



# Trends in the Vertical Distribution of Ozone

William J. Randel,<sup>1\*</sup> Richard S. Stolarski,<sup>2</sup> Derek M. Cunnold,<sup>3</sup> Jennifer A. Logan,<sup>4</sup> M. J. Newchurch,<sup>5</sup> Joseph M. Zawodny<sup>6</sup>

Analyses of satellite, ground-based, and balloon measurements allow updated estimates of trends in the vertical profile of ozone since 1979. The results show overall consistency among several independent measurement systems, particularly for northern hemisphere midlatitudes where most balloon and ground-based measurements are made. Combined trend estimates over these latitudes for the period 1979–96 show statistically significant negative trends at all altitudes between 10 and 45 km, with two local extremes:  $-7.4 \pm 2.0\%$  per decade at 40 km and  $-7.3 \pm 4.6\%$  per decade at 15 km altitude. There is a strong seasonal variation in trends over northern midlatitudes in the altitude range of 10 to 18 km, with the largest ozone loss during winter and spring. The profile trends are in quantitative agreement with independently measured trends in column ozone, the amount of ozone in a column above the surface. The vertical profiles of ozone trends provide a fingerprint for the mechanisms of ozone depletion over the last two decades.

Concentrations of ozone in the atmosphere change as a result of both natural and human activities, and accurate knowledge of past changes is important for attribution of causes and prediction of future changes. The distribution and trends in atmospheric ozone affect the climate system in several ways. Ozone concentration peaks in the stratosphere at an altitude of 20 to 27 km (Fig. 1A), and ozone's atmospheric distribution is maintained by a balance between photochemical production and loss and by transport from regions of net production to net loss (1). Ozone strongly absorbs solar ultraviolet (UV) radiation. A reduction in ozone therefore results in increased UV radiation levels in the troposphere and at the Earth's surface. These increased UV levels are associated with increases in tropospheric photochemical reactivity, which directly influences tropospheric CO and CH<sub>4</sub> trends (2). Ozone also absorbs and emits infrared radiation and is an effective greenhouse gas. Changes in stratospheric ozone concentration therefore strongly affect the temperature of the stratosphere itself and also have an impact on the troposphere. Recent estimates suggest that up to 30% of the surface and tropospheric warming resulting from greenhouse gas increases since 1980 may have been offset by the cooling effects of stratospheric ozone loss (3). In

order to assess and model the impacts of changes in ozone in recent decades quantitatively, knowledge of the vertical structure of these changes is crucial.

Measurements of the integrated amount of ozone in a vertical column above the surface (so-called column ozone) are provided on a routine basis by a global network of ground-based instruments and calculated from satellite data. Long-term records of these data show conclusive evidence of a decrease in column ozone over large regions of the globe during recent decades (4–6). The largest decreases in column ozone are observed over the Antarctic during spring and over middle to high latitudes of the northern hemisphere during winter and spring. In general, there is good agreement between ground-based and satellite-measured trends in column ozone (5). In contrast, vertical profiles of ozone trends (from ground-based, balloon-borne, and satellite measurements) have shown considerable uncertainty in the past because of many fewer stations with good quality long-term records, and because of large differences between balloon-borne and satellite results (6). A reevaluation of the vertical distribution of ozone trends for 1979–96 was recently carried out by a large group of research scientists (7). The basis for this reevaluation was a renewed effort to improve quality control in the long-term ozone sonde database, combined with improved treatment of the satellite data sets. Here we highlight the main scientific results of trend analyses from these improved data sets and update satellite and ground-based results through 1997–98.

## Sources of Data

No uniform data source exists to allow global estimation of long-term changes in the vertical ozone distribution. The data come from a num-

ber of ozone measuring platforms, which use different sensing techniques with varying spatial and temporal coverage. Four different measurement systems have been producing records long enough for the assessment of long-term ozone profile trends. A network of stations provides ozone profile information from balloon-borne sondes, which measure ozone from the ground to about 30 km, with a vertical resolution of ~150 m. The sondes use the reaction of ozone with potassium iodide in an aqueous solution for detection. Ozone sonde data are available since about 1970 from 11 stations located in northern middle and high latitudes; additional data are available from a few other stations since 1980 or later.

Ground-based measurements use the Umkehr technique for measuring the profile of ozone concentration (8). It consists of observing a series of ultraviolet wavelengths of solar radiation, some of which are strongly absorbed by atmospheric ozone. A sequence of radiance measurements is made as the sun rises or sets, and information about the ozone profile is obtained from an inversion algorithm applied to these measurements (9). The Umkehr technique provides ~5-km vertical resolution for altitudes between 25 and 40 km, along with low-resolution estimates above and below these altitudes. Eight Umkehr stations have records of sufficient length for trend evaluation, with four of them located in northern hemisphere midlatitudes.

Satellite measurements of the vertical profile of stratospheric ozone have been made as part of the Stratospheric Aerosol and Gas Experiment (SAGE) (10). SAGE I data are available from February 1979 to November 1981. SAGE II began operation in October 1984 and continues to make measurements to the present. The SAGE measurement technique is based on solar occultation, with profile measurements obtained at sunrise and sunset on each of 14 orbits per day. The technique provides good vertical resolution (~1 km) and very small long-term drift resulting from instrument calibration, but spatial sampling is limited, and it takes approximately 1 month to sample the latitude range 60°N to 60°S. A difficulty in combining the SAGE I and SAGE II ozone measurements for trend studies arises from an apparent error in the reference altitude for SAGE I. An empirical altitude correction has been applied to SAGE I data in an attempt to remove this bias (11). The SAGE II data used here are from retrieval version 5.96, with special attention paid to removing aerosol contamina-

<sup>1</sup>National Center for Atmospheric Research, Boulder, CO 80307, USA. <sup>2</sup>National Aeronautics and Space Administration (NASA), Goddard Space Flight Center, Greenbelt, MD 20771, USA. <sup>3</sup>Georgia Institute of Technology, Atlanta, GA 30332, USA. <sup>4</sup>Harvard University, Cambridge, MA 02138, USA. <sup>5</sup>University of Alabama in Huntsville, Huntsville, AL 35899, USA. <sup>6</sup>NASA Langley Research Center, Hampton, VA 23681, USA.

\*To whom correspondence should be addressed. E-mail: randel@ucar.edu

tion effects in the lower stratosphere. Because problems remain during periods of exceptionally high aerosol loading, SAGE II data are not used in the lower stratosphere for 1 to 2 years following the eruption of Mount Pinatubo in 1991 (12). Detailed comparisons between SAGE II and all available data sets [as reported in (7)] demonstrated no statistically significant drift in SAGE II data over the altitude range 15 to 50 km. Below 20 km the empirical altitude correction applied to SAGE I data (11) may be incorrect, and trends from combined SAGE I/II data are considered most reliable above 20 km (7).

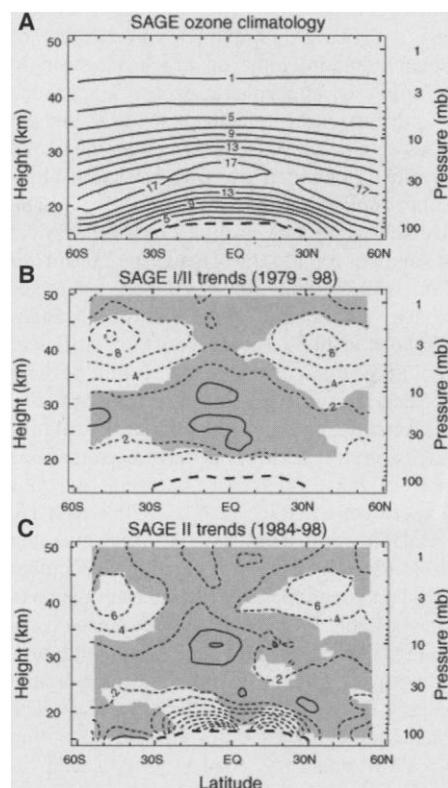
Satellite measurements of ozone profile were also made from the solar backscatter ultraviolet (SBUV) instrument on board the NIMBUS-7 satellite and SBUV/2 on NOAA-11, covering the periods November 1978 to May 1990 and January 1989 to October 1994, respectively. These data are based on measurements of the Earth's ultraviolet albedo at several wavelengths. An inversion algorithm then produces ozone profile estimates with

relatively low vertical resolution similar to the Umkehr profiles (~5 km over 25 to 45 km). A combined data set [termed SBUV(2)] has been created for trend analysis (13). An important aspect of the SBUV/2 data is that the orbit of the NOAA-11 satellite precessed in time, resulting in a loss of spatial coverage in the latter part of the record. Because of this loss of data and potential problems with the SBUV/2 calibration, less confidence is placed in the trends derived from SBUV(2).

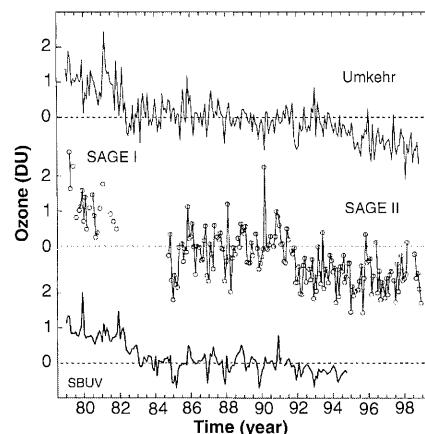
Linear trends were derived for each data set by use of multiple linear regression analyses (7). The regressions include terms that explain known sources of variability in stratospheric ozone resulting from changes in the 11-year solar cycle and the quasi-biennial oscillation (QBO) in stratospheric winds. Extensive analyses of test data sets showed that the overall trend results are insensitive to the details of the regression model used (14).

### Trends in the Upper Stratosphere

Changes in ozone were originally predicted to occur in the upper stratosphere (altitudes between about 30 and 50 km) as a result of anthropogenic chlorine increases (15). At these altitudes ozone is essentially in photochemical equilibrium, and the chemistry should be dominated by gas-phase reactions. Trends above 30 km can be derived from SAGE I/II, SBUV(2) and Umkehr data. Time series of these data over 40° to 50°N near 40 km (Fig. 2) show a general decline over the period from 1979 to 1998. The decreases appear most rapid during the periods 1982–83 and 1991–94, with relatively small changes otherwise; this time behavior is consistent with a decreasing trend superimposed onto the 11-year solar cycle. A comparison of trends derived from these data (Fig. 3) shows



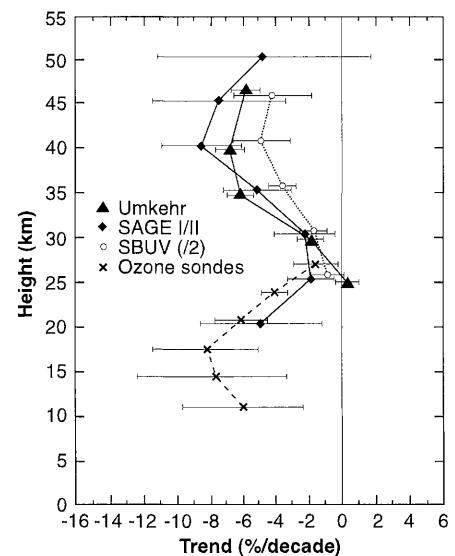
**Fig. 1.** (A) Time mean (1979–98) climatology of ozone density derived from SAGE I/II data. Units are Dobson units (DU)/km (29). The heavy dashed line represents the tropical tropopause. (B) Annual mean ozone trends calculated from SAGE I/II observations for 1979–98, expressed in percent per decade of the mean of the time series. Shading indicates where the trends are not different from zero within 95% confidence limits. Because of uncertainties in SAGE I data, results are not shown below 20 km. (C) Annual mean ozone trends calculated from SAGE II only data for 1984–98.



**Fig. 2.** Time series of ozone anomalies between 38 and 43 km for latitudes 40°–50°N from Umkehr, SAGE I/II, and SBUV(2) data. Umkehr data are the average of Arosa, Belsk, Boulder, and Haute Provence measurements. The time mean and average seasonal cycle have been removed from each time series.

statistically significant negative trends in all three data sets over 30 to 45 km, with peak values of –6 to –8% per decade near 40 km (16). There is good agreement between SAGE I/II and Umkehr results, whereas the (shorter) combined record from SBUV(2) shows less negative trends.

A global cross section of trends was derived from the combined SAGE I/II data for the extended period 1979–98 (Fig. 1B). Statistically significant negative trends are seen in the upper stratosphere (35 to 45 km), with maxima on the order of –8% per decade over middle to high latitudes in both hemispheres. Trends in the tropical upper stratosphere are less negative and not statistically significant, and there is a clear global symmetry in the trends above 30 km. The SAGE I/II trends (Fig. 1B) show a relative minimum over altitudes 25 to 30 km and increased negative trends in the lower stratosphere. The magnitude of the upper stratospheric ozone loss and the spatial pattern of maxima over high latitudes are consistent with idealized model calculations of ozone loss as a result of increasing stratospheric chlorine of anthropogenic origin (17). Ozone trends derived from the shorter SAGE II only record (1984–98) (Fig. 1C) show similar spatial structure, but a reduction in the magnitude of upper strato-



**Fig. 3.** Ozone trends (percent per decade relative to the mean for the period) over 40°–53°N for the period 1979–96 (16), calculated from SAGE I/II and SBUV(2) satellite data, the average of three Umkehr stations (Boulder, Haute Provence, and Belsk), and the average of seven ozone sonde stations (Boulder, Sapporo, Payenne, Hohenpeissenberg, Uccle, Edmonton, and Goose Bay). Horizontal error bars show 95% confidence intervals of the trend; SAGE I/II error bars include SAGE I altitude correction uncertainties, SAGE II sunrise/sunset trend difference uncertainties, and statistical uncertainties. Umkehr, SBUV(2), and ozone sonde error bars represent statistical uncertainty only.

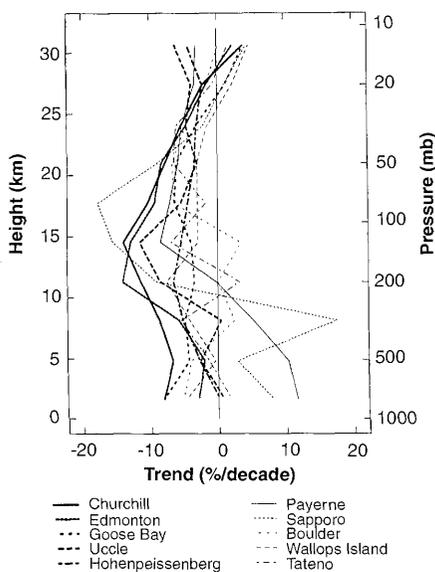
spheric trends to about  $-6$  to  $-7\%$  per decade [consistent with the recent flatness of the time series through 1998 (Fig. 2)].

### Trends in the Lower Stratosphere

Much of the change deduced from column ozone data is expected to occur in the lower stratosphere (altitudes below 30 km). The primary instruments that measure ozone in this region are sondes (up to 27 km) and SAGE (15 to 30 km). The annual mean global SAGE I/II data (Fig. 1B) show negative trends in the lower stratosphere over most of the globe, with statistically significant changes over northern hemisphere middle and high latitudes ( $40^\circ$  to  $55^\circ\text{N}$ ) and southern hemisphere low to midlatitudes ( $15^\circ$  to  $50^\circ\text{S}$ ). Geographical overlap between SAGE and sonde data over northern hemisphere midlatitudes allows detailed comparisons.

Annual mean trends for 1980 to 1996 from ten ozone sonde stations located from  $36^\circ$  to  $59^\circ\text{N}$  (Fig. 4) are negative at each station between about 12 and 25 km (7, 18). Maximum annual trends of about  $-2$  to  $-12\%$  per decade are found for 15 to 20 km for most stations in this latitude band, and the trends are statistically significant for 8 of the 10 stations.

Mean trends in ozone for the seven sonde stations located between  $40^\circ$  to  $53^\circ\text{N}$  were compared with zonal mean trends from SAGE I/II derived over approximately the same period (Fig. 3) (19). Both data sets show statistically significant negative trends over 20 to 25 km. The averaged ozone sonde trends in this region are somewhat more negative than zonal mean SAGE results, al-

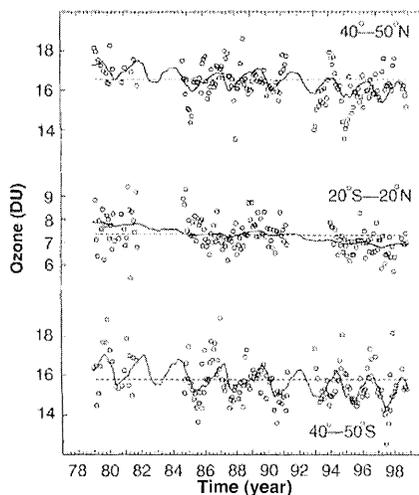


**Fig. 4.** Annual trends in ozone over 1980–96 derived from ozone sonde measurements. Shown here are superimposed trend results for individual stations located between  $36^\circ$  and  $59^\circ\text{N}$ .

though they agree within  $2\sigma$  uncertainty levels. This approximate agreement in the lower stratosphere is a significant improvement over previous comparisons (6), and mainly results from the inclusion of the latitude-dependent altitude correction to SAGE I data (11).

Trends calculated from SAGE II data only (spanning 1984–98) allow extension of the trends down to 15 km. The SAGE II data (Fig. 1C) exhibit negative trends over much of the globe in the lower stratosphere, with largest percentage changes (on the order of  $-10\%$  per decade) in the tropics below 20 km. Time series of SAGE data at 20 km (Fig. 5) show the reduction in tropical ozone with time; overall decreases are also observed over northern and southern midlatitudes, together with significant components of other interannual variability (20). Although the SAGE II v5.96 data show a negative trend in the tropical lower stratosphere, satellite measurements in this region are sensitive to details of the retrieval algorithm (21); preliminary analyses of a nonpublic, unvalidated new version of the data (v6.0a) show much smaller, statistically insignificant trends (about  $-2\%$  per decade) in the tropics near 20 km. Furthermore, no long-term ozone sonde records are available for independent estimation of tropical trends, and hence the possibility of large negative trends above the tropical tropopause remains a topic of continued research.

Averaging the northern midlatitude ozone sonde trends (Fig. 6) reveals large seasonal variation between 10 and 18 km altitude, with maximum negative trends during northern hemisphere winter and spring. Relatively little seasonal variation is observed above 20



**Fig. 5.** Time series of SAGE I/II ozone between 19.5 and 20.5 km, at northern and southern midlatitudes and over the tropics. The mean seasonal cycle has been removed at each location to accentuate interannual variations. The solid lines show the statistical regression model fit of the data at each location.

km. Direct comparisons between the vertically integrated ozone sonde profile trends and those derived from independent TOMS (total ozone mapping spectrometer) data (22) over northern midlatitudes (Fig. 6) show reasonable agreement in both magnitude and seasonality. This consistent behavior demonstrates that the observed seasonal maximum in column ozone trends during northern hemisphere winter-spring results from trend variations in the altitude range 10–18 km.

### Combined Trend at Northern Midlatitudes

An overall ensemble estimate of the ozone trends over northern midlatitudes for the period 1979–96 was calculated using results from all four measurement types (Table 1); the uncertainties are based on combined statistical and instrumental drift uncertainties (23). Because of the large geographic variability in tropospheric trends (Fig. 4), it is not reasonable to produce a mean troposphere estimate and ensemble results are not included below 10 km. Trends in northern midlatitudes from the combined analyses show statistically significant negative trends over all altitudes between 10 and 45 km. The combined trend (Table 1) has two local extremes, at  $-7.4 \pm 2.0\%$  per decade at 40 km and at  $-7.3 \pm 4.6\%$  per decade at 15 km. The smallest trend of  $-2.0 \pm 1.8\%$  per decade is deduced at 30 km at midlatitudes.

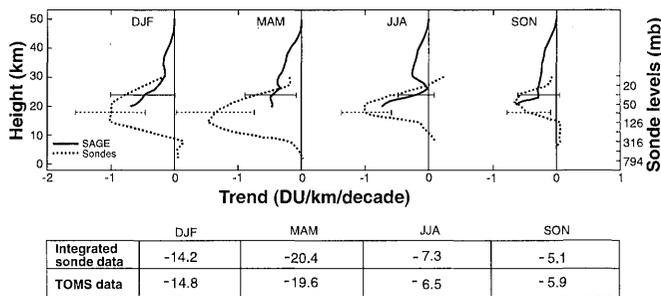
### Summary and Outlook

Ozone measurements spanning the last 20 years show clear evidence of ozone decreases in the upper stratosphere (maximum near 40 km) and lower stratosphere (15 to 20 km). Upper stratosphere ozone loss maximizes in middle to high latitudes of both hemispheres, with remarkable global symmetry. There is good quantitative agreement between the observed latitudinal and vertical structure of upper stratospheric ozone trends and idealized model calculations of ozone loss due to

**Table 1.** Combined trends and uncertainties (in percent per decade) for ozone profile trends over  $40^\circ$  to  $50^\circ\text{N}$ . The uncertainty estimates (given as  $1\sigma$ ) combine statistical and instrumental drift components.

Altitude (km)	Trend	Uncertainty
50	-4.5	3.3
45	-6.1	1.6
40	-7.4	1.0
35	-5.0	0.9
30	-2.0	0.9
25	-3.1	0.5
20	-5.1	0.9
17.5	-6.8	1.7
15.0	-7.3	2.3
12.5	-6.4	2.0
10.0	-3.3	1.8

**Fig. 6.** Altitude profiles of ozone trends for 1979–96 over 40° to 53°N for SAGE I/II and averaged ozone sonde data. These results show seasonal variation in the trends, that is, results for December–February, March–May, June–August, September–November. Results are expressed in terms of ozone density tendency (DU/km/decade) in order to directly illustrate the contribution to column ozone trends (29). The vertical integrals of the ozone sonde trends over 0 to 30 km are listed at the bottom (with units of DU/decade), together with independent estimates of column ozone trends derived from TOMS satellite data for 1979–94.



anthropogenic chlorine emissions (24), suggesting basic understanding of ozone depletion in this altitude region. The abundance of chlorine in the upper stratosphere is expected to peak in the near future and then slowly decline (25), and it is anticipated that this chlorine decrease will correspond to a slow recovery of upper stratospheric ozone towards pre-1980 values (26).

Long-term decreases in lower stratosphere ozone show reasonable agreement between SAGE satellite measurements and ozone sonde data over 15 to 27 km for northern midlatitudes. The major contribution to column ozone decline over these latitudes during the last two decades has occurred over altitudes 10 to 25 km. There is a strong seasonal variation to northern midlatitude ozone loss over 10–18 km, with maximum loss during winter and spring. The vertical, latitudinal, and seasonal variations of northern midlatitude ozone depletion are consistent with anthropogenic halogens as the cause (27). However, the abundance of lower stratospheric ozone is crucially dependent on transport, so that interannual variations in dynamic behavior can also be a source of ozone variability (20, 28). Based on projections of halogen content of the stratosphere, the maximum lower stratosphere ozone depletion is estimated to lie within the current decade or the next two decades, followed by slow recovery (26).

Current analyses of SAGE II data suggests large percentage ozone loss in the tropical lower stratosphere during 1984–98. Such tropical ozone depletion is not predicted by current model calculations (26). However, lower stratospheric ozone trends are difficult to estimate from satellite measurements in the tropics (21), and independent estimates from ozone sondes are not available. Estimates of long-term ozone changes in this region are a topic of continued research.

#### References and Notes

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- Time periods when SAGE II data are omitted follow the recommendations in Table 2.2 of (7).
- The SBUV and SBUV/2 data are combined by adjusting the SBUV time series by a small latitude-dependent offset, which is determined by differences observed during the overlap period January to December 1989.
- The comparison of trend statistical models in (7) suggests caution in analysis of time series with frequently missing observations (such as SAGE data). However, for ozone profile observations, the trend uncertainty levels due to natural and sampling variability are usually much larger than those associated with details of the regression model.
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- The trend comparisons in Fig. 3 are based on the period 1979–96 for SAGE I/II and Umkehr, 1980–96 for ozone sondes, and 1979–94 for SBUV/2. The 1- or 2-year differences in time periods do not have a large effect on trend estimates for these relatively long time records.
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- The space-time sampling of SAGE measurements do not allow a sufficient number of direct overlap comparisons with individual ozone sonde time series. In order to minimize statistical uncertainties, available ozone

sonde trends over northern midlatitudes (in Europe, North America, and Japan) are averaged and compared with trends derived from zonal mean SAGE data.

- Interannual variability in ozone density in the extratropical lower stratosphere is large because of enhanced meteorological variability inherent to the climate system. The QBO also contributes a large component of variance in this region; note the approximate 2-year variations in the regression fits at 40°–50°N and 40°–50°S in Fig. 5.
- Accurate satellite measurements of ozone in the tropical lower stratosphere are difficult because: (i) there is low ozone density in this region (Fig. 1A), (ii) this region is ~7 km below the tropical ozone maximum (Fig. 1A), through which the satellite line of sight must travel, (iii) there are problems in accurately tracking the sun at low altitudes, and (iv) the presence of enhanced aerosol contaminates the ozone retrieval in this region.
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- Trends from each measurement type were derived with an associated estimate of statistical uncertainty. An additional uncertainty results from the potential drift of the instrument over time, and the total uncertainty is a root sum of squares addition of statistical and instrumental terms. These uncertainties have been evaluated for each instrument type (7). The average trends and uncertainty estimates in Table 1 are derived from a weighted mean of the individual measurement systems, where available (with weighting by the inverse square standard error for each system).
- R. Müller et al., in "Scientific Assessment of Ozone Depletion: 1998" (WMO/U.N. Environ. Prog. Rep. 44, WMO, Geneva, 1999), chap. 6.
- Concentrations of chlorine in the troposphere peaked during 1992–94, and are now declining in response to the 1987 Montreal Protocol on Substances that Deplete the Ozone Layer and its subsequent amendments. Chlorine in the upper stratosphere is expected to peak before the year 2000; the approximate 6-year time lag reflects the average time required for surface emissions to reach the upper stratosphere.
- Detailed model simulations of past and future ozone changes are presented by D. J. Hofmann et al., in "Scientific Assessment of Ozone Depletion: 1998" (WMO/U.N. Environ. Prog. Rep. 44, WMO, Geneva, 1999), chap. 12. The future rate of ozone recovery in the upper stratosphere will probably be slower than the observed 1979–98 decreases, because of cooling stratospheric temperatures and increases in N<sub>2</sub>O and CH<sub>4</sub>. Calculated recovery in the lower stratosphere is sensitive to these variations as well as to details of the halogen and sulfate aerosol changes.
- A. Ravishankara et al., in "Scientific Assessment of Ozone Depletion: 1998" (WMO/U.N. Environ. Prog. Rep. 44, WMO, Geneva, 1999), chap. 7.
- Some component of the northern midlatitude lower stratospheric ozone trends may be associated with low frequency variations of stratospheric circulation, as suggested by L. L. Hood et al., *J. Geophys. Res.* **102**, 13079 (1997); K. Pentzoldt, *Ann. Geophysicae* **17**, 231 (1999); A. Fusco and M. Salby, *J. Clim.* **12**, 1619 (1999).
- The amount of ozone in a vertical column above Earth's surface is often expressed in terms of Dobson units (DU), where 1 DU ≈ 2.7 × 10<sup>20</sup> molecules of ozone per square meter. Typically observed values are of the order 300 DU. Ozone density vertical profile measurements may be expressed in terms of DU/km (1 DU/km ≈ 2.7 × 10<sup>17</sup> molecules/m<sup>3</sup>); the vertical integral of such data then provides column ozone in units of DU.
- The Stratospheric Processes and their Role in Climate (SPARC) and International Ozone Commission (IOC) ozone trends activities and report (7) were the result of a concerted effort of over 80 scientists spanning several years. These contributors are listed in (7). This effort was undertaken jointly by SPARC and the IOC, in close cooperation with the WMO Global Atmospheric Watch (GAW) program. The National Center for Atmospheric Research is sponsored by the National Science Foundation.