to 5% or better [*Cunnold et al.*, 1989]. The accuracy of SAGE II ozone measurements is estimated to be 6%.

Although the altitude separation between individual profile points can be estimated very precisely from the scan mirror data and the satellite orbit information, an important uncertainty in the measurements is establishing a reference, base altitude for the profile. For SAGE II the reference altitude is determined entirely from the spacecraft ephemeris information. Based on an analysis of these data, *Buglia* [1989] concludes that SAGE I reference altitude errors can increase to 350 m (1σ) at the end of a 7-day period before an adjustment to the ephemeris is made; he also concluded that the errors are less for SAGE II but can reach 200 m. This uncertainty produces constituent profile errors consisting not only of a direct altitude offset in the profile but also a small and mostly negligible error in removing the Rayleigh scattering contribution from the retrieval of the trace gas profiles [*Chu et al.*, 1989; *Cunnold et al.*, 1989]. The Rayleigh scattering contribution is estimated from meteorological data supplied for each profile by the National Weather Service (NWS) and given on pressure surfaces.

In performing comparisons against other sensors, such as the UARS instruments, it is necessary to relate altitudes to pressures. This conversion is performed for the SAGE II data using the individual profile NWS data which also contains hydrostatically derived altitudes. The SAGE II mixing ratios are first interpolated to pressure levels differing by a factor of $10^{-1/18}$ in pressure; this is approximately equivalent to the 1-km altitude resolution of the SAGE measurements. Three of these sub-UARS layers then make up a UARS layer for which the end point pressures differ by $10^{-1/6}$. The average SAGE II mixing ratio in the UARS layer is then calculated in such a way that the columnar ozone is conserved and the ozone column contribution from individual UARS layers is just the average of the three sublayer contributions. Thus SAGE II ozone profiles are being degraded to the UARS layer resolution using

$$\bar{\chi}_i = \frac{\sum_{i=1}^3 \chi_i \Delta p_i}{\sum_{i=1}^3 \Delta p_i}$$

where Δp_i is the pressure difference between the boundaries of the sub-UARS layers and χ_i is the mixing ratio in the sub-UARS layer.

Another uncertainty in the lower stratosphere results from the need to separate the ozone contribution at $0.6\ \mu\text{m}$ from the aerosol contribution. Near the peak of the stratospheric aerosol layer, the ozone and aerosol contributions are approximately equal under typical aerosol conditions [e.g., *Chu et al.*, 1989]. For this reason the accuracy of the SAGE II ozone measurements may degrade by 4% below 25 km in the tropics and below 20 km at midlatitudes under normal aerosol loading conditions [*Cunnold et al.*, 1989]. There have always been some concerns about how accurately this separation can be achieved [e.g., *Cunnold and Veiga*, 1991], and the possibility that chang-

ing atmospheric conditions (i.e., aerosol loading) could have some influence on SAGE-II-derived ozone trends should not be neglected. During the UARS mission, stratospheric aerosol concentrations were exceptionally high because of the Mount Pinatubo eruption. Therefore comparisons between the SAGE II and the UARS ozone measurements provide data for investigating this possibility.

The only known change in the SAGE II satellite instrument, which has affected the retrieved profiles, was a problem in the scan mirror mechanism which commenced in mid-1989. This affected the ozone retrievals at pressures less than 1 mbar (i.e., at altitudes where the ozone signature is small). The retrieval algorithm contains a correction for this problem and the effect on the retrieved profiles is believed to be just a small increase in SAGE II ozone noise levels above approximately 50 km after mid-1989.

Some SAGE II profiles have been excluded from the comparisons reported here based on experience gained in comparisons against SBUV [Wang, 1994; H. Wang, manuscript in preparation, 1995]. The excluded data consists of the SAGE II which has been flagged as other than type zero (for example, retrievals for which no NWS temperature data were available are always flagged and often possess large errors). Furthermore, a retrieved profile sensitivity to the SAGE II β angle has been identified following times when the β angle exceeded 60° [Wang, 1994]; at such times, only profiles for which β was less than 40° have been used; this eliminates just a few days of profiles 4 times per year. There are also some occasional indications of possible pressure-altitude errors in the data, perhaps related to the ephemeris errors described by Buglia [1989]; however, these are still under investigation. The latter errors should, however, average out to zero and no data have been excluded for this reason.

The diurnal variation of ozone is small ($\leq 5\%$) for altitudes below the 1-mbar level. Between 0.3 and 1 mbar the conversion of atomic oxygen to ozone occurs just after sunrise and just before sunset, so that SAGE II ozone values should be typical of nighttime conditions [e.g., Chu, 1989; Chu and Cunnold, 1992]. In order that the comparisons are useful over the entire pressure range of the MLS measurements, only nighttime MLS measurements are employed in the comparisons.

3. MLS Measurements

The MLS ozone measurements compared here are the version 3 205-GHz data, which intrinsically provide more accurate results than the 183-GHz data [Froidevaux *et al.*, 1994, this issue]. The MLS retrieval solves for ozone on alternate UARS levels starting at 0.46 mbar (i.e. the vertical resolution of the reported MLS ozone profiles is 2 UARS levels)—actually the MLS measurements go somewhat higher than this, but at the higher altitudes they are noisier and possess significant a priori information. Level 3 MLS profiles provide ozone mixing ratios on every UARS level but the values in between MLS retrieval levels are simply an average of the values above and below (thus leading to reduced variability at the intermediate levels); these intermediate level values were not used, even in the forward model calculation, in the MLS version 3 retrievals [Froidevaux *et al.*, this issue]. The retrieval algorithm is a sequential estimation procedure (based on Rodgers [1976]) through which the MLS measurements are combined with an a priori, climatologically based profile. The procedure provides error bars at each point on the retrieved profiles and altitudes

where the retrieved profile contains more than a 25% contribution from the a priori are flagged with negative error bars. MLS ozone values with negative error bars have not been used in these comparisons. The expected accuracy of the MLS ozone measurements is about 0.3 parts per million by volume (ppmv) which translates into an accuracy of 15–20% at 0.46 and 46 mbar and of 3% at 4.6 and 10 mbar [Froidevaux *et al.*, this issue]. The precision of the measurements is approximately 0.2 ppmv from 2.2 to 46 mbar with larger values outside this range; this translates into a precision of 2–4% from 2.2 to 22 mbar. However, the error bars supplied with the MLS ozone profiles are typically 5–10% because the latter include contributions from the a priori profiles and some nonrandom components.

To provide almost global coverage, the UARS spacecraft undergoes a 180° rotation (the yaw maneuver) approximately every 36 days. As a result, the MLS measurements alternate between covering the latitudes of 32°N to 80°S and 32°S to 80°N . The latitudinal coverage by MLS is illustrated in Figure 1, where it is overlaid with the SAGE II measurement locations.

4. MLS/SAGE II Comparisons

SAGE II profiles have been compared against coincident MLS profiles. Coincidence here is defined as the closest nighttime MLS measurement in time and space to the SAGE II measurement; on the average, the temporal separation is 6 hours and the spatial separation is 1° in latitude and 6° in longitude. All the available SAGE II and MLS ozone profiles satisfying the coincidence criteria and the SAGE II selection criteria mentioned in section 2 over the period September 1991 to December 1993 have been included.

Mean Values Sorted by Latitude and Pressure Level

Figure 2 depicts these comparisons at the MLS levels. The differences change sufficiently slowly with latitude that almost all the features are captured by subdividing the comparisons into just tropical and midlatitudes (as shown in Figure 3), which however also shows the intermediate levels. The comparisons indicate that between 2.2 and 22 mbar (the altitude range where both measurement systems have their best precision) the MLS ozone values are systematically approximately 5% larger than the SAGE II values and the standard deviations of the daily zonal mean differences are approximately 3%. Note, however, that SAGE II sunset and sunrise ozone measurements possess different means at pressures less than 4.6 mbar but again agree at 0.31 mbar; the largest sunrise/sunset differences are at 1 mbar. We are investigating the possibility that these differences are caused by the diurnal tide; this would affect the conversion from SAGE II altitudes to pressure levels which are currently based on NWS temperatures supplied once per day (i.e., without any diurnal variation). At most latitudes the diurnal tide would be expected to result in maximum temperatures at sunset and minimum temperatures at sunrise [Forbes, 1982].

Between 1 and 0.31 mbar the SAGE II/MLS differences are less systematic than at lower altitudes. It is important to note, however, that in Figure 3 ozone values at all the UARS levels are plotted, but only even-numbered layers are actually used in the MLS retrievals (to emphasize this point, the differences at the odd levels on Figures 3, 4, and 11 are not given any error bars). A comparison of HALOE and MLS ozone measure-

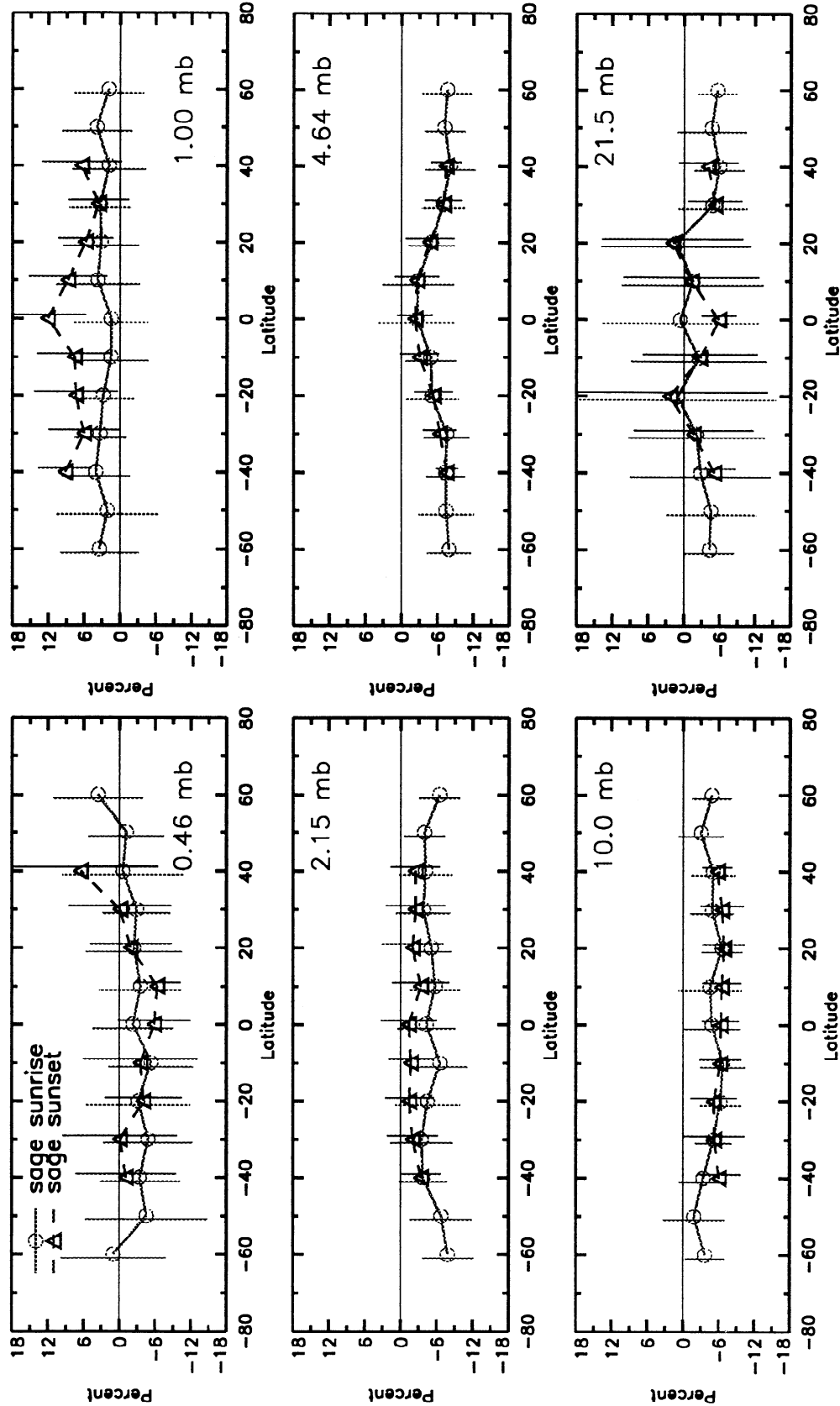


Figure 2. Mean ozone differences in 10° latitude bands at the MLS retrieval levels for coincident nighttime MLS and SAGE II sunrise (solid line) and sunset (dashed line) profiles over the period September 1991 to December 1993. Differences are given as (SAGE II-MLS)/MLS * 100. The error bars are ±1 standard deviation in the zonal mean daily differences.

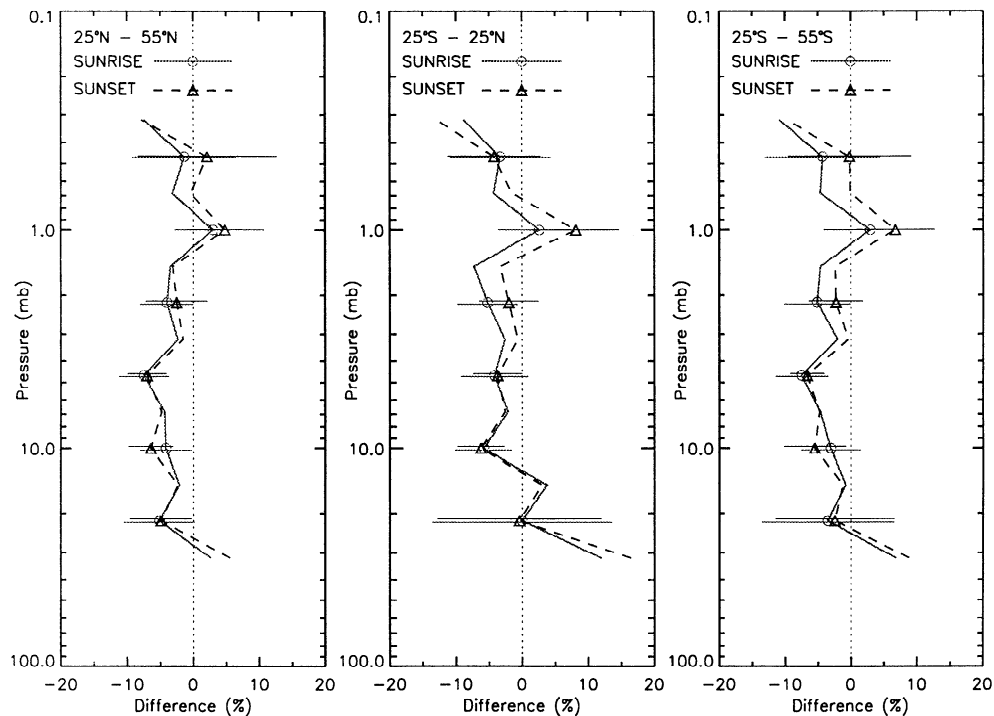


Figure 3. Mean SAGE II/MLS ozone differences and standard deviations at the UARS standard levels as in Figure 2 but binned into just the latitude bands 55° – 25° N, 25° N– 25° S, and 25° – 55° S. The lack of significance of the MLS values on odd-numbered UARS levels is indicated by the omission of error bars on the differences at these levels.

ments for the month of September 1992 shows a similar vertical structure in the differences in this height range. In this comparison the HALOE (version 16) level 2 measurements have been processed in a similar way to the SAGE II measurements. Figure 4 shows that the HALOE and MLS measurements are in good agreement, except for a systematic difference of approximately 10%; SAGE II measurements lie approximately midway between the higher MLS and the lower HALOE measurements over this period. These comparisons show that the MLS ozone values at 1 mbar are approximately 5% smaller, relative to the other levels, when compared to SAGE II and HALOE measurements. The poorer closure in the MLS retrievals at this level than at other levels [Froidevaux *et al.*, this issue] might also be contributing to this feature of the MLS profiles. Although this is a much more limited comparison than that for SAGE II, the HALOE observations in the tropics do not exhibit the sunrise/sunset differences at 1 mbar that are seen in the SAGE II observations. However, HALOE measures its own temperature profiles and the conversion of HALOE data to pressure levels are not therefore based on once per day temperature measurements.

Lower Stratosphere

Figure 3 shows that at pressures greater than 10 mbar, the character of the differences between the SAGE II and the MLS measurements changes and the SAGE II values can exceed the MLS values. Note that the transition from negative to positive differences occurs at slightly lower pressures in the tropics than at midlatitudes. A comparison of the ozone monthly means as a function of time in the tropics and monthly mean time series of aerosol optical depths in each UARS layer (Figure 5) shows that the SAGE II/MLS differences are small when the aerosol

optical depths at $1.02 \mu\text{m}$ are less than approximately 0.005 and a somewhat smaller value at 14.6 mbar than at the other levels. The version 3 MLS ozone measurements in the tropics at 46.4 mbar are lower than ozonesonde data by 0.5 ppmv [Froidevaux *et al.*, this issue], but this is small compared to the SAGE/MLS differences at this level. Figure 6 depicts time series comparisons at the MLS levels at 45° N, showing excellent consistency between the two sets of measurements at these levels with standard deviations of the differences of approximately 3% at 10 mbar and above. Figures 5 and 6 provide a strong indication that the SAGE II ozone retrievals are being affected by the presence of large aerosol concentrations at levels below 10 mbar because, in contrast to visible and infrared measurements, microwave measurements are not effected by aerosols. Furthermore, Figure 5 also suggests that the average of the MLS values above and below, as is provided in the MLS level 3 profiles to obtain the odd-numbered UARS level mixing ratios (here at 14.6 and 31.6 mbar), contributes to identifying the SAGE II aerosol effect.

Figure 7 shows scatterplots of SAGE II/MLS ozone differences versus layer aerosol optical depths at $0.525 \mu\text{m}$. The aerosol effect on SAGE II ozone retrievals is clearly demonstrated by these diagrams. The effect can be quantified by fitting a straight line to the ozone difference as a function of the aerosol loading. The equations for the fitted lines are given in the individual diagrams (the lines appear as curves because logarithmic spacing has been used on the abscissas). The parameter X is the layer aerosol optical depth multiplied by 10^3 . The layer aerosol optical depth used throughout this manuscript is the extinction/kilometer integrated over the vertical extent of a UARS layer (each of which is approximately 2.5 km thick). The vertical extent of each UARS layer is determined

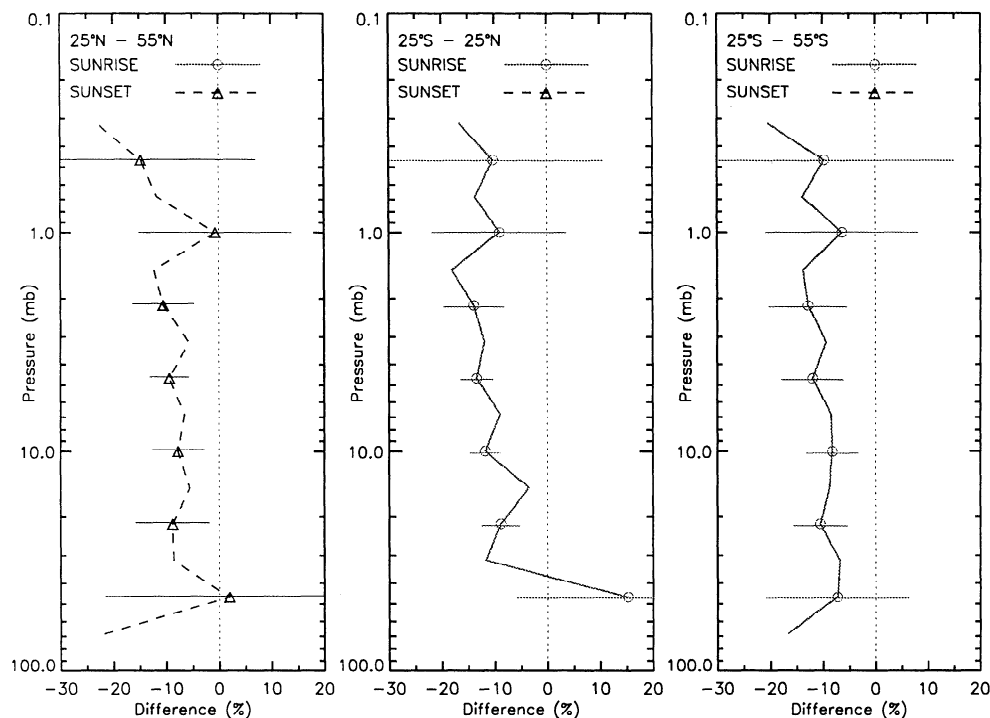


Figure 4. Mean Halogen Occultation Experiment (HALOE)/MLS ozone differences binned into the three latitude bands as in Figure 3 for coincident profiles over the month of September 1992. Differences are given as $(\text{HALOE-MLS})/\text{MLS} * 100$. The error bars are standard deviations of the zonal mean daily differences.

from the NWS temperature profiles. Figure 7 shows that the SAGE II/MLS ozone differences at low aerosol concentrations are negative (as they are at altitudes above the 10-mbar level) and that the sensitivity to aerosols, expressed in $\text{ppm}/10^{-3}$ aerosol layer optical depth, increases with height (the error bars on the sensitivities are less than 0.01).

Scatterplots based on the aerosol optical depths at $1.02 \mu\text{m}$ are shown in Figure 8; the plots are similar to those at $0.525 \mu\text{m}$. The sensitivities are seen to increase with altitude at this wavelength also. The parameters to which the SAGE II measurements are sensitive are the slant column optical depths of ozone and aerosols. Therefore it should be expected that an error in removing the aerosol effects at $0.6 \mu\text{m}$ would directly translate into an error in the ozone concentration, not in the mixing ratio. This means that to compare the sensitivities at the different altitudes, the sensitivities should be multiplied by the pressure in order to express them in units of $\text{cm}^{-3}/10^{-3}$ aerosol layer optical depth. In the remainder of this paper all sensitivities have been converted to these units using a standard atmospheric temperature profile. The pressures differ by a factor of 1.47 between levels and by $(1.47)^3 = 3.16$ between 14.6 and 46.4 mbar. The sensitivity is then found to be approximately constant with altitude at a value of $7 \times 10^{10} \text{cm}^{-3}/10^{-3}$ aerosol layer optical depth. The slopes of the scatterplots would have to increase slightly more with altitude than is shown in Figure 7 for the sensitivity in these units to also be constant at $0.525 \mu\text{m}$.

An attempt has been made to simulate this effect. Dustsonde data supplied by the University of Wyoming following the Mount Pinatubo eruption [Deshler *et al.*, 1992] were used to calculate aerosol extinction values at the SAGE II wavelengths. Synthetic transmission profiles were then calculated in each channel, simulated noise was added, and constituent profiles were estimated using the operational retrieval algorithm.

The process was repeated 30 times to provide estimates of bias and dispersion. The aerosol extinction in this experiment was a factor of 3 to 4 larger at $0.525 \mu\text{m}$ than at $1.02 \mu\text{m}$, and the optical depth of the aerosol layer at $1.02 \mu\text{m}$ was roughly 4×10^{-2} . These conditions were typical of July 1991 over Wyoming [Russell *et al.*, 1993]. The ozone retrievals were found to be biased high by approximately 5% between 25 and 28 km and ozone could not be retrieved below approximately 24 km. Quantitatively, this sensitivity to aerosols agrees within a factor of 2 with the sensitivities derived from SAGE II/MLS differences. There was also an error of approximately 10% in the aerosol extinction at $0.448 \mu\text{m}$ at all heights. This experiment therefore established that SAGE II ozone retrievals can be biased upward under very high aerosol loading conditions.

Inaccuracies in the extrapolation of the SAGE-II-measured aerosol extinctions at 0.385, 0.448, 0.525, and $1.02 \mu\text{m}$ to $0.6 \mu\text{m}$ would be expected to depend on the smoothness of the relationship between the extinctions at the measured wavelengths or equivalently, since the SAGE II algorithm [Chu *et al.*, 1989] solves for the number of particles of the corresponding sizes, the smoothness (and lack of curvature) of the size distribution. Immediately following the eruption, a large number of small particles, especially of sizes less than $0.1 \mu\text{m}$ were observed in the stratosphere [Thomason, 1992; Russell *et al.*, 1993]. Over the next year [Russell *et al.*, 1993] the centroid of the aerosol layer size distribution increased to approximately $0.35 \mu\text{m}$. As a result, the wavelength dependence of the extinction tended to become fairly flat, although there remained some curvature in that dependence. The curvature in the size distribution was larger at that time [Russell *et al.*, 1993].

The behavior of the aerosol extinction is illustrated in Figure 9, where the ratio of the $0.525 \mu\text{m}$ to the $1.02 \mu\text{m}$ aerosol extinction is plotted as a function of the aerosol layer optical

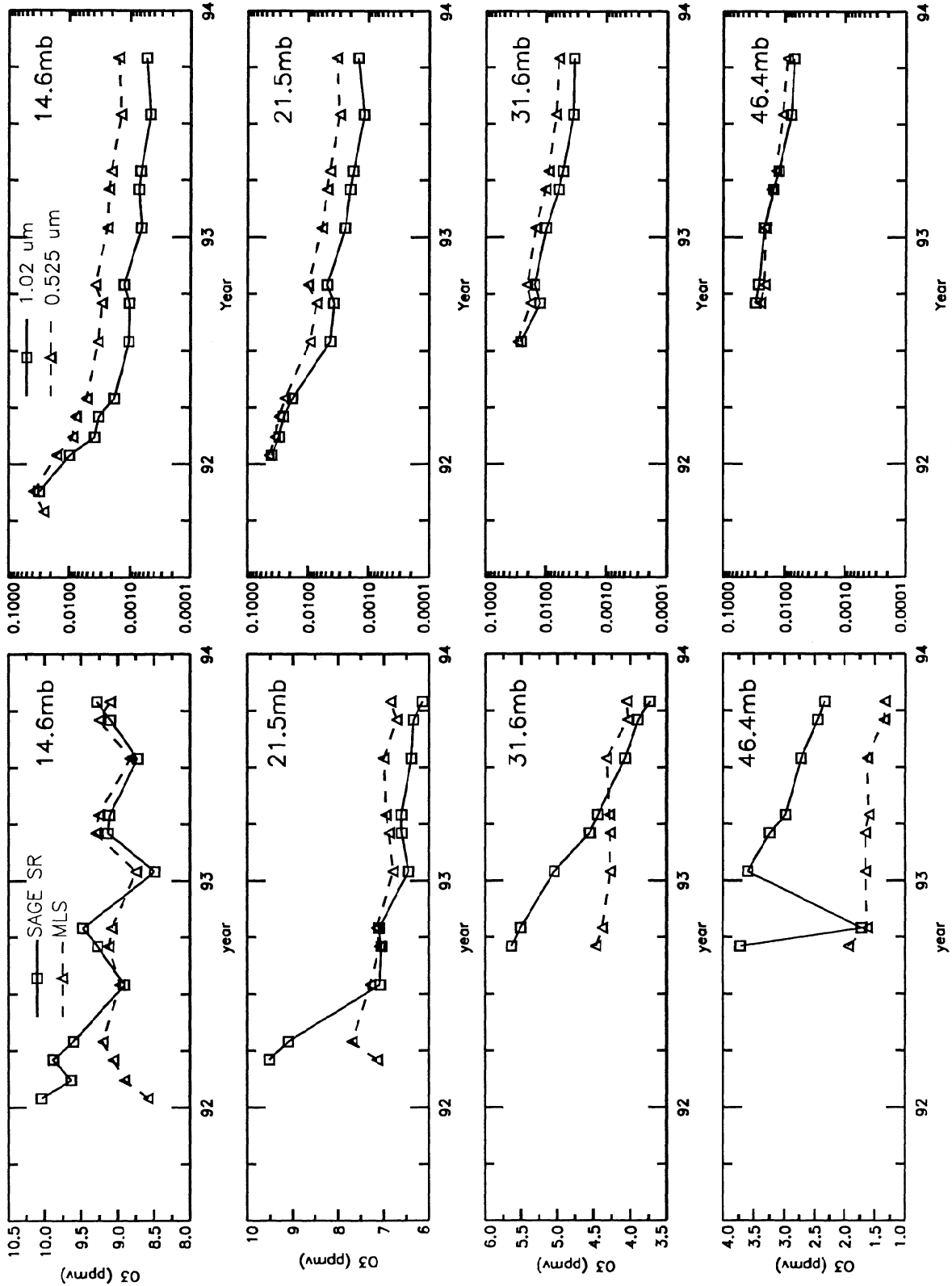


Figure 5. Comparison of the monthly mean time series of coincident SAGE II (sunrise, solid line) and MLS (dashed) ozone measurements in the latitude band 5°N to 5°S in the lower stratosphere (the curves on the left). The curves on the right show simultaneous SAGE II measurements of layer aerosol optical depth at 1.02 μm (solid line) and 0.525 μm (dashed line) at the same levels.

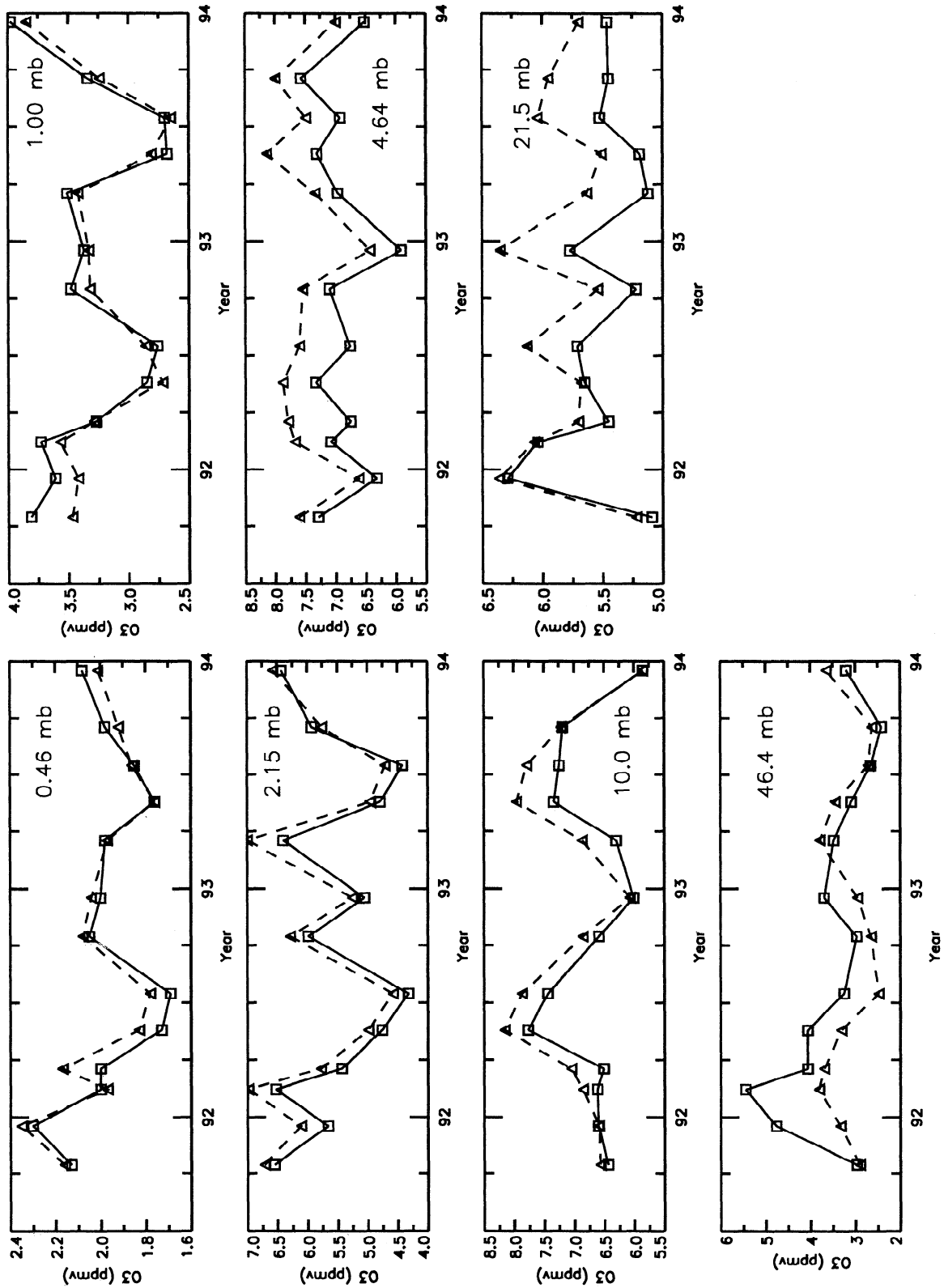


Figure 6. Comparison of the monthly mean time series of coincident SAGE II (sunrise, solid line) and MLS (dashed) ozone measurements in the latitude band 45° to 55°N and at the MLS retrieval pressure levels.

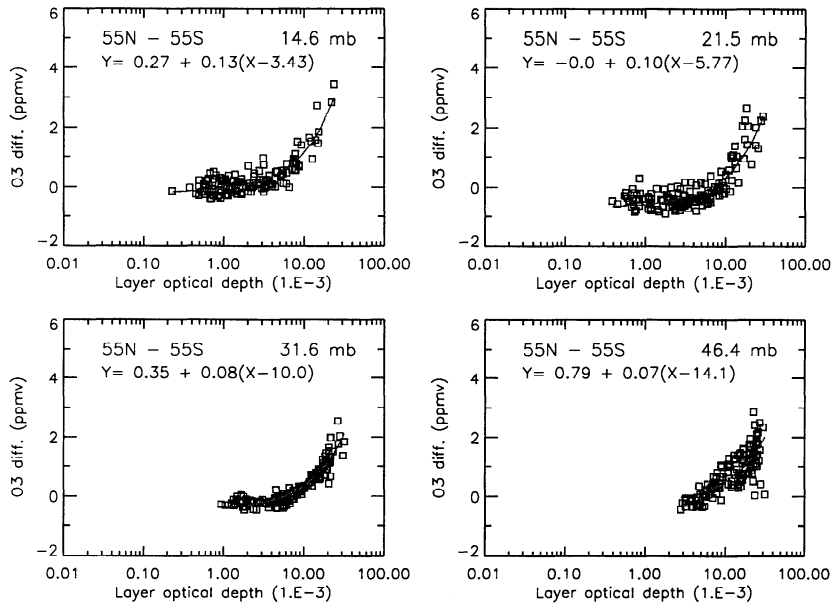


Figure 7. Scatterplots of lower-stratospheric SAGE II/MLS ozone differences in coincident measurements versus layer aerosol optical depth at $0.525 \mu\text{m}$ based on SAGE II coincident measurements. The plots include all the coincident profiles between 55°N and 55°S . The equations (and the lines through the points) show the linear fits to the points with X equal to the layer aerosol optical depth multiplied by 10^3 . These lines are shown as curves in the plots because the values on the abscissas are on a logarithmic scale.

depth at several levels. The ratio of the extinctions is close to unity at the largest aerosol layer optical depths, which is consistent with the fairly flat extinction dependence on wavelength reported by *Russell et al.* [1993] for the first year after the eruption (but excluding early July 1991). The behavior of this ratio as the aerosol layer decays is similar at all four levels except that the extinction at both of these wavelengths decays faster at the higher altitudes, and the extinctions at 14.6 mbar never attained the values seen at the lower altitudes. The extinctions are consistent with a more rapid decay of particles of size larger than approximately $1 \mu\text{m}$ at all altitudes. Figure

9 also indicates that it is not possible to separately infer the dependence of aerosol interference on the aerosol optical depth and the ratio of the extinctions at the two wavelengths; these parameters are obviously highly correlated.

Since the reported sensitivities are based on straight line fits to data in Figures 7 and 8 and the y intercepts correspond to the ozone differences in the absence of aerosols, the sensitivities will be dominated by the effects at the largest aerosol concentrations. Based on the strong relationship between the aerosol extinctions at 0.525 and $1.02 \mu\text{m}$, it is not possible that the sensitivities at both the wavelengths are independent of the

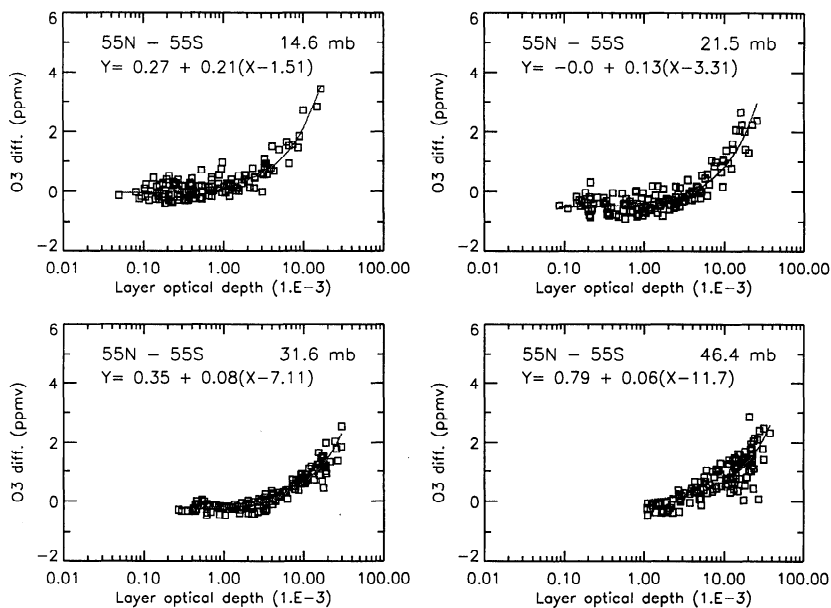


Figure 8. As in Figure 9 but using the SAGE II aerosol optical depths at $1.02 \mu\text{m}$.

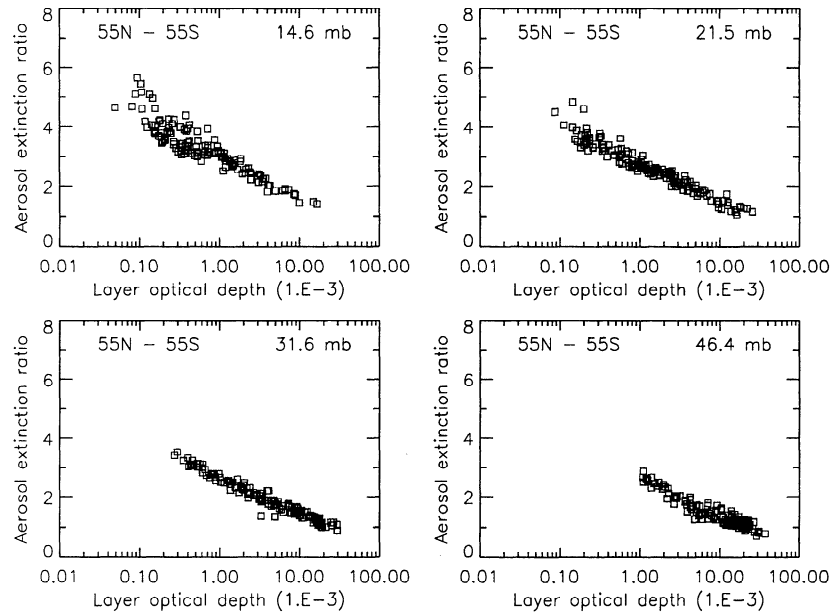


Figure 9. Ratio of the SAGE II aerosol extinction at $0.525 \mu\text{m}$ to that at $1.02 \mu\text{m}$ plotted as a function of the aerosol layer optical depth and depicted for four UARS layers in the lower stratosphere.

aerosol extinction (i.e., at least one of the two sets of scatterplots would be better fit by a curve than by a straight line). The altitudinal consistency of the Figure 8 results at $1.02 \mu\text{m}$ suggests that a straight line fit is acceptable in that case. The inconsistency in the sensitivities at $0.525 \mu\text{m}$ as a function of altitude may then be understood because of the curvature of the sensitivity and the smaller layer optical depths at 14.6 mbar than at the other levels. It would of course have been possible to fit curves instead of straight lines in Figure 7 and 8, but these curves would also have been a strong function of the ozone differences at the largest aerosol extinctions. The resulting sensitivities might still not have much relevance to more typical atmospheric aerosol loading conditions.

The sensitivities that have just been derived correspond to an underestimation in the aerosol extinction at $0.6 \mu\text{m}$ (from the SAGE II interpolation procedure) of approximately 8% at all of the depicted pressure levels. A test of the reasonableness of this result might be to remove the $0.525 \mu\text{m}$ results from the SAGE II interpolation procedure used in the retrievals (during the same time period) and to determine how well the interpolation procedure based on just the 0.385 -, the 0.448 -, and the $1.02 \mu\text{m}$ results simulated the measured values at $0.525 \mu\text{m}$. For the moment, however, a similar type of test can be based on the wavelength dependence of the extinction reported by Russell *et al.* [1993] during three several day periods in the first year after the Pinatubo eruption. The curvature of the extinction dependence on wavelength would seem to permit an interpolation error in the extinction of up to approximately 10%.

5. Potential Effects on SAGE II Ozone Trends

The sensitivities given in Figures 7 and 8 suggest that long-term trends in lower stratospheric ozone deduced from SAGE II measurements could be aliased by aerosol interference. The possibility that changes in atmospheric aerosol loading over time could alias SAGE II trends in the lower stratosphere has been a cause for concern for some time [e.g., Cunnold and

Veiga, 1991]. Because of the possible variation in the sensitivities with aerosol optical depth, it is first necessary to review the aerosol optical depths prior to the Mount Pinatubo eruption in order to determine what was the level of aerosol loading during that period. Figure 10 shows the layer optical depths in the tropics at several levels. The Ruiz eruption was responsible for increasing the aerosol concentrations in this region at the end of 1985. Prior to that time, there had been a general decay of the El Chichon-produced aerosol. Clearly over the SAGE II observational period from 1984 to 1991, there was a downward trend in the atmospheric aerosol loading. Moreover, typical layer optical depths for which the ozone retrieval errors should be estimated are 5×10^{-3} at $0.525 \mu\text{m}$ and 2×10^{-3} at $1.02 \mu\text{m}$. From Figures 9 and 10 it may be seen that the $0.525 \mu\text{m}$ to $1.02 \mu\text{m}$ extinction ratio is approximately 2.5 during both time periods, suggesting that the aerosol size distributions over the appropriate size range may have been similar in these two periods.

The sensitivities have therefore been reevaluated by fitting straight lines to the scatterplots in Figures 7 and 8 but only including measurements having aerosol layer optical depths less than 5×10^{-3} at $0.525 \mu\text{m}$ and 2×10^{-3} at $1.02 \mu\text{m}$. The estimated sensitivities under these conditions are shown in Table 1; sensitivity estimates could not be made at the 46.4-mbar level or below because aerosol concentrations had not decayed sufficiently by the end of 1993. The sensitivity at 14.6 mbar at $1.02 \mu\text{m}$ (and at 10 mbar) is the same as that previously calculated for the higher aerosol concentrations. Furthermore, the sensitivities at $0.525 \mu\text{m}$ at 14.6 and 10 mbar are consistent with these values based on the ratio of the extinction at $0.525 \mu\text{m}$ to that at $1.02 \mu\text{m}$ being approximately 2.5 at the limiting aerosol layer optical depths.

The sensitivities at 21.5 and 31.6 mbar, however, are not consistent with the other calculations and they are approximately a factor of 3 smaller than were calculated for the larger aerosol layer optical depths. Now, in fact, consistency should

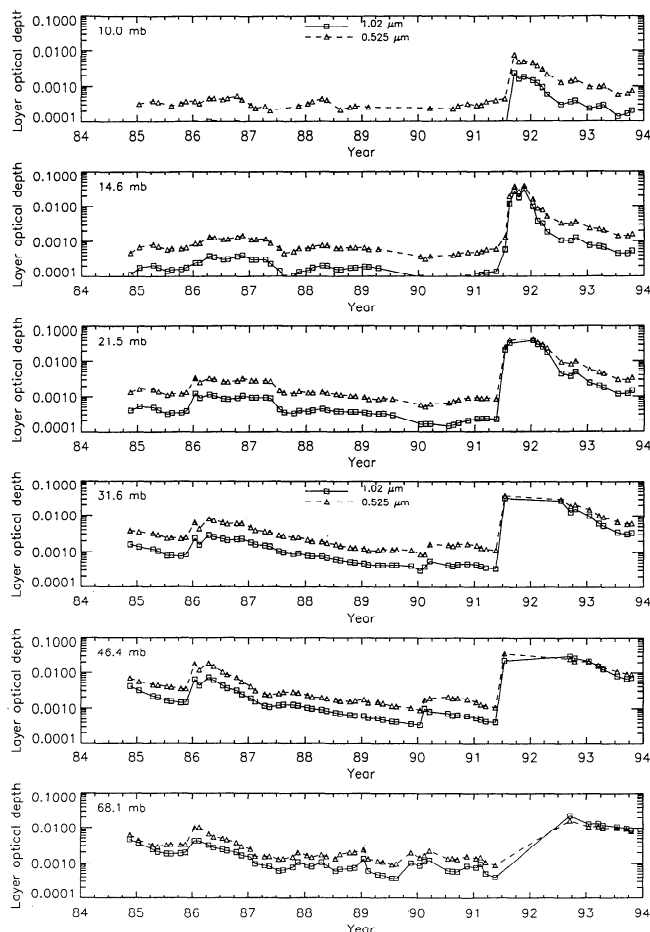


Figure 10. Monthly mean SAGE II layer aerosol optical depths (sunrise values) in the lower stratosphere in UARS layers between 10 and 68.1 mbar in the latitude band 5°N to 5°S. The solid line is for the 1.02- μm measurements and the dashed line is for the 0.525- μm measurements. After the Mount Pinatubo eruption in 1991 the aerosol concentrations were so large that it was not possible to retrieve any profiles in these layers until the aerosol layer optical depths decreased below approximately 0.03.

not really be expected because the error in the aerosol optical depth at 0.6 μm is likely to be a function of the evolving size distribution. The results imply that the size distribution is evolving differently above and below 20 mbar. It is important to

note, however, that the sensitivities at 21.5 and 31.6 mbar are the same (based on the expected difference factor of 1.47), although admittedly the error bars on the sensitivity estimates are very large. Nevertheless, it is significant that at these lower aerosol concentrations, SAGE II/MLS differences are consistently indicating that the SAGE II ozone measurements are biased upward. The differences between the sensitivities at high and more typical aerosol concentrations indicate that at these two levels the sensitivity is significantly dependent on the aerosol optical depths and that the corresponding scatterplots in Figures 7 and 8 must possess significant curvature. This curvature must also account for why the ratio of the sensitivities at 1.02 and 0.525 μm is not approximately 2.5. The sensitivities indicate that at these aerosol layer optical depths approximately 3% of the aerosol extinction at 1.02 μm is being aliased into the SAGE II ozone retrievals.

Before leaving Table 1, it should be noted that it includes the y intercepts of the line fits to the SAGE II/MLS ozone differences dependencies on the aerosol layer optical depths. These indicate the expected ozone differences in the absence of any aerosol contamination. These values are almost the same in Figures 7 and 8 and Table 1. Figure 11 is a revised version of Figure 3 and shows the mean percentage ozone differences in the “absence” of aerosol effects. This figure was actually calculated by limiting the ozone comparisons to those times and locations when the aerosol layer optical depth was less than 2×10^{-3} for pressures greater than 20 mbar and was less than 5×10^{-4} for lower pressures. It may be noted that the SAGE II/MLS ozone differences at low aerosol concentrations are now approximately 5% below the 10-mbar level and are consistent with the differences above this level. Note that this is true, in particular at 46.4 mbar, in midlatitudes but that aerosol concentrations at this level have not yet decreased sufficiently to allow the difference to be estimated in the tropics. The line fit in Figure 7 at this level is also dominated by the midlatitude differences and it therefore does not show a positive intercept which would be expected based on the tropical MLS/ozonesonde comparisons. Pre-Pinatubo SAGE II ozone values at this level in the tropics are approximately 0.5 ppm larger than the measured MLS values [Cunnold *et al.*, this issue].

To estimate the potential effects of this aliasing on long-term trends derived from the version 5.9 SAGE II ozone retrievals, a simple procedure is adopted in this paper. The linear trends in the aerosol optical depths at 1.02 μm over the period October 1984 to June 1991 have also been estimated and are given for each layer in Table 2. We shall base our estimates of

Table 1. Estimated Sensitivities of SAGE II Ozone Retrievals to the Presence of Stratospheric Aerosols Based on Comparisons Against MLS Ozone Measurements for Layer Aerosol Extinctions (in Layers ~ 2.5 km Thick) Using Only Layer Extinctions $< 5 \times 10^{-3}$ at 0.525 μm and 2×10^{-3} at 1.02 μm

UARS Pressure Level, mbar	0.525 μm Result		1.02 μm Result		Optimal Estimates	
	Sensitivity, $10^{10}/\text{cm}^{-3}/10^{-3}$ Layer Extinction	Mean SAGE II/MLS Ozone, ppm	Sensitivity, $10^{10}/\text{cm}^{-3}/10^{-3}$ Layer Extinction	Mean SAGE II/MLS Ozone, ppm	Sensitivity, $10^{10}/\text{cm}^{-3}/10^{-3}$ Layer Extinction	Mean SAGE II/MLS Ozone, ppm
10.0	3.4 ± 1.1	-0.62 ± 0.04	8.1 ± 3.2	-0.56 ± 0.05	8.3 ± 2.2	-0.59 ± 0.03
14.6	3.6 ± 0.8	-0.11 ± 0.02	9.3 ± 2.0	-0.07 ± 0.02	9.2 ± 1.4	-0.09 ± 0.01
21.5	1.5 ± 1.3	-0.48 ± 0.03	2.1 ± 3.2	-0.46 ± 0.03	2.8 ± 2.3	-0.47 ± 0.02
31.6	1.3 ± 1.3	-0.24 ± 0.02	2.1 ± 3.0	-0.22 ± 0.02	2.7 ± 2.2	-0.23 ± 0.01

Also shown are estimated mean Stratospheric Aerosol and Gas Experiment (SAGE) II/MLS differences after removal of aerosol effects and uncertainty limits. MLS, microwave limb sounder.

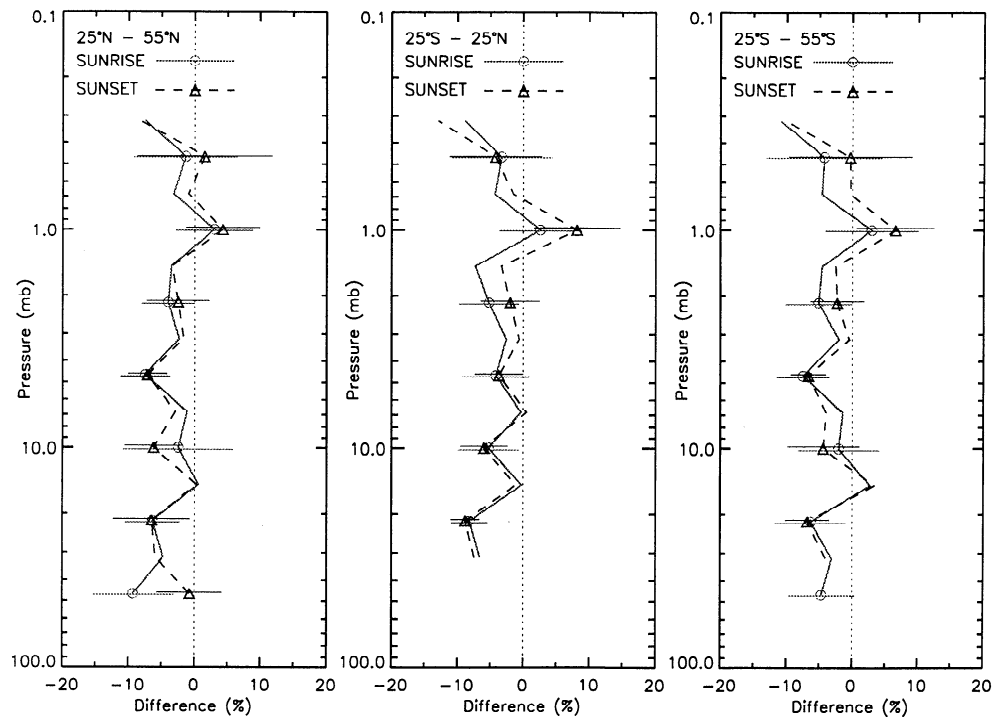


Figure 11. Mean SAGE II/MLS ozone differences as in Figure 3 expressed as $(\text{SAGE II}-\text{MLS})/\text{MLS} \times 100$ but only including those measurements when the $1.02\text{-}\mu\text{m}$ aerosol loading was less than 2×10^{-3} for pressures greater than 20 mbar and less than 5×10^{-4} for pressures less than 20 mbar.

the aliasing on the $1.02\text{-}\mu\text{m}$ aerosol layer optical depth time series. To include the inferred $0.525\text{-}\mu\text{m}$ sensitivities in this estimate, we use the sensitivities given in column 6 of Table 1 instead of those given in column 4. The column 6 sensitivities were obtained by averaging the sensitivity at $1.02\text{ }\mu\text{m}$ and 2.5 times the sensitivity at $0.525\text{ }\mu\text{m}$. Here the factor of 2.5 is based on the ratio of the $0.525\text{-}\mu\text{m}$ extinction to the $1.02\text{-}\mu\text{m}$ extinction where and when aerosols might be expected to influence the ozone retrievals, i.e., in the tropics, between 21.5 and 46.4 mbar, over the period 1985 to 1988. The effect on the ozone trend is then estimated by multiplying the aerosol layer trends by the derived sensitivity factors.

The procedure just described for estimating effects on ozone trends obviously contains a number of approximations. However, because of the large error bars on the sensitivities, we judge it to be sufficiently accurate for this purpose. Neverthe-

Table 2. Trends in UARS Layer Aerosol Optical Depths at $1.02\text{ }\mu\text{m}$ Over the Period October 1984 to June 1991 Estimated From SAGE II Measurements and Inferred Effects on SAGE II Ozone Trends Over This Period Based on Sensitivities Given in Table 1

UARS Layer Pressure, mbar	Aerosol Layer Optical Depth Trends, $10^{-4}/\text{year}^{-1}$			Induced Errors in SAGE II Ozone Trends, %/yr		
	50°N	0°	50°S	50°N	0°	50°S
14.6		-0.2			-0.1	
21.5		-1.0			-0.1	
31.6	-0.4	-2.5	-0.2		-0.2	
46.4	-1.0	-5.5	-0.6	-0.1	-0.8	-0.1
68.1	-2.4	-4.1	-1.6	-0.2	-0.9	-0.1

less, the approximations are being checked by estimating ozone trend differences before and after correcting the individual ozone monthly means for the aerosol effect. Separate estimates are being made based on the sensitivities at 0.525 and $1.02\text{ }\mu\text{m}$. These results are being included in the work of Wang [1994]. Preliminary indications are that the results are very similar to those given in Table 2.

Table 2 indicates that the largest aliasing of the ozone trends occurs in the tropics at 46.4 and 68.1 mbar. However, it was not possible to directly derive sensitivities at these levels because there were still large aerosol concentrations at these levels at the end of 1993. Moreover, there are strong indications that MLS ozone concentrations at 46.4 mbar are anomalously low in the tropics [Froidevaux *et al.*, this issue]. The sensitivities used at these levels in this study are the same as those used at 21.5 and 31.6 mbar. Table 2 then shows that the SAGE II ozone trends in the tropics at 46.4 and 68.1 mbar are being aliased downward by almost 1%/yr. The individual layer contributions can also be summed on the basis of typical mean ozone profiles to derive an error in the SAGE II ozone column (which excludes the tropospheric contribution). The trend error is $-0.2\%/yr$ in the tropics and between zero and $-0.1\%/yr$ at midlatitudes.

Figure 12 shows the trends in columnar ozone over the period of November 1984 to May 1991 inferred from the SAGE II ozone measurements and from the total ozone mapping spectrometer (TOMS) ozone measurements. Each is expressed as a percentage of the mean column measured by that instrument. The SAGE II trends will refer to the stratosphere only, whereas the TOMS trends will include both stratospheric and tropospheric contributions. The TOMS measurements utilized in this analysis include only profiles coincident with the SAGE II measurements, i.e., using the closest TOMS profile

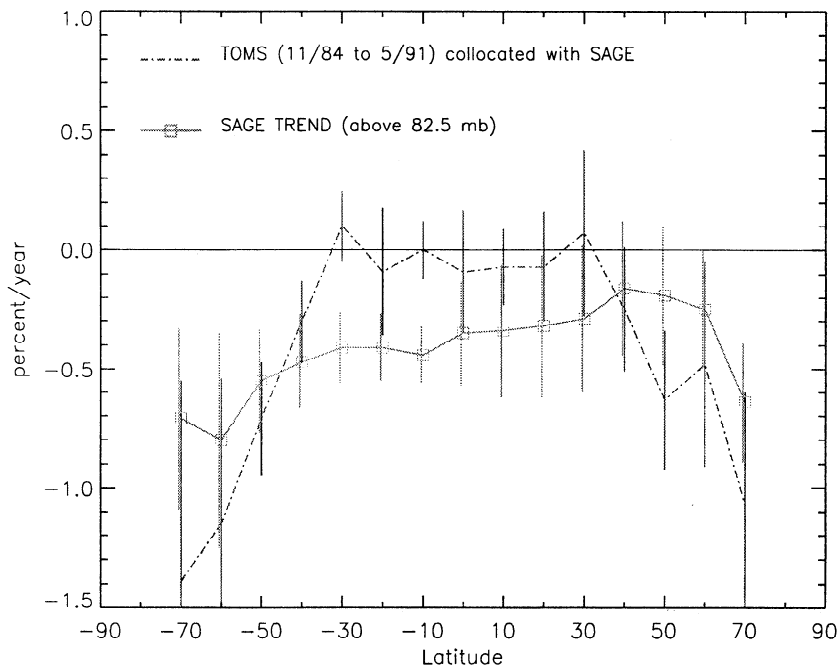


Figure 12. Columnar ozone trends above 82.5 mbar in 10° latitude bands over the period November 1984 to May 1991. Similar models have been applied to SAGE II measurements and coincident total ozone mapping spectrometer measurements.

for each SAGE II measurement. The model which has been fitted here consists of a linear trend, annual and semiannual cycles, a quasi-biennial contribution (assuming a 30-month period), and a solar cycle contribution based on the 10.7-cm flux.

The SAGE II columnar ozone trends (using the version 5.9 data) down to the bottom of the 68.1-mbar UARS level (82.5 mbar) are reported in Figure 12. This is consistent with the 17-km cutoff on the lower stratosphere ozone trends reported by McCormick *et al.* [1992]. The important result shown in Figure 12 is that at latitudes less than 30° the TOMS ozone trend exceeds that from SAGE II by approximately 0.4%/yr. The calculations in this paper suggest that one half of this difference may be associated with a downward bias in the SAGE II trends caused by aerosol retrieval errors. The remaining difference in the trends may be caused by increases in tropospheric ozone. However, because of the uncertainties in the calculated sensitivities to aerosol interference and possible TOMS-related uncertainties (e.g., cloud effects [Thompson *et al.*, 1993]), it would be dangerous to infer trends in tropospheric ozone in the tropics from these trend differences.

Figure 9 indicates that stratospheric aerosol concentrations decreased until 1988 when they reached a fairly stable background level. Between 1988 and May 1991 the effects of aerosol interference on SAGE II ozone retrievals should certainly be negligible. Therefore to complement the above analysis, it is useful to compare SAGE II and TOMS columnar ozone trends over just this period. Figure 13 then has been constructed in a similar way to Figure 12 but just for the period January 1988 to May 1991. It may be seen that during this period the differences between the TOMS and the SAGE II trends in the tropics in particular are insignificant (although the uncertainties are, of course, larger than for the longer period). The similarity in the trends from the two data sets suggests that the tropospheric ozone trends during this period were similar to those in the lower stratosphere.

6. Conclusions

SAGE II ozone profile measurements have been compared against coincident measurements by the MLS instrument over the period September 1991 to December 1993. It is found that the differences are consistent with the stated precisions and accuracies of the individual measurement systems except in the lower stratosphere where the SAGE II ozone measurements are evidently affected by the large aerosol concentrations produced by the Mount Pinatubo eruption. Over the altitude range from 1.5 to 15 mbar, each system is expected to measure ozone in UARS layers, which are approximately 2.6 km thick, with a precision and accuracy close to 5%. Over this altitude range the MLS measurements systematically yield approximately 5% larger values than the SAGE II measurements. Version 4 MLS retrievals are projected to reduce the ozone values by a few percent (at least above 10 mbar), as discussed by Froidevaux *et al.* [this issue].

In the altitude range above 10 mbar it has been demonstrated that the monthly mean differences are remarkably independent of space and time of the year. There is, however, an altitude structure to the differences with the MLS measurement being equal to or slightly less than the SAGE II measurements at 1 mbar and with the differences undergoing a vertical oscillation at lower pressures. This may be related at least in part to the MLS measurements currently being retrieved on even UARS levels only. It has been noted that SAGE II sunrise and sunset measurements do not give the same ozone values near the 1-mbar level and that the reasons for this are under investigation.

At pressure levels greater than 10 mbar the comparisons indicate and simulated retrievals confirm that SAGE II ozone values are being positively biased by a failure to accurately remove the contributions from the exceptionally large aerosol concentrations arising from Mount Pinatubo. At these large

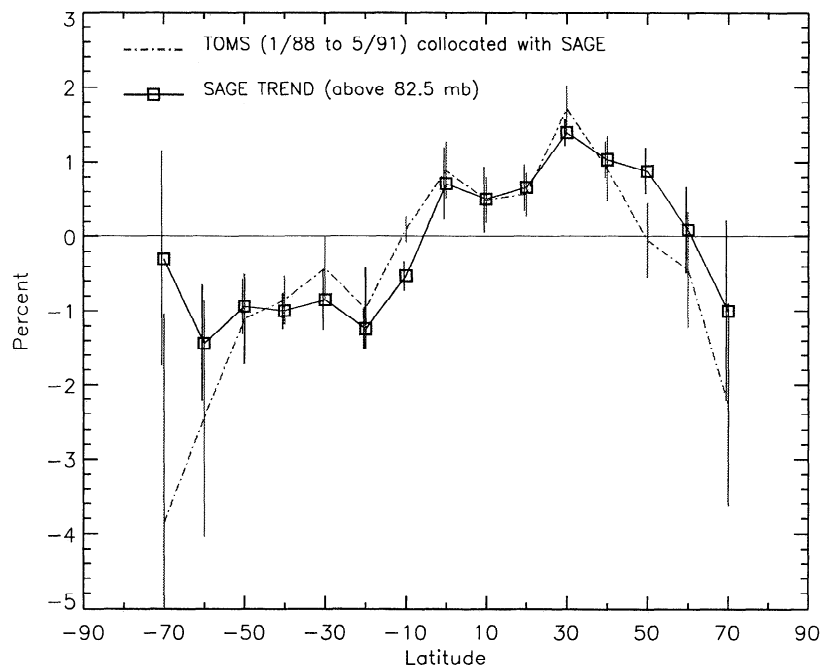


Figure 13. As in Figure 12 but for the period January 1988 to May 1991; note, however, a factor of approximately 3 change in scale on the ordinate compared with Figure 12.

aerosol concentrations, for which the aerosol extinction at $1.02 \mu\text{m}$ is approximately equal to that at $0.525 \mu\text{m}$, the ozone concentration is biased in direct proportion to the aerosol layer optical depth, with a sensitivity factor of $7 \times 10^{10} \text{ cm}^{-3}/10^{-3}$ aerosol optical depth in a UARS layer. This is equivalent to an underestimation of the aerosol extinction at $0.6 \mu\text{m}$ by approximately 8%.

This sensitivity to aerosol retrieval errors has been reestimated for aerosol conditions typical of the 1980s. For this purpose the relationship between SAGE II/MLS ozone differences and the aerosol layer optical depth was reexamined for aerosol optical depths of less than 5×10^{-3} at $0.525 \mu\text{m}$ and of less than 2×10^{-3} at $1.02 \mu\text{m}$. The previously calculated sensitivities still applied at $1.02 \mu\text{m}$ at altitudes above the 20-mbar level and consistent with the ratio of the $0.525\text{-}\mu\text{m}$ aerosol extinction to that at $1.02 \mu\text{m}$ being approximately 2.5, the sensitivities at $0.525 \mu\text{m}$ were a factor of approximately 2.5 smaller. The sensitivities at 21.5 and 31.6 mbar were, however, found to be much less for the reduced aerosol loading conditions but to both equal $3 \times 10^{10} \text{ cm}^{-3}/10^{-3}$ aerosol layer optical depth at $1.02 \mu\text{m}$. This sensitivity was assumed to also apply at 46.4 and 68.1 mbar. This is equivalent to an underestimation of approximately 3% in the inferred aerosol extinction at $0.6 \mu\text{m}$ in the SAGE II ozone retrievals.

Based on this sensitivity, tropical ozone trends inferred from version 5.9 SAGE II retrievals were estimated to be biased downward by approximately 1%/yr at 46.4 and 68.1 mbar over the period 1984 to 1991, and therefore the total stratospheric ozone column trends were estimated to be biased downward by approximately 0.2%/yr in the tropics. This accounts for approximately one half of the difference between SAGE II and TOMS ozone trends over this period in the tropics. Because of the large error bars on the sensitivities, the remaining differences in the tropical trends are not considered to be statistically significant. The aerosol effect on SAGE II tropical column ozone trends above 38.3 mbar and on midlatitude column

trends above 82.5 mbar is less than 0.1%/yr. Moreover, the effect at individual UARS levels above these lower boundaries is estimated to be no more than 0.2%/yr, again over the period 1984 to May 1991. It was shown that atmospheric aerosol concentrations were particularly low from 1988 to May 1991 and that over this period, SAGE II and TOMS ozone trends were similar. The magnitudes of these trends is of considerable importance for climate studies [*World Meteorological Organization (WMO)*, 1991]; these comparisons should therefore be continued as Mount Pinatubo aerosols decay further and as revised versions of the MLS retrievals become available, which may provide improved ozone values at pressures greater than 40 mbar.

Acknowledgments. We thank James Russell, head of the UARS-HALOE team for providing the excellent ozone measurements and for allowing us to include the month of HALOE data in these comparisons. This research was supported by contracts NAS5-27264 and NAS1-19954 to Georgia Tech and by the Upper Atmospheric Research Satellite project at the Jet Propulsion Lab.

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(Received November 16, 1994; revised March 4, 1995; accepted May 27, 1995.)