

An analysis of ozonesonde data for the lower stratosphere: Recommendations for testing models

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Abstract. I present an analysis of ozonesonde data with a focus on using these data to evaluate stratospheric models of transport and chemistry. Ozonesondes are the only instruments that provide year-round profiles of ozone throughout the lowermost stratosphere for middle and high latitudes of the Northern Hemisphere. I show vertical profiles and the annual cycle of ozone at selected pressure levels and stations and use TOMS data to evaluate the spatial bias of the sonde stations with respect to zonal mean values of column ozone. Between 10 and 30% of the ozone column is located between 100 hPa and the tropopause at middle and high latitudes, and this region drives much of the seasonal variation of the ozone column. The sonde data allow quantification of the buildup of ozone in the lowermost stratosphere of the Northern Hemisphere in winter and of its loss in late spring and summer. The amount of ozone between the tropopause and 100 hPa in the Northern Hemisphere decreases from 175 to 75 Tg from March to September, with the maximum rate of decrease in May to June; about half of the decrease is caused by the increase in the height of the tropopause. There is a lag of about a month in the time when ozone starts to accumulate in the lowermost stratosphere, from September near 40°N to October or November near 80°N, and a similar lag in the time when ozone starts to decrease, from February or March near 40°N to March or April near 80°N. I propose that the sonde data be used to provide a test of transport in two- and three-dimensional models in addition to those provided by long-lived tracers, such as N₂O, CFCs, CO₂, CH₄, and their correlation patterns, and age of air. The annual variation of the ozone content of the lowermost stratosphere in the Northern Hemisphere is suggested as a particularly useful constraint on transport parameterizations. I make recommendations for the use of particular stations and provide summary statistics for ozone concentrations and for the mean tropopause height for each station.

1. Introduction

Assessment of the effects of human activity on the distribution of stratospheric ozone has relied primarily on two-dimensional models during the last decade [World Meteorological Organization (WMO), 1986, 1990, 1992, 1995, 1999]. These models have been used to predict the effects of halocarbon emissions and of a hypothetical fleet of supersonic aircraft on ozone [e.g., WMO, 1995; Stolarski *et al.*, 1995]. Recently, some of these models have been used to simulate the observed decrease in ozone in the lower stratosphere [Solomon *et al.*, 1996, 1998; Jackman *et al.*, 1996; Callis *et al.*, 1997] with a reasonable degree of success; the models tend to underestimate ozone loss in the lowermost stratosphere. Three-dimensional models are starting to be used for assessment purposes [e.g., Shindell *et al.*, 1998].

The ability of these models to simulate the present-day distribution of ozone has been tested using data for the total column and selected data for the vertical profile of ozone in the stratosphere. The ozone column predicted by two-dimensional (2-D) models for middle to high latitudes is particularly sensitive to advection below 100 hPa, where computation of the heating rates used to drive the residual circulation is most uncertain [Jackman *et al.*, 1989]. In many models, horizontal diffusion coefficients

in the lower stratosphere and vertical diffusion coefficients in the troposphere are adjusted to obtain a reasonable depiction of column ozone. Even so, most models tend to overestimate ozone in the lower stratosphere at middle and high latitudes in winter and spring [e.g., Prather and Remsberg, 1993; WMO, 1995; Park *et al.*, 1999]. Evaluation of the vertical distribution of trace gases in two- (and three-) dimensional models has generally been limited to the region above 100 hPa (~17 km) [e.g., Prather and Remsberg, 1993; Rasch *et al.*, 1995; Jackman *et al.*, 1996], and sonde data have not been used although they provide unique information on the vertical distribution of ozone in the lowermost stratosphere. To my knowledge, Garcia and Solomon's [1994] is the only published study that used sonde data to evaluate a 2-D model, and they show profiles from two locations. The first Models and Measurements Workshop used vertical profiles of ozone above 16 km derived from satellite data to evaluate 2-D models, in addition to the ozone column, but the comparison emphasized the region above the ozone peak at ~24 km [Prather and Remsberg, 1993]. The recent Models and Measurements Workshop (MM II) [Park *et al.*, 1999] used a preliminary climatology based on ozone sonde data analyzed here and SAGE II data, and examined the performance of both 2-D and 3-D models in the lowermost stratosphere as well as at higher altitudes. Long-lived tracers such as N₂O, CFCs, CO₂, CH₄, SF₆, and aerosols and analyses of age of air were used to test the dynamics in these models [e.g., Garcia and Solomon, 1994; Jackman *et al.*, 1996; Park *et al.*, 1999; H. R. Schneider *et al.*, Analysis of residual mean transport in the stratosphere, 1, Model description and

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comparison with satellite data, submitted to *Journal of Geophysical Research*, 1999], as were correlations of long-lived tracers [e.g., *Plumb and Ko*, 1992; *Prather and Remsberg*, 1993], but the data available at present do not provide good constraints on transport in the extratropical lowermost stratosphere. A comparison of five two-dimensional models showed that they computed about the same midlatitude profile for ozone above 26 km (22 hPa) but that they gave ozone values that differed by 30-100% from 12 to 20 km (200-55 hPa), with the differences attributed in part to transport in the models [*Stolarski et al.*, 1995]. The response of these models to a projected fleet of supersonic aircraft was a modest change in the ozone column, but the predicted difference in ozone in the region from 12 to 20 km varied over a factor of 5 and changed sign, depending on the model. Results from MM II showed that models tend to exceed measured ozone values by about 10% near 25-30 km. Differences between modeled and observed ozone were largest in the lowermost stratosphere, with errors up to 50% for the best performing models and errors exceeding 100% for the worst performing models [*Logan and McPeters*, 1999].

I propose here that ozonesonde measurements of the vertical profile of ozone in the lowermost stratosphere should be used as an additional constraint on transport in 2-D and 3-D models, particularly in the Northern Hemisphere, where the density of stations is highest. Ozonesondes are the only instruments that provide year-round profiles of ozone throughout the lowermost stratosphere, and there are sufficient data to provide long-term averages for comparison with models that represent an average atmosphere, such as 2-D models. These data should be useful also for evaluation of 3-D models based on dynamics from general circulation models. Three-dimensional data assimilation models are evaluated with data from particular periods, and the sonde data are being used for this purpose [e.g., *Rood et al.*, 1991].

Ozone builds up in the lowermost stratosphere during winter, a consequence of poleward and downward transport and its long lifetime with respect to chemical loss [e.g., *Holton et al.*, 1995]. I use the sonde data from the Northern Hemisphere to quantify this buildup of ozone in late autumn and winter and its dissipation in late spring and summer. About 10-30% of the ozone column is located between 100 hPa and the tropopause at middle and high latitudes, and this region drives much of the seasonal variation of the ozone column. As much as 15-50% of the decrease in column ozone in the stratosphere at middle to high latitudes takes place between 90 hPa and the tropopause [*Logan*, 1994]. It is important that models be able to reproduce the characteristics of ozone in this region since they are being used to assess perturbations to it.

I begin with a brief review of measurements of the ozone profile in section 2. The major features of the distribution of ozone in the stratosphere are considered to be well known on the basis of measurements of the ozone column from the ground and from space and of the profile from balloons, rockets, and satellites [e.g., *Dutsch*, 1978; *McPeters et al.*, 1984; *London*, 1985; *Perliski and London*, 1989; *Barnes et al.*, 1989; *London and Liu*, 1992; *WMO*, 1995]. I discuss the data used in this study in section 3 and relate the sonde data to the longitudinally varying features of the ozone column using Total Ozone Mapping Spectrometer (TOMS) data [*McPeters and Beach*, 1996] in section 4. Characteristic features of the ozone profiles are discussed in section 5, which includes an analysis of the budget of ozone in the lowermost stratosphere of the Northern Hemisphere. I use mean values for the period 1980-1993 (selected to overlap with the TOMS column measurements) and focus on the utility of these data for model evaluation. A similar analysis of the tropospheric

data is presented in a companion paper [*Logan*, this issue]. Recommendations for the use of particular data are given in section 6, and a summary is given in section 7.

2. A Brief Review of Measurements of the Ozone Profile

The vertical distribution of ozone in the stratosphere was determined in the early 1930s with the Umkehr technique, and the maximum was found to be near 22 km rather than 50 km as thought previously [*Gotz et al.*, 1934; *Walshaw*, 1989]. Balloon-borne ozonesondes provided the first detailed information on the vertical and seasonal distribution of ozone in the 1960s [*Hering*, 1966; *Dutsch*, 1966]. Data from a network of 12 stations that operated from 1963 to 1966 between Greenland and Panama allowed construction of meridional cross sections for ozone in the lower stratosphere and demonstrated their similarity to those for potential vorticity and radioactive debris [*Hering and Borden*, 1964, 1967; *Hering*, 1966]. Ozonesonde measurements using more reliable instrumentation than that employed initially by *Hering and Borden* [1964] were started in Europe, North America, and Japan in the 1960s and early 1970s. Analysis of these data showed in detail the seasonal behavior of ozone at middle latitudes, which shifts from a spring maximum in the lowest part of the stratosphere to a summer maximum by about 20 hPa, above the ozone peak [e.g., *Dutsch*, 1966; *Dutsch et al.*, 1969; *Dutsch and Ling*, 1973; *Dutsch*, 1974; *Attmanspacher and Hartmannsgruber*, 1976; *De Muer*, 1977]. The first decade of these data, with limited spatial coverage, was used to derive a climatology for stratospheric ozone [*Dutsch*, 1978], and the sonde data have been reviewed subsequently by *London* [1985] and *London and Liu* [1992].

The late 1970s saw the advent of satellite measurements of ozone in the middle and lower stratosphere. These instruments offer the potential of almost global coverage. *London* [1985] reviews earlier satellite data primarily for the upper stratosphere. Measurements from solar backscattered ultraviolet (SBUV) instruments (which rely on a nadir viewing technique) give vertical profiles of ozone for 80°N to 80°S from 100 to 1 hPa, with resolution of ~ 8 km above 10 hPa and about 15 km below [*McPeters et al.*, 1984, 1994; *Perliski and London*, 1989]. Stratospheric Aerosol and Gas Experiment (SAGE) I and II, solar occultation instruments, provide profiles of ozone with 1-km resolution from 70 km down to the middle troposphere, or cloud top [*McCormick et al.*, 1989]. In practice, the ozone profiles from SAGE II (Version 5.96) appear to be reliable to ±10% or better down to about 15 km for middle and high latitudes, but differences between sonde measurements and colocated SAGE II data are as large as ~30% (with SAGE II larger) at 15 km for lower latitudes, where ozone concentrations are smaller [*WMO*, 1998]. SAGE II provides year-round measurements for ozone from 50°N to 50°S. More recently, ozone has been measured by several instruments on the Upper Atmosphere Research Satellite (UARS), but none of these gives reliable data below 100 hPa [*Bailey et al.*, 1996; *Bruhl et al.*, 1996; *Cunnold et al.*, 1996; *Froidevaux et al.*, 1996; *WMO*, 1998]. Polar Ozone and Aerosol Measurement (POAM), also a solar occultation instrument, gives ozone measurements from 55° to 71° and 18 to 35 km, but different latitudes are sampled in different months [*Randall et al.*, 1995].

3. Analysis of Ozonesonde Measurements

Data from the ozonesonde stations in Table 1 were considered in this analysis. The measurements were made with two

Table 1. Ozonesonde Data

WMO Code	Station	Latitude	Longitude	Type	Number	Data Record
18	Alert	82°N	62°W	ECC	347	1/88-12/93
89	Ny Alesund	79°N	12°E	ECC	231	10/90-12/93
24	Resolute	75°N	95°W	ECC	540	1/80-12/93
262	Sodankyla	67°N	27°E	ECC	246	1/89-12/92
77	Churchill	59°N	147°W	ECC	515	1/80-12/93
21	Edmonton	53°N	114°W	ECC	498	1/80-12/93
76	Goose Bay	53°N	60°W	ECC	546	9/80-12/93
221	Legionowo ^a	53°N	21°E	GDR ^a	234	1/80-12/93
174	Lindenberg ^a	52°N	14°E	GDR ^a	845	1/80-12/93
99	Hohenpeissenberg	48°N	11°E	BM	1615	1/80-12/93
156	Payerne	47°N	7°E	BM	1520	1/80-12/93
132	Sofia	43°N	23°E	GDR	195	2/82-12/91
12	Sapporo	43°N	141°E	KC	250	1/80-12/95
67	Boulder ^b	40°N	105°W	ECC	333	1/85-12/93
107	Wallops Is. ^b	38°N	76°W	ECC	401	1/80-12/93
14	Tateno	36°N	140°E	KC	444	1/80-12/95
7	Kagoshima	32°N	131°E	KC	227	1/80-12/95
190	Naha	26°N	128°E	KC	185	9/89-12/95
109	Hilo ^b	20°N	155°W	ECC	359	12/84-12/90
219	Natal ^c	6°S	35°W	ECC	281	11/78-10/92
191	Samoa ^b	14°S	170°W	ECC	164	4/86-1/96
26	Aspendale/Laverton	38°S	145°E	BM	285	1/80-12/95
256	Lauder ^d	45°S	170°E	ECC	286	8/86-12/90
233	Marambio	64°S	57°W	ECC	241	11/88-12/95
101	Syowa	69°S	39°E	KC	303	1/86-12/95
280	Forster	71°S	12°E	GDR	339	5/85-2/91

WMO, World Meteorological Organization; ECC, electrochemical concentration cell; BM, Brewer Mast; KC, type of sonde made in Japan; GDR, BM type of sonde made in the former East Germany. The data were obtained from the World Ozone and Ultraviolet Data Center, with the exceptions noted below. Column 8 shows which data were included in the analysis; 1980-1993 was chosen as the period if such data were available. Column 7 gives the numbers of soundings that met the selection factor criteria: 0.8-1.2 for ECC and KC sondes and 0.9-1.35 for the Brewer Mast type of sondes (BM, GDR sondes), except for Hohenpeissenberg, for which 0.9-1.2 was used. The data for Samoa, Natal, and Lauder are not normalized to the ozone column.

^aLegionowo changed to ECC sondes in June 1993; Lindenberg changed to ECC sondes in June 1992.

^bData provided by S. Oltmans. The Wallops Island data from WOUDC were reprocessed, so each profile is scaled to reevaluated ozone column data on the Bass-Paur scale [Oltmans *et al.*, 1998].

^cData provided by V. Kirchhoff.

^dData provided by G. Bodeker.

types of sonde: the electrochemical concentration cell (ECC) and variations on the Brewer Mast (BM) bubbler, as discussed by Logan [this issue and references therein]. Most of the sonde data used in this study have been normalized to concurrent measurements of the ozone column, with exceptions noted in Table 1. The magnitude of the scaling, or correction factor, is used as a quality check [Logan, this issue].

Several intercomparisons have shown that BM sondes record more ozone than do ECC sondes in the stratosphere, but usually by less than 5% [Atmanspacher and Dutsch, 1970, 1981; Hilsenrath *et al.*, 1986; Beekman *et al.*, 1994]. The accuracy of ECC sondes is estimated to be about $\pm 5\%$ in the stratosphere, and the precision is estimated to be $\pm 3\%$; values for BM sondes are slightly larger [Komhyr *et al.*, 1995; Beekman *et al.*, 1994; WMO, 1998]. Several of the sondes made in the former East Germany (type GDR) performed unreliably in the 1970 and 1978 intercomparisons, but when soundings with large correction factors were omitted from the analysis, the agreement with the BM and ECC sondes was reasonable in the stratosphere [Atmanspacher and Dutsch, 1970, 1981]. The GDR data were included in this analysis, but 30-50% of the soundings were excluded on the basis of the correction factor criteria, a higher fraction than for BM or ECC soundings [Logan, 1994]. The GDR sondes were replaced with ECC sondes in the early 1990s.

There has been a significant trend in ozone in the lower stratosphere over the past two decades, with a maximum decrease of

7% per decade for 90-200 hPa for 1970-1996 at northern midlatitudes [Logan *et al.*, 1999]. Ozone values in the lower stratosphere were particularly low in late 1992 and 1993 because of the enhanced aerosol loading in the stratosphere following the eruption of Mt. Pinatubo [e.g. Kerr *et al.*, 1993; Hofmann *et al.*, 1994; Solomon *et al.*, 1998]. I selected 1980-1993 as the period for which to provide long-term averages of ozone in large part because it coincides with the period of TOMS measurements. Averages for 1980 to 1995 are provided for the Japanese and Australian stations, where a longer record is needed to improve statistics for the summer months. Results are shown also for stations which operated for shorter periods to describe the morphology of ozone, but I do not recommend using all of these stations for model evaluation as discussed in section 6.

Ozone concentrations for each sounding that met the correction factor criteria in Table 1 were interpolated to give values at 22 pressure levels from 1000 to 10 hPa. Tables were compiled for each station, giving the sample monthly mean, the standard deviation s (the square root of the sample variance), and the number of soundings (N) that went into the mean; the standard error s/\sqrt{N} is readily calculated. The means weight each sounding in a month equally. There were insufficient measurements made each month at most stations (usually 3-4) to derive statistics that reflect interannual variability [Logan, this issue]. For most stations the measurement frequency did not change between 1980 and 1993. The measurement frequency increased around 1990

for the Japanese stations, particularly Sapporo, so that the means are weighted toward the end of the given time period, especially for the summer data; this is unavoidable, given the sparseness of data in the previous years [Logan, 1994]. Thermal tropopause heights for each sounding were determined using the standard meteorological definition [Craig, 1965], and monthly medians are shown by Logan [this issue]. Monthly statistics for all stations are available from <http://www-as.harvard.edu>.

The 95% confidence intervals for the monthly means (2 SE) depend on both the inherent variability of ozone and the number of measurements. There are sufficient measurements each month for midlatitude and tropical stations that 2 SE are less than 10% of the monthly means for 50-20 hPa, as shown in Figures 1 and 2. Errors are only slightly larger for 60-70 hPa for middle and high latitudes, but the errors increase for 125-80 hPa; here 2 SE are <15% of the monthly means for 48°-82°N and are usually <20% of the monthly means for 36°-43°N for $N>20$ (Figure 1). Near the tropopause, 2 SE are usually <25% of the monthly mean for high latitudes and <30% for middle latitudes for $N>20$ [Logan, this issue]. In the tropics, 2 SE are <5% of the monthly means for 70-90 hPa for $N>20$, while the variability is larger in the subtropics, where 2 SE are 10-25% of the monthly means. The means are less well defined in the lower stratosphere in the tropics and subtropics, where $N<20$ for several months at most stations.

The TOMS data, version 7.0 [McPeters and Labow, 1996; McPeters and Beach, 1996], were used to evaluate (1) the spatial bias in the location of the sonde stations with respect to the zonal

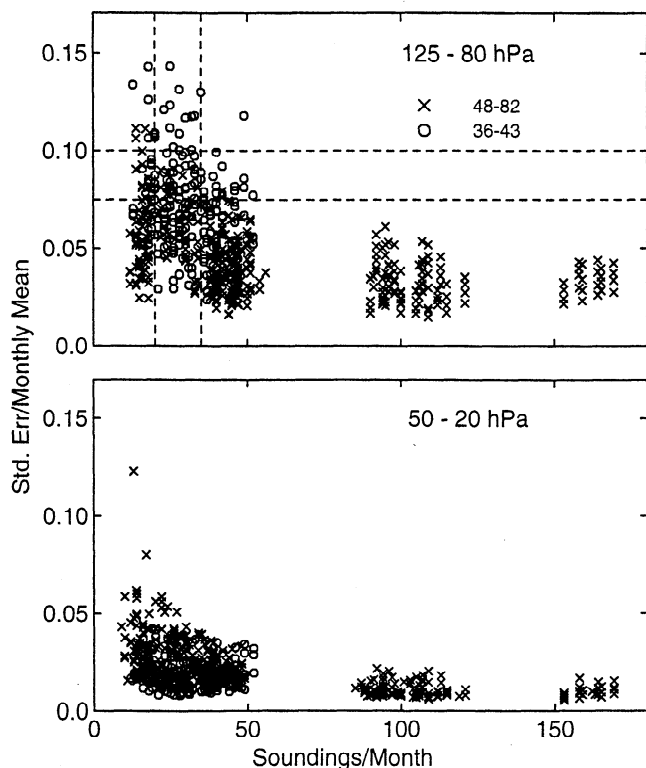


Figure 1. The ratio of the standard error to the monthly mean versus the number of soundings in a month. The vertical dashed lines in the top plot are for $N=20$ and $N=35$; the horizontal dashed lines are for relative standard errors of 0.075 and 0.1. Results are shown for selected stations located between 48° and 82°N (crosses) and 36° and 43°N (circles). Hohenpeissenberg, Payenne, and Lindenberg (December-April) have more than 60 soundings per month.

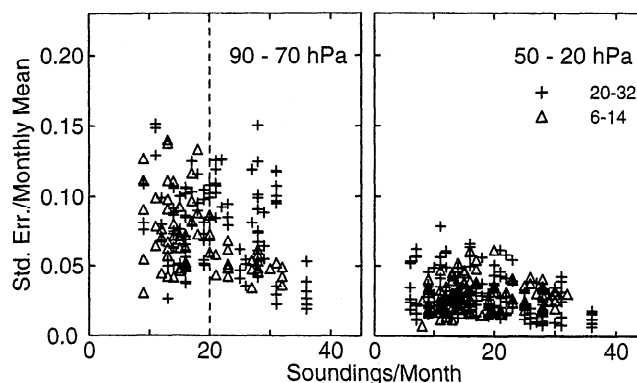


Figure 2. The ratio of the standard error to the monthly mean versus the number of soundings in a month. Results are shown for Kagoshima, Naha, and Hilo (pluses) and Natal and Samoa (triangles).

mean value of ozone and (2) the sampling and instrumental bias caused by the relative infrequency of the sonde measurements compared to the satellite measurements, and any offset between the satellite versus the ground-based measurements of the ozone column. For present purposes the biases in 2 appear to be small, as shown below.

4. Sonde Data for Ozone in the Context of the TOMS Data

Plate 1 shows the mean ozone column in January and July for 1980 to 1993 and the locations of the sonde stations. Because of the longitudinal gradient in ozone, which is strongest in winter, certain stations are in regions that are biased low with respect to the zonal mean value of ozone (e.g., the European stations in winter), while others are biased high (e.g., some Canadian stations and Sapporo, Japan). These biases were quantified by taking the ratio of ozone values in a box (3° latitude by 3.75° longitude) around each station to the appropriate 3° zonal mean value (Figure 3). The systematic biases with respect to the zonal mean are less than ~5% with some exceptions: the European sites are biased low by ~10% in November-February, but there is little bias from April to August; Sapporo is biased high by up to 15% in winter and by more than 5% all year except July to October; Resolute is biased high by a little more than 5% in March, but the TOMS instrument does not see the high latitudes in winter; Syowa, influenced by the Antarctic ozone hole [Chubachi, 1997], is biased low in October and November.

Most sonde data are normalized with respect to a ground-based measurement of the ozone column. Figure 4 shows the ratio of the ground-based data for the ozone column, obtained on the same day as each sounding, and the TOMS data for the 3° by 3.75° box around each station. This comparison shows that the biases in the ozone column are usually <5%. TOMS overpasses tend to give ozone columns that are 1% higher than collocated Dobson stations [McPeters and Labow, 1996]. Some of the systematic offsets are caused by different time periods for the data and by the downward trend in column ozone. For example, the ozone column recorded at Syowa for the period 1986-1993 is ~10% lower than the TOMS data in August-December for 1980-1993. There was a substantial decline in Antarctic ozone through the 1980s [WMO, 1995], so the sonde data are biased low. The column for Sapporo is systematically low, especially in summer, consistent with the fact that the mean for the sonde data is biased

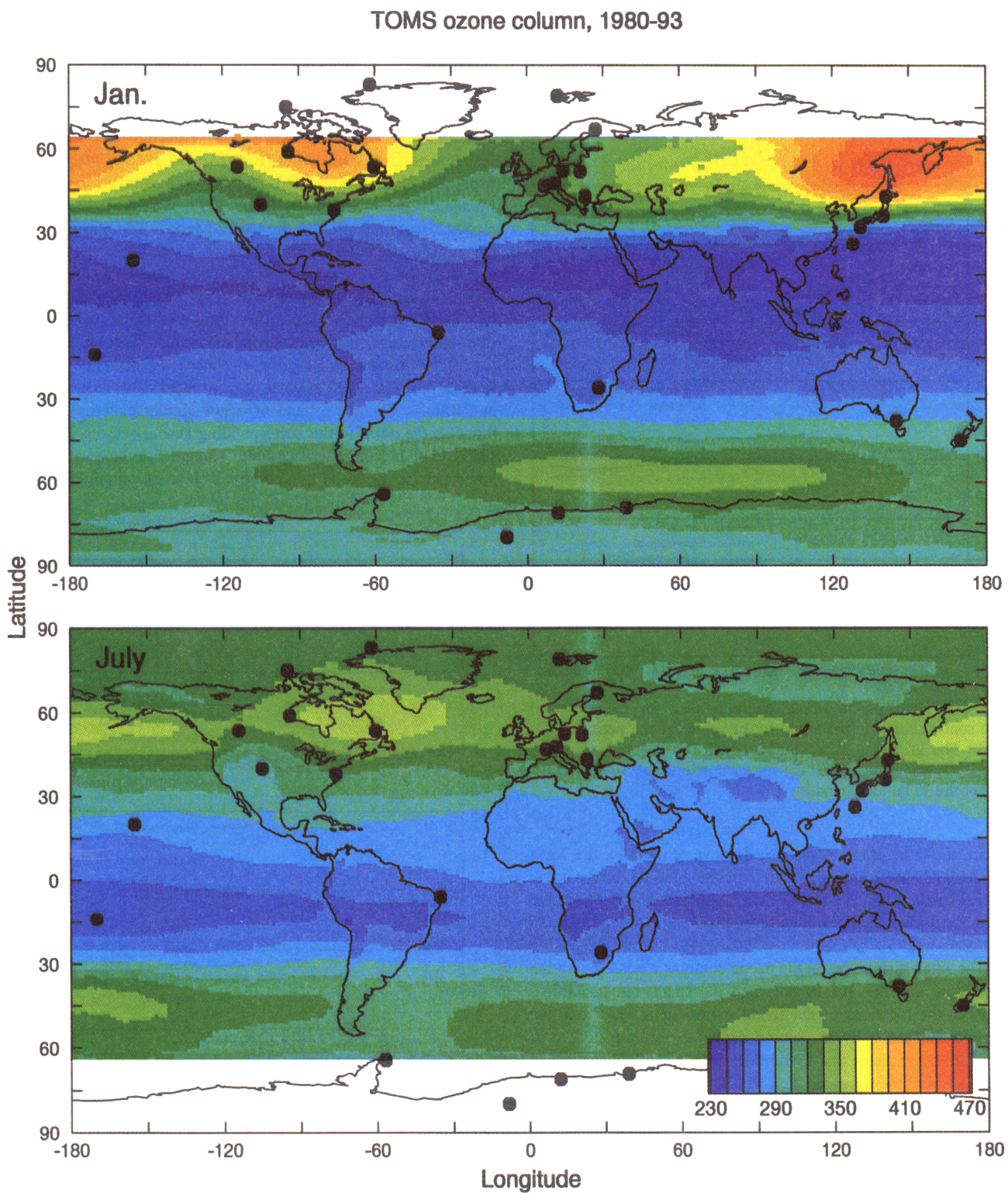


Plate 1. Ozone columns measured by the Total Ozone Mapping Spectrometer (TOMS) for 1980 to 1993, in January and July, with the locations of ozonesonde stations marked (circles). The data were taken from the gridded monthly mean files (1° latitude by 1.25° longitude) on the TOMS CD-ROM (version 7) [McPeters and Beach, 1996].

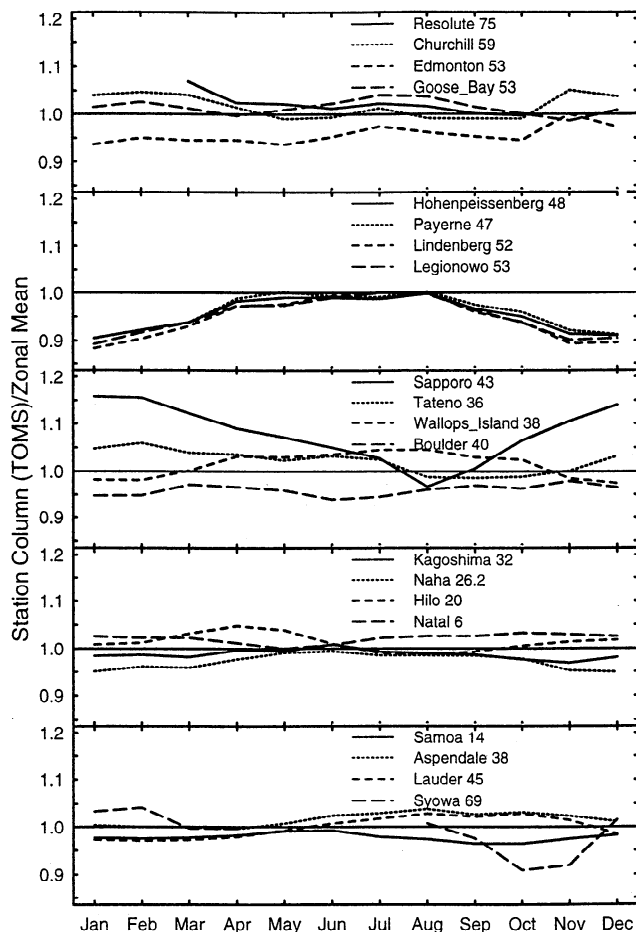


Figure 3. Ratio of the TOMS ozone column for a grid around each sonde station to the zonal mean value for the period 1980-1993. The grid was 3° in latitude by 3.75° in longitude. The ratio is for the 14-year average for each grid to the 14-year average for the 3° latitude band.

toward the later years, as discussed above. Similarly, the Naha data for 1989-1995 and Hilo data for 1985-1993 tend to be a few percent smaller than the TOMS data; there is less trend in ozone in the subtropics than at midlatitudes [McPeters *et al.*, 1996], so the bias is small.

The small differences in the ozone columns recorded with the sonde data and the TOMS data and the small, but quantified, biases in the locations of the sonde stations with respect to the longitudinal variability in column ozone at northern middle to high latitudes indicate that such biases do not preclude the use of the sonde profiles for evaluation of 2-D models. The biases are significantly smaller than the differences among model values for ozone in the lower stratosphere, as discussed in section 1.

5. Analysis of the Sonde Data

5.1. Vertical Profiles and the Annual Cycle of Ozone

Vertical profiles of ozone below 10 hPa are shown in Figure 5 while the annual cycle of ozone is shown for selected stations and pressure levels in Figure 6. Ozone is given in units of partial pressure, so the integral of these profiles gives the ozone column. Figures 5 and 6 emphasize well-known features of the ozone distribution, such as the gradient with latitude, and the increase in

altitude of the maximum in the ozone profile from high to low latitudes.

One of the most characteristic features of the vertical profiles from 75°N to 36°N is the inflection, or secondary maximum, that builds up between the tropopause and 100 hPa over winter (Figures 5a-5c). This feature, reflecting the poleward and downward transport of ozone into the lowermost stratosphere in the winter hemisphere, is discussed extensively in papers interpreting the first few years of sonde data [e.g., Dutsch, 1966; Dutsch *et al.*, 1969; Dutsch and Ling, 1973; Dutsch, 1974; Attmanspacher and Hartmannsgruber, 1976; De Muer, 1977; Pittock, 1977] but has received scant attention in the past 20 years. The phase shift in the annual cycle at midlatitudes (Figures 6b and 6c) from a spring maximum in the lowermost stratosphere, where the lifetime of ozone is long and controlled largely by dynamics, to a summer maximum by 10 hPa, where the lifetime of ozone is relatively short and controlled by photochemistry, is also discussed in these papers and in subsequent reviews [Dutsch, 1978; London, 1985; London and Liu, 1992]. Much more limited data from southern midlatitudes (Figures 5d and 6f) show similar patterns to those found in the north in the corresponding austral seasons [Pittock, 1977]. For high northern latitudes (Figure 6a) the spring maximum persists up to 20 hPa and by 10 hPa the amplitude of the

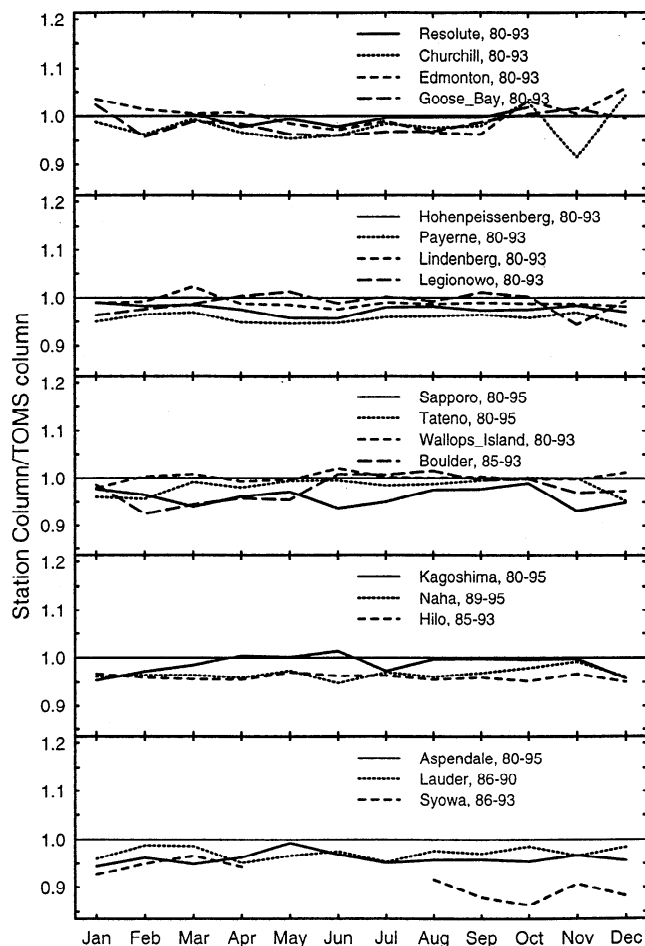


Figure 4. Ratio of ozone columns provided with each ozone sounding to the ozone columns measured by TOMS for a grid 3° in latitude by 3.75° in longitude centered on each station. The ratio was formed from the average for the years given in the legend for each sonde station and from the average for 1980-1993 for the TOMS data.

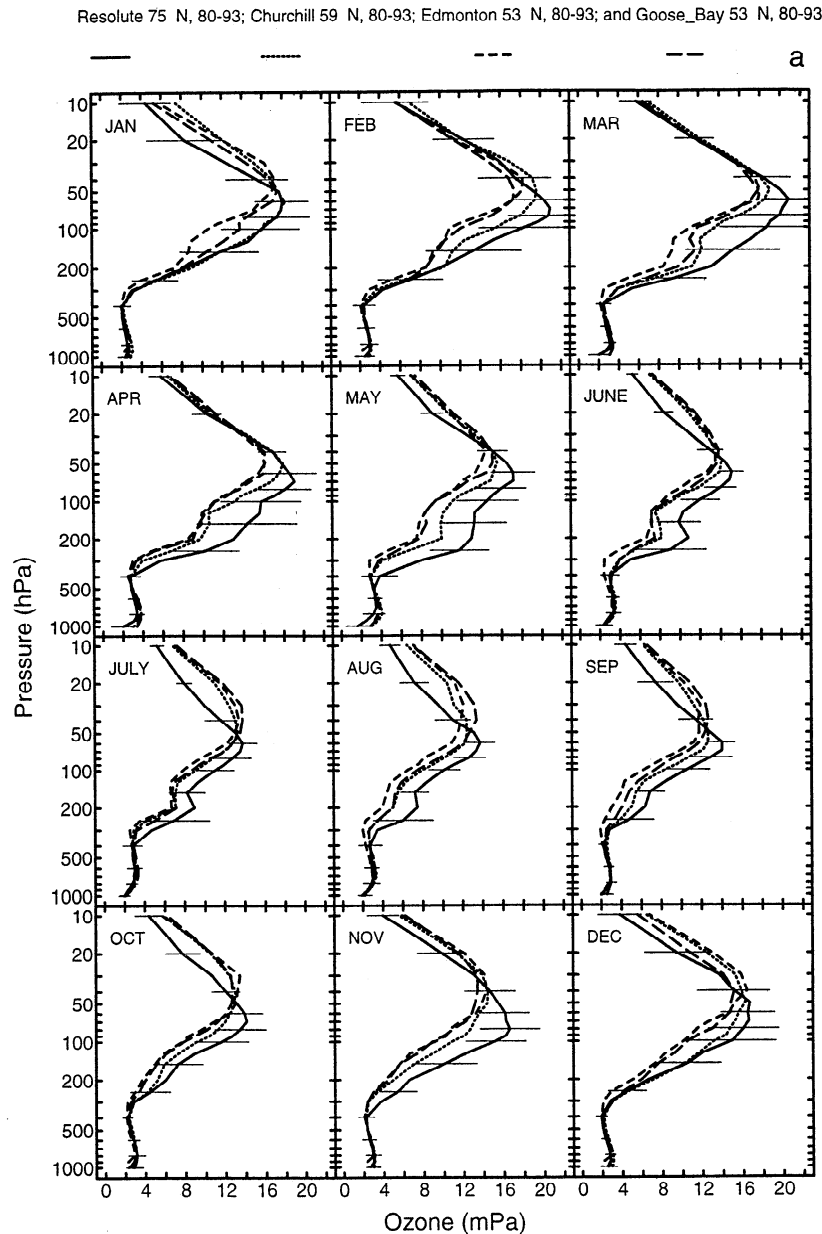


Figure 5. Monthly mean vertical profiles for ozone, for selected sonde stations. The means are averages of each individual sounding that met the appropriate correction factor criteria [Logan, this issue] for the period given in the legend. The horizontal lines show the mean ± 1 standard deviation for selected stations and levels. The data are arranged by latitude.

cycle is small, but the maximum remains in the early part of the year. Recent measurements by the POAM instrument indicate a summer maximum in ozone by 35 km (~ 6 hPa) for latitudes 65° – 70° N [Randall *et al.*, 1995].

The data from the two tropical stations, Natal (6° S) and Samoa (14° S) (Figure 6e), show that the annual cycle has a winter or spring maximum at 90 hPa, near the tropopause, and a slight winter maximum in the stratosphere below 20 hPa. At 10 hPa, there are maxima in late summer and late winter at Natal but only in late summer at Samoa. The amplitude of the annual cycle in stratospheric ozone is small, as shown in Figure 7 for the column from 100 to 10 hPa. The maximum in the upward air

mass flux across the 100-hPa surface in the tropics is in northern winter [Holton, 1990; Rosenlof and Holton, 1993]. Rosenlof [1995] suggested that this could be responsible for the minimum values of tropical stratospheric ozone in northern winter since stronger upwelling would transport lower ozone air from the troposphere into the stratosphere. There is a slight minimum in the stratospheric ozone column in January and February at Natal. There is no evidence in the sonde data that this minimum propagates up from the lower stratosphere (Figure 6e), and it seems more likely that the lower values in northern winter near 20 hPa are caused by enhanced transport from the tropics to midlatitudes in northern winter. The annual cycle of the total column of ozone

Hohenpeissenberg 48 N, 80-93; Sapporo 43 N, 80-95; Boulder 40 N, 85-93; and Wallops_Island 38 N, 80-93

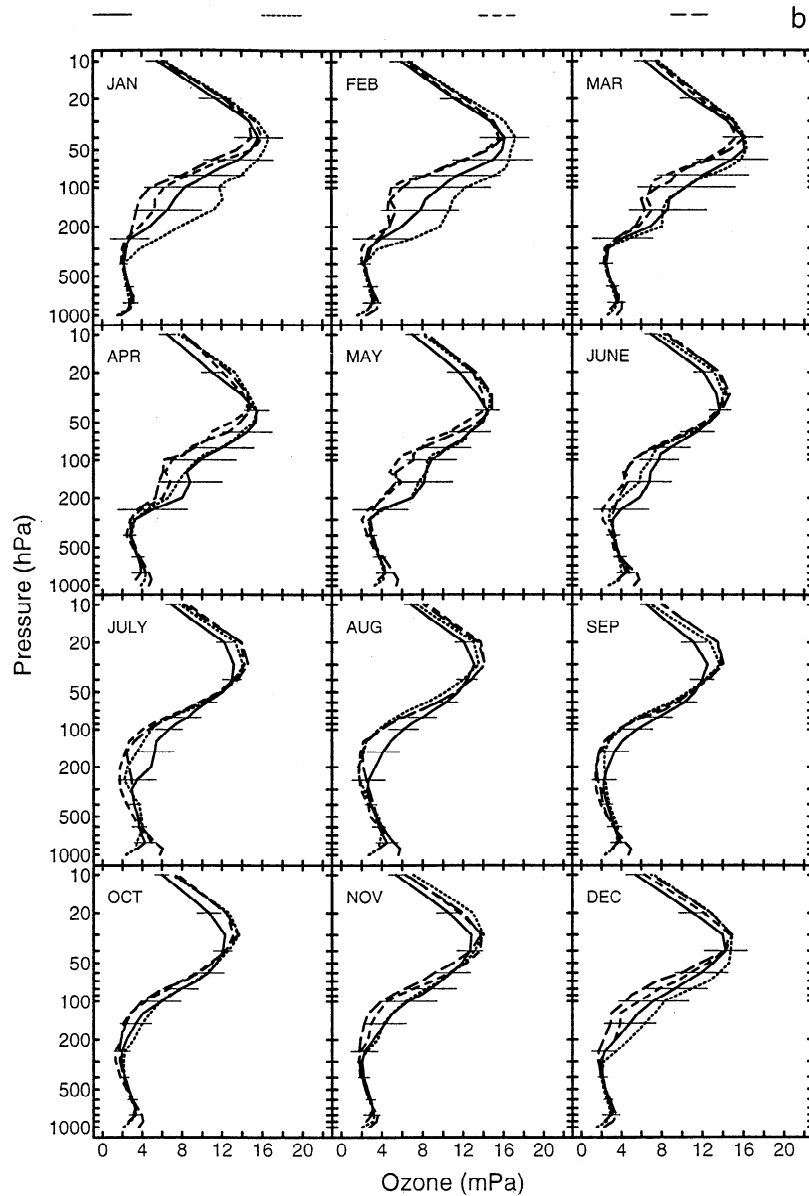


Figure 5. (continued)

over Natal is driven in large part by the strong annual cycle in tropospheric ozone [Logan and Kirchoff, 1986; Kirchoff et al., 1991].

Year-round measurements are available for three stations on the edge of the Antarctic continent since the discovery of the ozone hole but for only one, Syowa, before (Figures 5e and 6g). The annual cycle of ozone before the ozone hole is different from that at northern high latitudes [Dutsch, 1974, 1978]. Highest values of ozone were found in late winter and in early summer at 100 hPa with shallow minima in between, from 1966 to 1974; by contrast, data from the South Pole showed a summer maximum and spring minimum before the ozone hole [Oltmans et al., 1994]. The annual cycle at Syowa was small at 50 hPa with a slight peak in late spring, and by 20 hPa there was a pronounced maximum in late spring and a minimum in autumn. There was only a hint of a secondary maximum in ozone at Syowa in the lowermost stratosphere, and it occurred from December to April, in late summer, rather than in winter as found in the north (Figure

5e). At southern midlatitudes, however, the secondary maximum is in the same season as in the north (Figure 5c). Since the onset of the ozone hole, highest values of ozone are found in autumn and winter at 100 hPa but are found in summer at 20 hPa (Figure 6g). The mean profiles for the three stations, Marambio, Forster, and Syowa, are similar, showing dramatic ozone losses compared to the preozone hole profile for Syowa in September-November [cf. Logan, 1994; Oltmans et al., 1994].

5.2. Variability in Ozone in the Lowermost Stratosphere

Individual profiles of ozone from middle and high latitudes frequently show a layered structure in the lowermost stratosphere in winter and spring [e.g., Dutsch et al., 1969; Dobson, 1973]. Reid and Vaughan [1991] found that the layers in the sonde data are most common from 200 to 75 hPa, with a peak altitude of 14 km (~140 hPa), and occur from the tropopause to ~50 hPa. Their frequency increases from late fall to spring, then decreases to almost none in summer; it also decreases from higher to lower

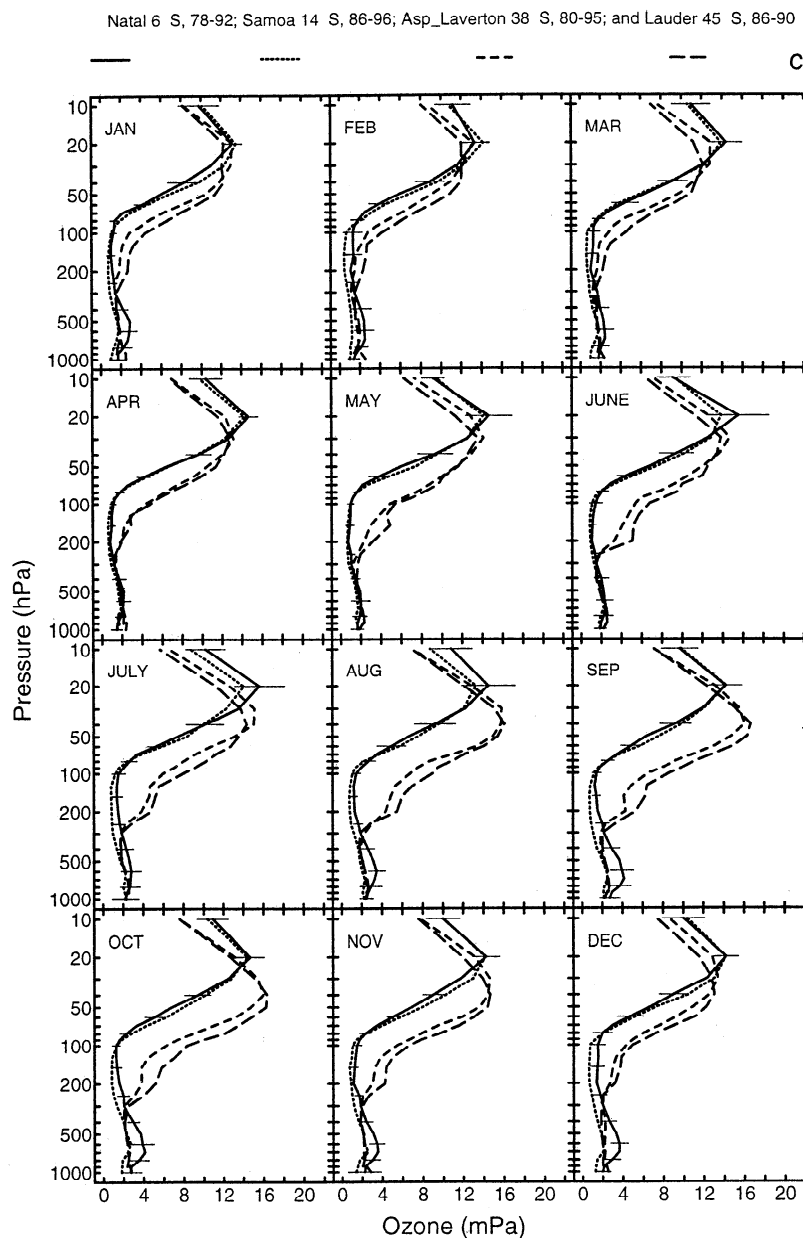


Figure 5. (continued)

latitudes, and they are not found below 30° . They are deeper and narrower at higher than at middle latitudes. Reid and Vaughan suggested that the layers are caused by exchange of air across the boundary of the polar vortex. Reid *et al.* [1993] found that layers were absent or rare in the north polar vortex. Using lidar data, they showed the extrusion of layers of high ozone from the polar regions in the lowermost stratosphere. Newman *et al.* [1996] found that high ozone layers at midlatitudes were correlated with low N_2O , indicating a polar origin, which was confirmed by tracing the air motion back to the polar vortex. They concluded that the layers were a consequence of Rossby wave breaking during the breakdown of the polar vortex.

Appenzeller and Holton [1997] developed a climatology for the ozone lamination rate using Microwave Limb Sounder (MLS) data and also derived the lamination rate for modified potential vorticity. They showed that the formation of layers is a consequence of dynamical activity providing vertical shear, in combi-

nation with strong horizontal ozone gradients on which quasi-horizontal advection can act. They found similar features in the lamination rate to those derived from the sonde data and also found high lamination rates in the subtropical lower stratosphere in winter and in the lowermost stratosphere year round, with a maximum in winter near 30° - 40° , near the subtropical tropopause break. Appenzeller and Holton argue that the earlier studies discriminated against layers where ozone partial pressures are low, such as the subtropical lower stratosphere, as their definition of a layer was based on an absolute rather than a relative increment in ozone.

The layers appear to be an important contributor to the variability in ozone below 50 hPa. For example, in the lowermost stratosphere the variability in ozone, indicated by the horizontal lines in Figure 3 (± 1 standard deviation), is largest in winter and spring. However, the relative variability, the ratio of the standard deviation to the monthly mean, is largest near the tropopause

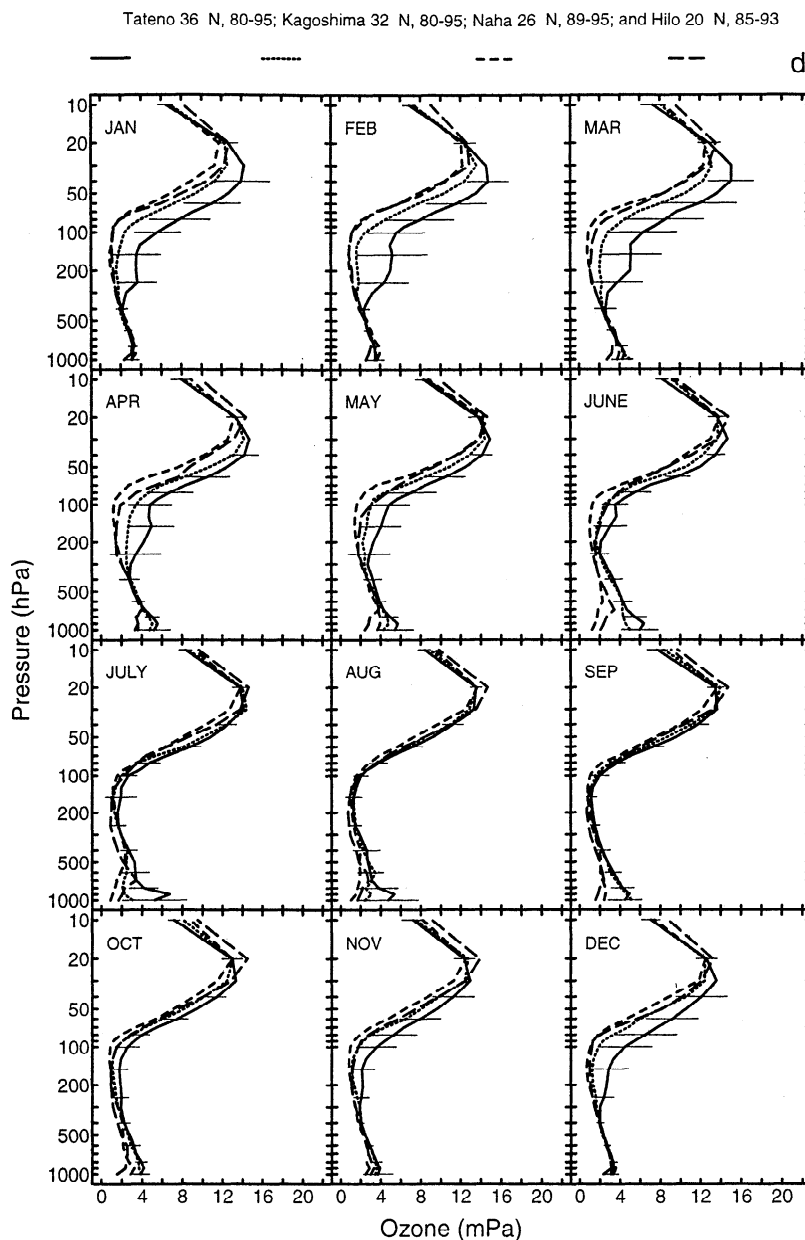


Figure 5. (continued)

[Logan, this issue]. The layers identified by Reid and coworkers occur primarily in the region of the secondary ozone maximum shown in Figures 5a and 5b. The phase of the frequency of lamination [Reid and Vaughan, 1991] is about the same as that of the ozone content of the lowermost stratosphere shown below in Figure 11.

Figure 1 shows that the relative standard errors of ozone in the lowermost stratosphere are larger for stations near 40° than those northward of 50°N (for similar N), consistent with higher lamination rates inferred from modified potential vorticity. Similarly, Figure 2 shows that the relative standard errors in the lower subtropical stratosphere (70-90 hPa) are larger than those in the tropical stratosphere (and those near 40°N), consistent with the higher lamination rates shown by Appenzeller and Holton [1997] for the lower subtropical stratosphere.

5.3. Contribution of Different Ozone Layers to the Ozone Column

The sonde data provide a unique opportunity to examine the contribution of different altitudes to the behavior of the total ozone column. We divided the profiles into (1) the troposphere, using the standard definition of the thermal tropopause [Craig, 1965]; (2) the tropopause to 100 hPa (roughly, the region of the secondary maximum, and at midlatitudes, below the 380-K surface favored as the division between the lowermost stratosphere and the "overworld" [Holton *et al.*, 1995]); (3) 100-50 hPa (roughly, up to the ozone maximum at midlatitudes); and (4) 50-10 hPa, to the top of the region with good statistics for measurements for many stations. The ozone columns for these four regions are compared to the total ozone column in Figure 8,

Syowa 69 S, 86-95; Syowa 69 S, 66-74; Marambio 64 S, 88-95; and Forster 70 S, 85-91

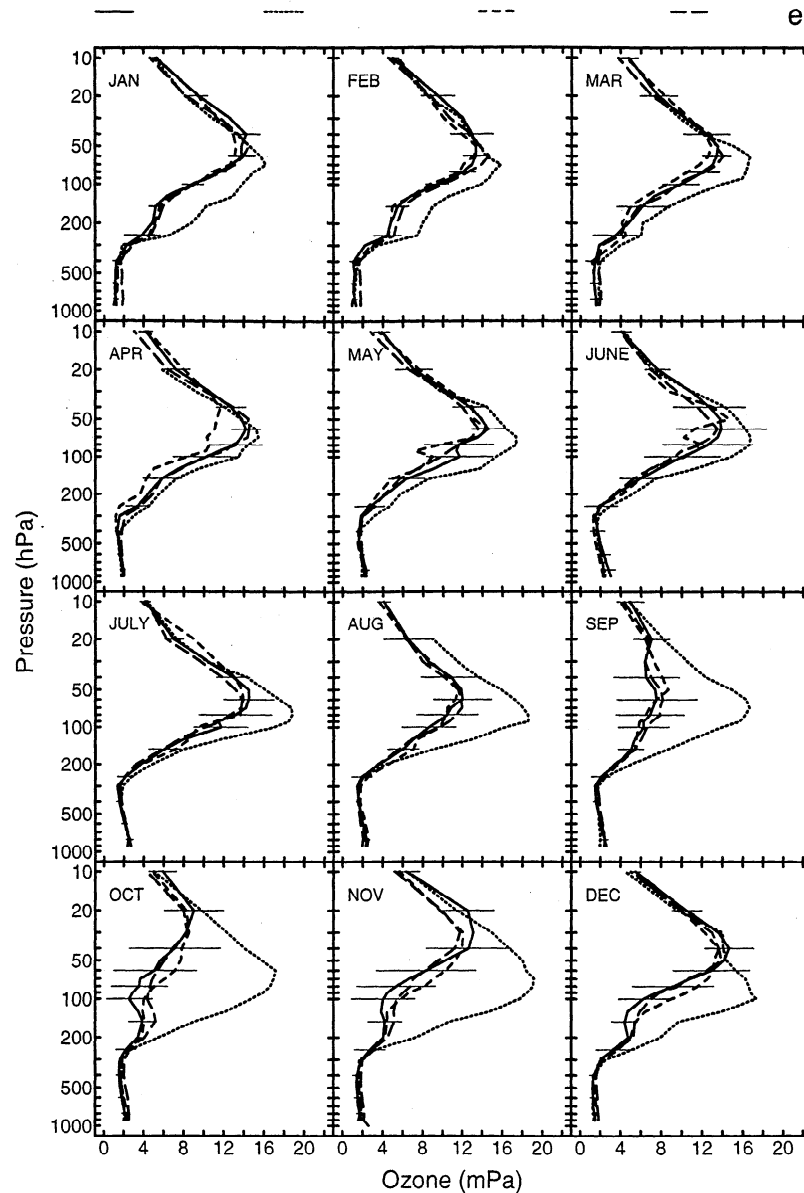


Figure 5. (continued)

which illustrates the well-known fact that the seasonal cycle of the column is due primarily to the evolution of ozone in the lower stratosphere. The sonde data show that for latitudes 75° - 35° , 35%-50% of the increase in the total column from October to March is caused by the increase in the amount of ozone between the tropopause and 100 hPa; for the high-latitude stations the region from 50 to 10 hPa contributes 35-40% of the increase in the column during these months, while for midlatitude stations it contributes 15-25%, reflecting the move to a summer maximum in ozone at the higher altitudes. The amplitude of the seasonal cycle is smaller above the ozone maximum than below, as well as being shifted in phase.

The region between the tropopause and 100 hPa contains 15-27% of the ozone column poleward of 45° N, with the largest fraction at higher latitudes and in spring, as shown in Figure 9. Even though this region contributes only 10-15% of the total

column around 40° N it contributes about 50% of the amplitude of the annual variation of the column. By about 30° N the region below 100 hPa is almost entirely in the troposphere.

It is usually stated that the troposphere contains about 10% of the ozone column. The contribution of the troposphere can be as low as 5% in winter and as high as 15% at more polluted midlatitude locations in summer, as shown in Figure 10. The fraction is close to 20% in regions of the tropics where tropospheric ozone is influenced by emissions from biomass burning and where the stratospheric column is low [Logan and Kirchoff, 1986]. The amount of ozone from the ground to 100 hPa is as much as 20-33% of the column at middle and high latitudes.

5.4. Ozone in the Lowermost Stratosphere

The annual cycle of ozone from the tropopause to 100 hPa is shown in Figure 11, with the stations grouped by latitude; Figure

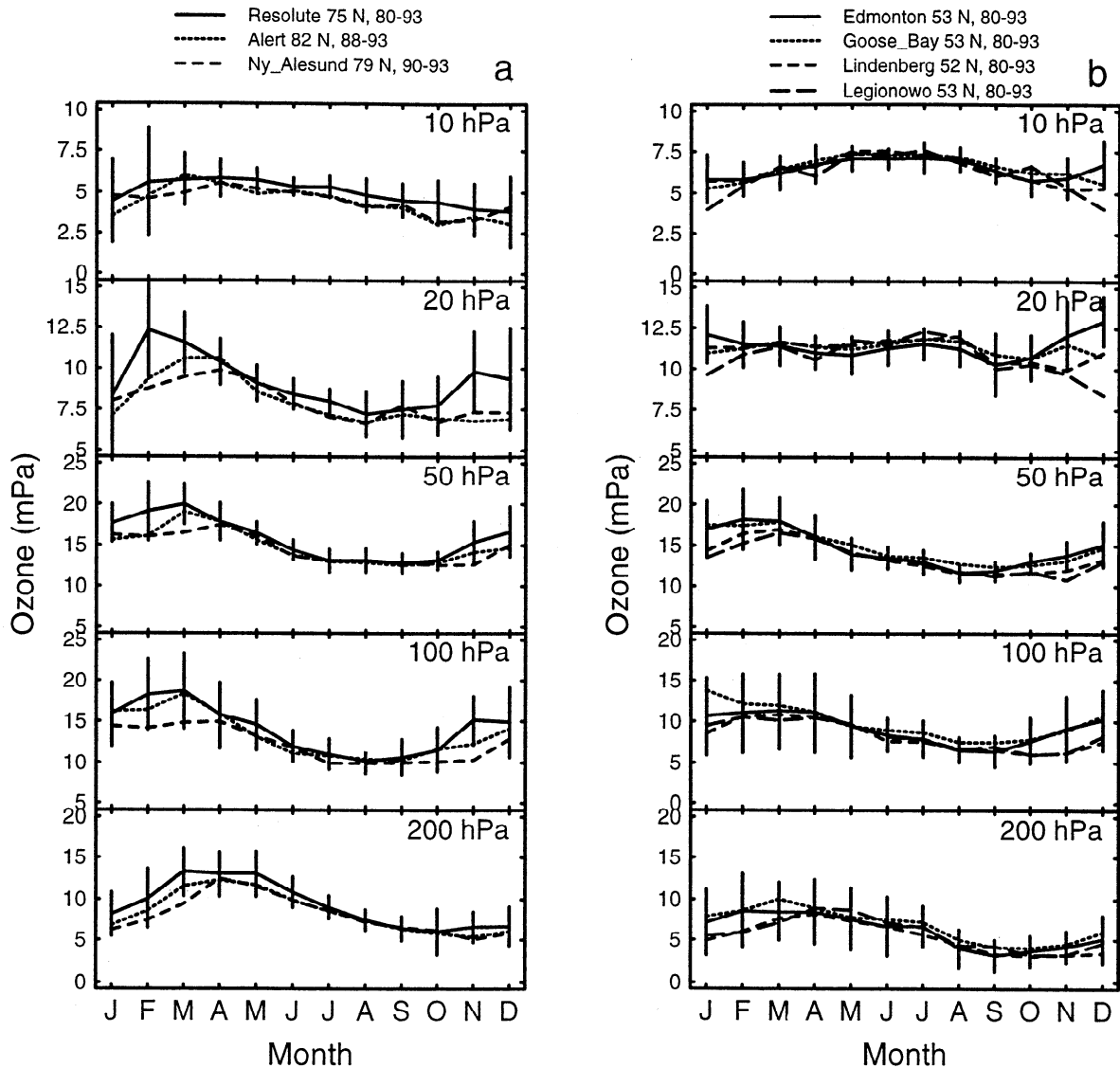


Figure 6. Annual cycle of ozone for selected pressure levels and stations. The vertical bars show the mean ± 1 standard deviation for one station on each plot. The data are arranged by latitude.

12 shows the corresponding monthly change in the amount of ozone. For a given latitude belt the rate of change of ozone is similar, with the exception of Sapporo, even though the stations are located in different positions with respect to the longitudinal variation of column ozone. (Sapporo is affected by a strong north-south mean trough at 50-100 hPa in winter, resulting in northwesterly flow from eastern Siberia and mean subsidence, as discussed by *Dutsch* [1974] and *London* [1985]). The stations near 40°N start to accumulate ozone from September to February or March and lose ozone at the fastest rate from May to July; those near 50°N accumulate ozone for the same period and lose it at the fastest rate from June to August; those near 80°N accumulate ozone from October or November to March or April and lose it at the fastest rate from May to June. There is a lag of about a month in the time when ozone starts to accumulate in the lowermost stratosphere, from ~40°N to high latitudes, and a similar lag in the time when the ozone content starts to decrease.

The data shown in Figure 12 could be used, in principle, in combination with estimates of the flux of ozone across the 100-

hPa surface to compute the cross-tropopause flux of ozone, following the approach of *Appenzeller et al.* [1996]. They calculated the mass flux of air across the tropopause as the sum of mass flux into the lowermost stratosphere (defined by the 380-K surface) and the rate of change of the mass of the lowermost stratosphere, using UARS data and daily stratospheric analyses prepared for the UARS project. For ozone the net effect of chemistry would also have to be included to estimate the net flux of ozone across the tropopause. The sonde data are the only available measurements that can provide the rate of change of the ozone content in the lowermost stratosphere, and I present an estimate for the Northern Hemisphere below. Accurate estimates of the mass flux of ozone across the 380-K or 100-hPa surface are limited at present by the lack of reliable global ozone data at 100 hPa during 1992-1993, the period for which several authors calculated the global-scale meridional circulation using UARS data [*Appenzeller et al.*, 1996; *Yang and Tung*, 1996; *Eluszkiewicz et al.*, 1996] and by the uncertainties in these calculations. The UARS instruments do not give reliable ozone measurements as

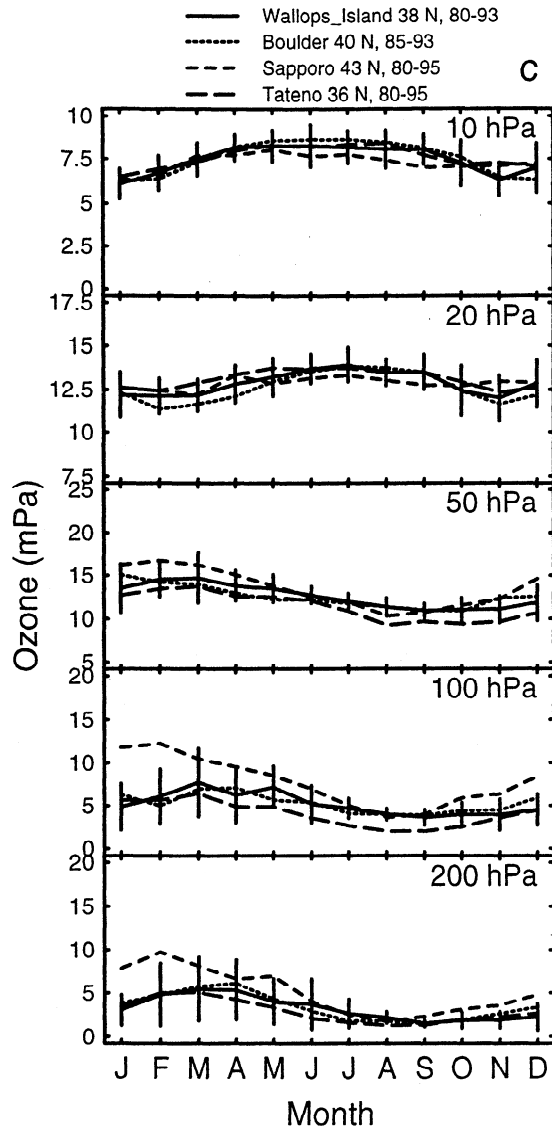


Figure 6. (continued)

low as 100 hPa [Bailey *et al.*, 1996; Bruhl *et al.*, 1996; Cunnold *et al.*, 1996; Froidevaux *et al.*, 1996], and the SAGE II ozone data are confounded by aerosols from Pinatubo at the lower altitudes for 1991-1993 [WMO, 1998].

For air the maximum in the downward flux across the 380-K surface is in midwinter, while the maximum in the net flux of air across the tropopause caused by changes in mass of the lowermost stratosphere is in April-June; the net flux of air across the extratropical tropopause is highest in January-July, with a peak in May-June, caused primarily by the flux out of the lowermost stratosphere. Gettelman *et al.* [1997] calculated the mass flux of ozone across the 50-hPa surface using estimates of the global-scale air mass circulation and MLS measurements of ozone and derived a downward flux of 380 Tg in the northern extratropics and 290 Tg in the south. The largest downward fluxes of ozone are in the winter hemisphere between 60° and 70°N, and between 40° and 55°S, while the largest upward fluxes are in the subtropics in summer. They used a two-box model to derive flux of ozone across 100 hPa of 510 Tg/yr (450-590 Tg/yr) (with ~290 Tg in the Northern Hemisphere), similar to other estimates of the flux of ozone into the troposphere [e.g., Murphy *et al.*, 1993; Roelofs and Lelieveld, 1995].

The sonde data in Figure 11 were used to form an empirical average of the amount of ozone in the extratropical lowermost stratosphere, defined here as the tropopause to 100 hPa, and 30°N to the pole (Figure 13a). The amount of ozone decreases from about 175 Tg in March to about 75 Tg in October, or by 60%. About half of the decrease in ozone is caused by the change in height of the tropopause, which leads to a decrease in the air content of this region of 30% from March to September (Figure 13b). The fractional loss of air calculated from the tropopause heights for the sonde data is in good agreement with the more detailed calculation of Appenzeller *et al.* [1996]. The largest change in the amount of ozone from the tropopause to 100 hPa is between May and June (Figure 14). The integrated amount of ozone removed from the lowermost stratosphere between March and September (and accumulated from September to March) is ~100 Tg from 100 hPa to the tropopause, while it is ~64 Tg for the region from 380 K to the tropopause for the same period. The largest change in the amount of ozone occurs in April-June, the same months as the largest change in the amount of air. This region accumulates ozone at the highest rate in December-February, while it accumulates air at the highest rate in September-November [Appenzeller *et al.*, 1996]. The fate of the ozone that is removed from this region between March and September is uncertain. The amount removed chemically is likely small. Calculations of the residual circulation show that in spring the vertical velocities are downward across the 100-hPa surface poleward of ~40°N, upward for 30°-50°N in summer, and down-

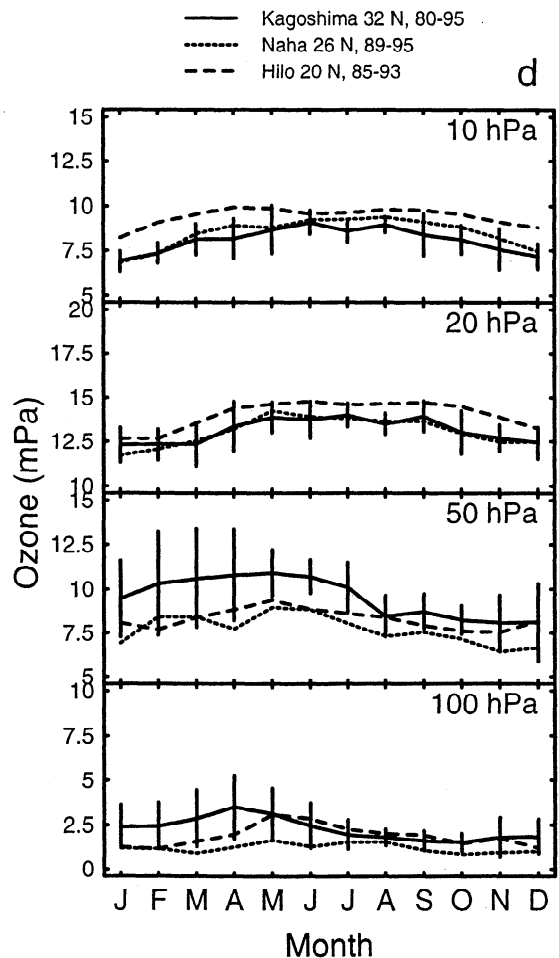


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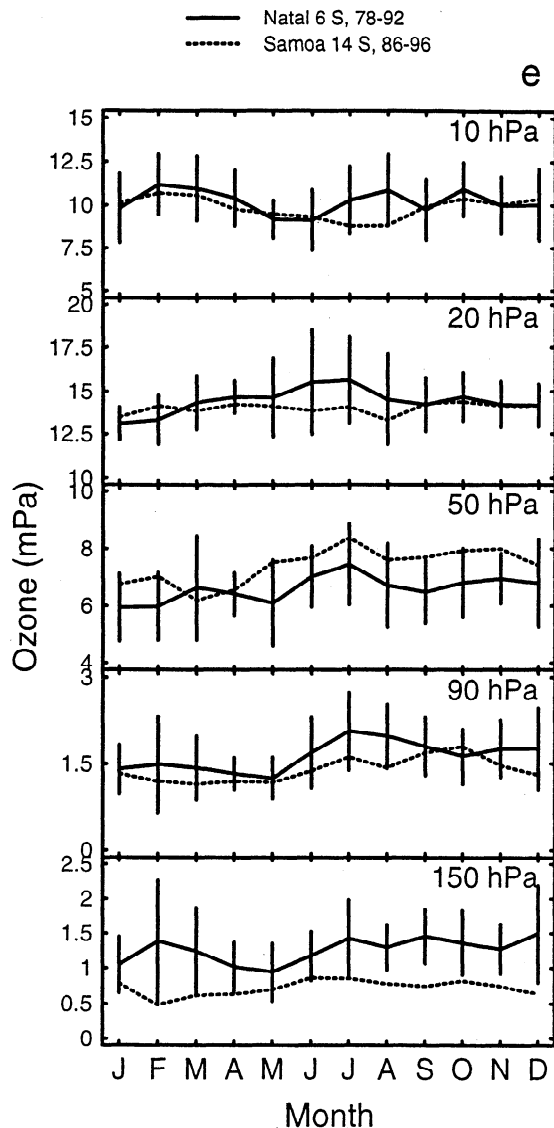


Figure 6. (continued)

ward further north [Eluszkiewicz *et al.*, 1996; Rosenlof, 1995]. Using the velocities given by Eluszkiewicz *et al.* [1996], I estimate that as much as one third of the ozone that leaves the region from 100 hPa to the tropopause between March and September may enter the overworld, while two thirds moves down into the troposphere. Yang and Tung [1996] show that all of the mass flux across the 385-K surface is downward north of 35°N year round.

In the Southern Hemisphere, ozone and temperature data are available for only one midlatitude station, Lauder (45°S). The change in the amount of ozone between the tropopause and 100 hPa from October to April is only 70% of the analogous change for the northern midlatitudes stations from March to September. For higher latitudes, there is no buildup of ozone in the lowermost stratosphere in southern winter, as evidenced by the pre-ozone hole data from Syowa, 67°S (Figure 5e).

6. Evaluation of Models With the Ozonesonde Data

The sonde data are sufficient to describe the abundance and annual cycle of ozone below 10 hPa for the extratropics of the

Northern Hemisphere, as shown in Figures 5 and 6. The unique information they provide, compared to remote-sensing techniques, is quantification of the buildup of ozone in the lowermost stratosphere in late autumn and winter and its dissipation in late spring and summer (Figure 11-14). The sonde data from the tropics and Southern Hemisphere are sparse but provide useful information on vertical structure and annual cycles. I recommend that data from the majority of stations given in Table 1 be used to test models of stratospheric dynamics and chemistry.

Given the trend in ozone in the lower stratosphere, the stations most suitable for testing models are (1) those with data from a common time period, 1980 to 1993, and (2) those with data from a similar period, if located in a region with small trends in column ozone, such as the tropics and subtropics [McPeters *et al.*, 1996]. The former category includes four Canadian stations from 53° to 75°N, four European stations from 48° to 53°N, and Wallops Island (38°N) in the U.S. The latter category includes three Japanese stations from 26° to 36°N, Hilo, Hawaii (20°N), Natal, Brazil (6°S), and Samoa (14°S). I recommend also the use of data from Boulder, Colorado (40°N), Aspendale/Laverton (45°S), and Lauder, New Zealand (45°S), and the three Antarctic stations which do not quite meet these criteria but which have

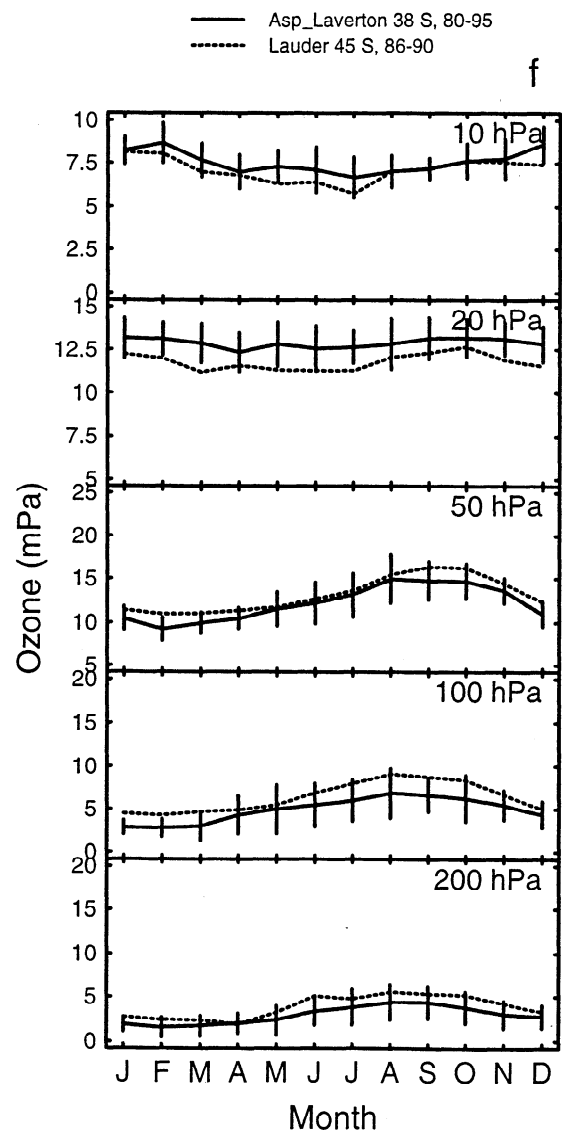


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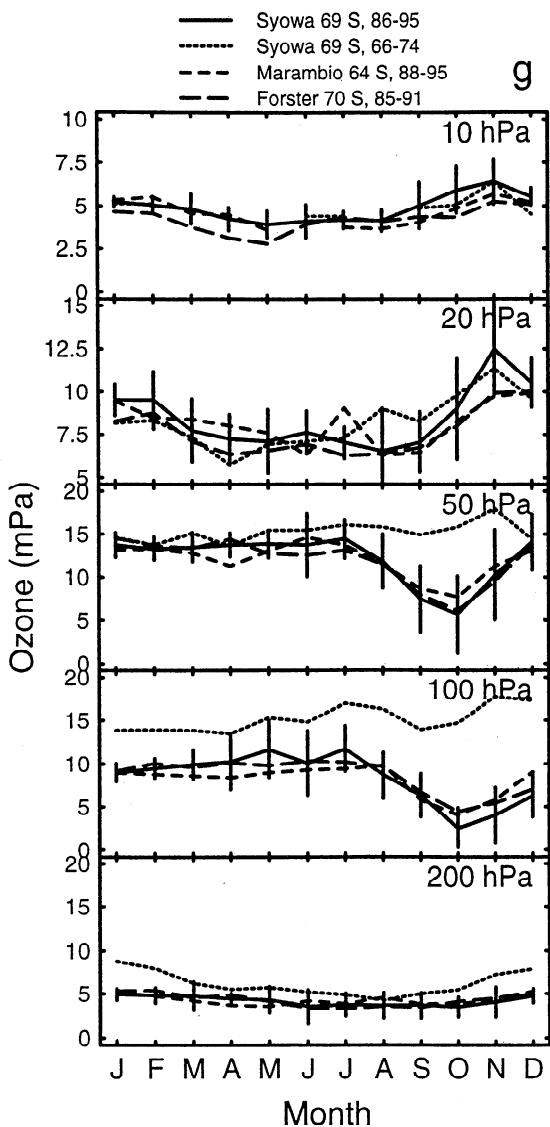


Figure 6. (continued)

measurements for a closely overlapping period. The monthly means are well defined for most of these stations, but I caution that the 95% confidence intervals can be as high as $\pm 25\%$ for tropical and subtropical stations below 70 hPa. The variance of the data is likely not well defined when there are less than 20 soundings per month, which is the case for some months at Kagoshima, Naha, Natal, Samoa, and Marambio.

The observed profiles for ozone should be compared with model simulations with chlorine and aerosol loading appropriate for the late 1980s. The inclusion of data for 1992 and 1993 has only a minor effect on the 14-year mean profiles for stratospheric ozone. Mean values for 1980-1991 are larger than those for 1980-1993 by $<5\%$ (usually 2-3%) for the European stations, Resolute, and Wallops, except for differences of 6-12% in January at Lindenberg. Differences are $<5\%$ for more than 90% of monthly means for the other Canadian stations and are nearly always $<10\%$.

In terms of evaluating 2-D models, the biases of the sonde stations with respect to zonal mean values of the ozone column (Figure 3) need to be taken into account, as well as the sampling and instrumental biases in column ozone measurements at the

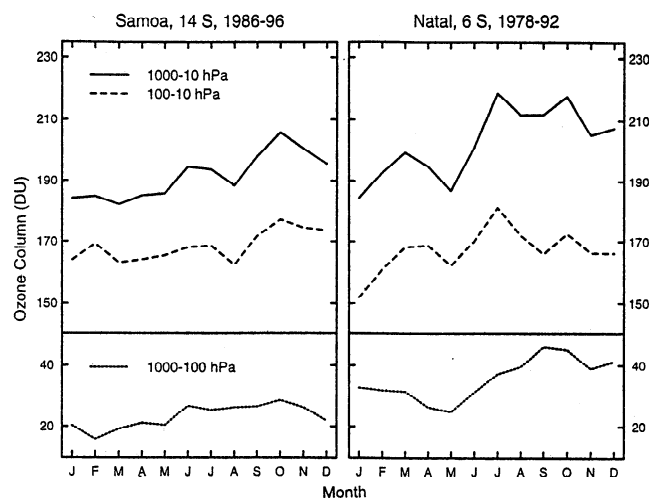


Figure 7. The annual cycle in ozone for (top) 1000 to 10 hPa, (top) 100-10 hPa, and (bottom) 1000-100 hPa for Natal and Samoa. The tropopause is close to 100 hPa, so these divisions separate the tropospheric and stratospheric contributions to the column below 10 mbar. The total column data were not provided with the sonde data.

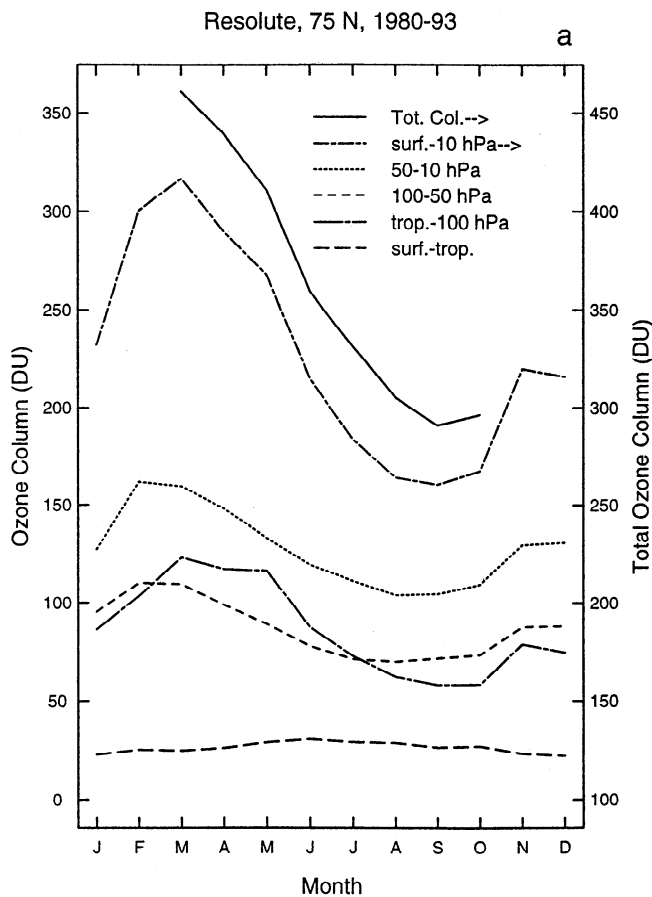


Figure 8. The annual cycle of the ozone column in four layers (left-hand scale) and of the total ozone column (right-hand scale). The layers are (1) the surface to the tropopause, (2) the tropopause to 100 hPa, (3), 100-50 hPa, and (4) 50-10 hPa. For Resolute, also given are the annual cycle from the ground to 10 hPa, as column data are lacking in winter. The ozone column data are those provided with the sonde profiles.

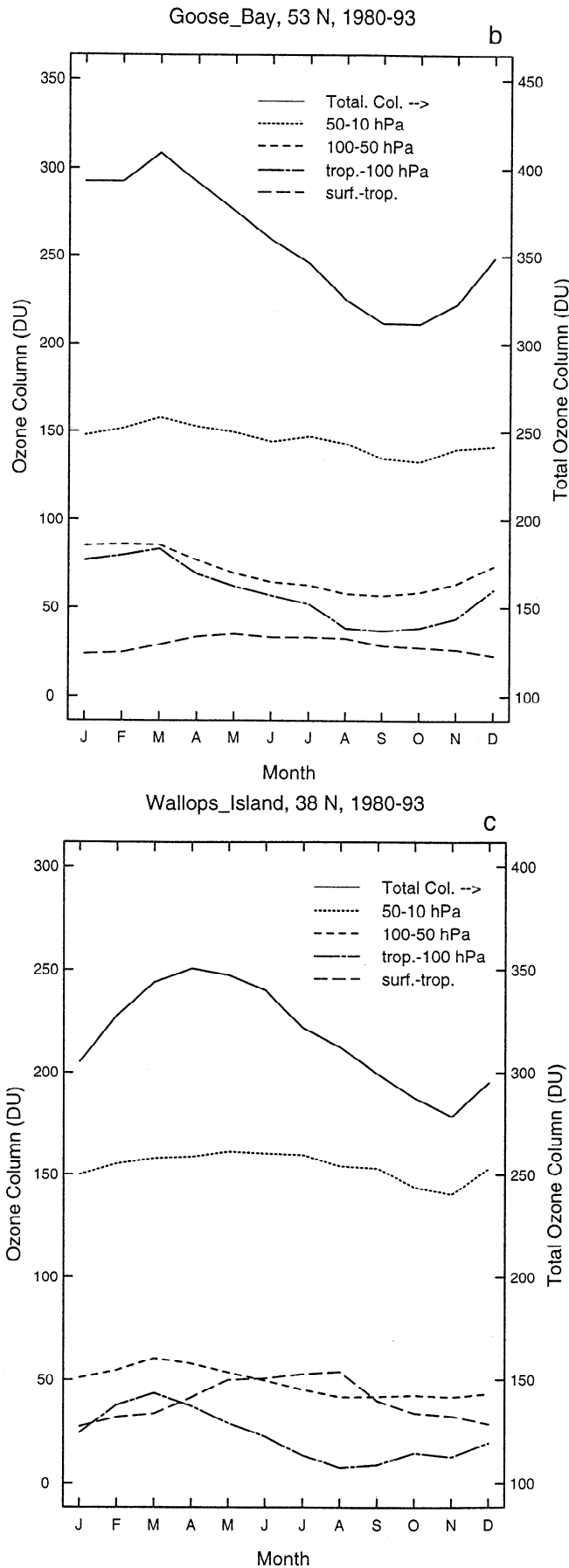


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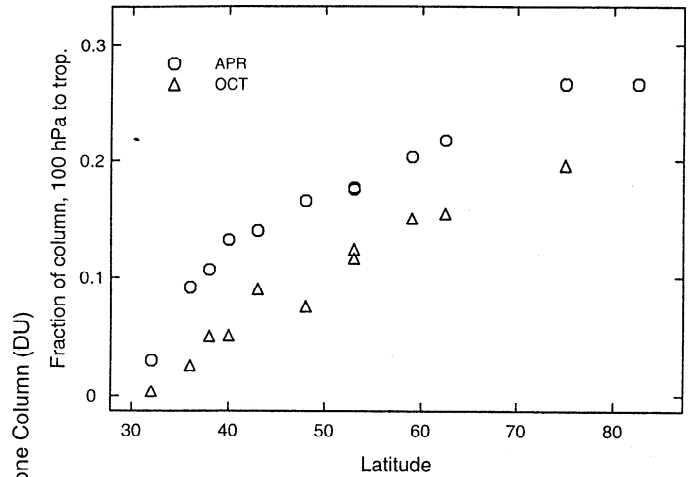


Figure 9. The fraction of the ozone column from the tropopause to 100 hPa versus latitude, calculated from the sonde data for the Northern Hemisphere. Results are shown for April and October, roughly the maximum and minimum of the ozone column.

stations (Figure 4). The combined effect of these biases (which are additive) is generally <10%. I do not recommend the use of Sapporo to test 2-D models because of the significant bias shown in Figure 3. For 3-D models, only the biases shown in Figure 4 need to be considered, and these are usually <5%. The bias in the sonde measurements themselves is less than $\pm 5\%$.

Earlier evaluations of 2-D models used profile information only above 16 km, derived from SBUV and SAGE II; Prather and Remsberg [1993, p. 22] stated "no-one has proposed how to usefully compare the high-frequency, single locale sondes with a zonally averaged model." As shown above, the region below ~16 km (100 hPa) contains 20-33% of the ozone column at middle to high latitudes, and any biases in the locations of the sonde stations with respect to zonal mean ozone are small compared to model-model differences in the lower stratosphere. The proposal here is to compare the monthly mean statistics for each station with the appropriate latitude band of the model; the information on the spatial bias of the stations with respect to the zonal mean ozone column, and on the tropopause height for the station, will

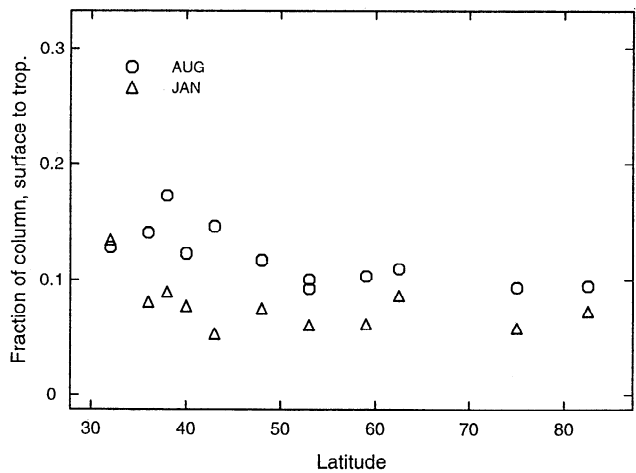


Figure 10. The fraction of the ozone column from the ground to the tropopause versus latitude, calculated from the sonde data for the Northern Hemisphere. Results are shown for January and August, roughly when the fraction is smallest and largest.

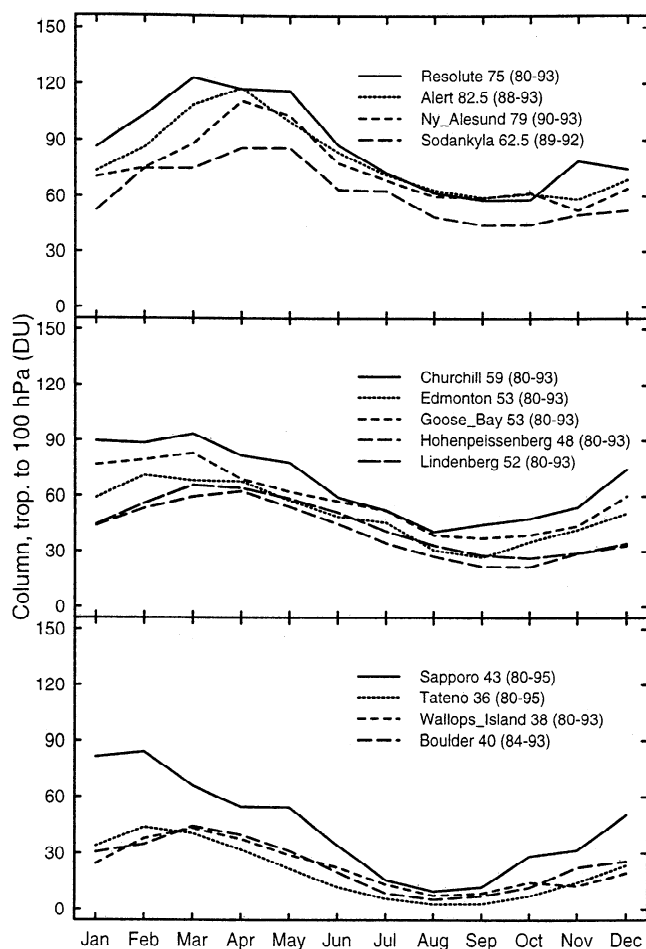


Figure 11. The annual cycle of the ozone content from the tropopause to 100 hPa, in Dobson units, for selected stations.

aid in interpretation of the results. Since the focus here is on the lower stratosphere, I recommend that vertical profiles be shown in units of either ozone partial pressure or number density. My experience in using the sonde data to evaluate 3-D tropospheric models indicates that it is useful to compare the annual cycle at different altitudes and the vertical profiles for different months to characterize the quality of the simulation [Friedl, 1997; Wang *et al.*, 1998]. The annual cycle of the ozone content of the lowermost stratosphere in the Northern Hemisphere, and its variation with latitude, is another useful test of stratospheric models. The sonde data should provide constraints on adjustable transport parameters in 2-D models in addition to those provided by dynamical tracers [e.g., Prather and Remsberg, 1993; Park *et al.*, 1999], and these constraints may lead to improvements in the formulation of model dynamics and, ultimately, to an improved predictive capability for ozone.

The data provided here allow evaluation also of the errors caused in the total ozone column by the representation of tropospheric ozone in stratospheric models. Although the troposphere is a small fraction of the column, if it is in error by a factor of 2, it could cause an error of as much as 5-10% in the column in northern midlatitude summer or in regions affected by biomass burning in the tropics. Analysis of SAGE II and MLS data show that much of the zonal asymmetry in the total ozone column in the southern tropics is not present in stratospheric ozone and is

caused by zonal asymmetries in tropospheric ozone [Shiotani and Hasabe, 1994; Ziemke *et al.*, 1996]; the annual cycles of the sonde data for Samoa and Natal (Figure 7) support this view. The contribution of tropospheric ozone should be taken into account when using TOMS data to evaluate tropical ozone in models.

The analysis presented here served as a foundation for development of a climatology that incorporates information from sonde data and from SAGE II. Comparison of sonde and SAGE II profiles for 1988-1996 shows good agreement for 20-30 km for most latitudes, and a preliminary stratospheric climatology was developed using these data for MM II [Logan and McPeters, 1999]. This climatology is being improved and will be reported elsewhere.

7. Summary

This paper summarizes current knowledge of the distribution of stratospheric ozone determined from ozonesondes, with an emphasis on the lowermost stratosphere, the region where the sondes provide unique information. Between 10 and 30% of the ozone column is located from 100 hPa to the tropopause poleward of 35°N, and this region drives 35-50% of the amplitude of the annual cycle in the ozone column. One of the major features of the ozone distribution at middle to high latitudes is the secondary maximum that builds up below 100 hPa in winter and early

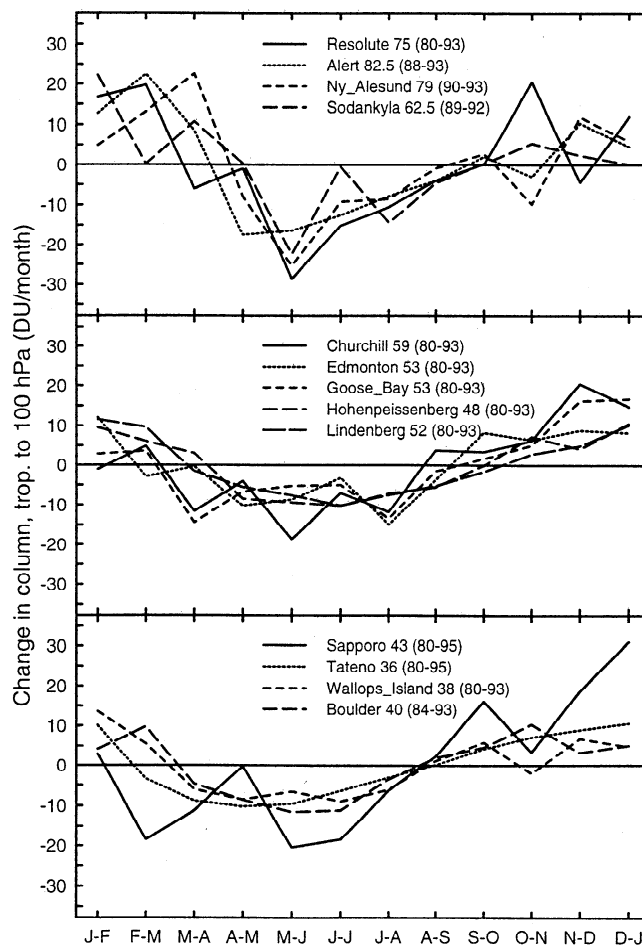


Figure 12. The rate of change of the ozone column from the tropopause to 100 hPa, in Dobson units per month.

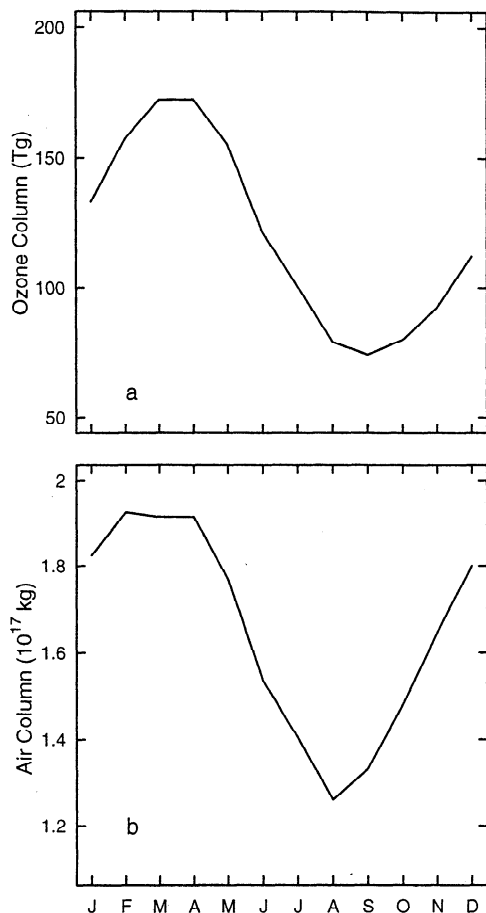


Figure 13. (a) The annual cycle of the amount of ozone in Tg from the tropopause to 100hPa, and from 30° to 90°N, calculated by weighting the results in Figure 11 by the area of each latitude band: 75°N, Resolute, Ny Alesund, and Alert; 60°N, Churchill and Sodankyla; 53°N, Edmonton, Goose Bay, and Lindenberg; 45°N, Hohenpeissenberg; 38°N, Wallops Island, Boulder, and Taten; 30°N, Kagoshima. (b) The annual cycle of the amount of air in the same region, calculated from the monthly mean tropopause heights for the sonde stations used for Figure 13a.

spring and recedes in late spring and summer. Ozone concentrations are highly variable in the region of the secondary maximum, with significant lamination in individual profiles. The frequency of occurrence of laminae given by Reid and Vaughan [1991] is similar to the annual cycle in the column of ozone in the lowermost stratosphere shown here.

The ozone content of the lowermost stratosphere decreases from 175 to 75 Tg from March to September, with about half this decrease caused by the increase in height of the tropopause; the maximum rate of decrease is in May-June. There is a lag of about a month in the time when ozone starts to accumulate in the lowermost stratosphere, from September near 40°N to October or November near 80°N, and a similar lag in the time when ozone starts to decrease, from February or March near 40°N to March or April near 80°N. On the basis of calculations of the residual circulation, it appears that most of the 100 Tg of ozone removed from the lowermost stratosphere enters the troposphere, with perhaps one third of it entering the overworld.

The sonde data suggest that the slight annual cycle in tropical stratospheric ozone, with a winter minimum, is likely caused by

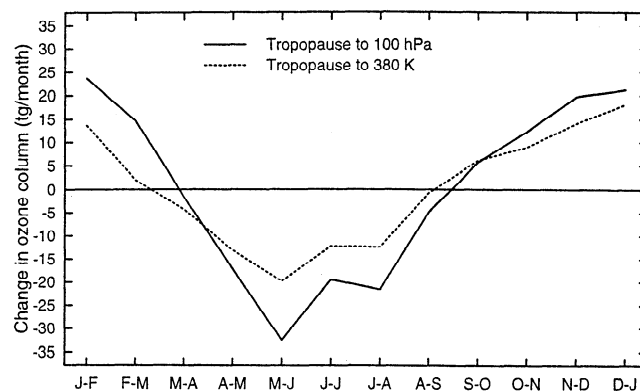


Figure 14. The rate of change of the ozone column from the tropopause to 100 hPa, for 30° to 90°N, in Tg ozone per month, for the results in Figure 13. Also given is the rate of change of the ozone column from the tropopause to the 380-K surface.

enhanced transport from tropics to midlatitudes in northern winter. The data from three stations on the periphery of the Antarctic continent show a consistent picture of polar ozone loss for the past decade.

The sonde data provide a test of transport in the lower stratosphere in addition to those provided by other long-lived tracers, such as N_2O , CFCs, CO_2 , CH_4 , SF_6 , and aerosols, and age of air, and recommendations are made for the use of particular stations to evaluate models. The biases in the location of sonde stations with respect to longitudinal variations of the ozone column are quantified using TOMS data and are small (<5%) compared to current model-model differences in the lower stratosphere. The annual variation of the ozone content of the lowermost stratosphere in the Northern Hemisphere is suggested as a particularly useful constraint on transport parameterizations in models.

Acknowledgments. I would like to thank Amy Munson for her able management and manipulation of the sonde data and H. Schneider for useful discussions about transport in 2-D models. Many thanks are due to Edward Hare, who has made WOUDC, the primary source of sonde data, a user friendly environment. S. Oltmans, J. Staehelin, A. Torres, and V. Kirchhoff kindly provided sonde data. Data were also obtained from the GTE archive at NASA Langley. This work was funded with support from the National Air and Space Administration, grants NAG5-2688, NAGW-2632, NAG1-1909, NAS1-19955, and NAS1-96022, and the National Science Foundation, grant ATM-9320778.

References

- Appenzeller, C., and J. R. Holton, Tracer lamination in the stratosphere: A global climatology, *J. Geophys. Res.*, *102*, 13,555-13,659, 1997.
- Appenzeller, C., J. R. Holton, and K. R. Rosenlof, Seasonal variation of mass transport across the tropopause, *J. Geophys. Res.*, *101*, 15,071-15,078, 1996.
- Attmanspacher, W., and H. U. Dutsch, International ozonesonde intercomparison at the Observatory Hohenpeissenberg, January 19 to February 5, 1970, *Ber. Dtsch. Wetterdienstes*, *16*(120), 1970.
- Attmanspacher, W., and H. U. Dutsch, Second international ozonesonde intercomparison at the Observatory Hohenpeissenberg, April 5-20 1978, *Ber. Dtsch. Wetterdienstes*, *157*, 1981.
- Attmanspacher, W., and R. Hartmannsgruber, Some results of 6 years (1967-72) of regular ozone soundings at the meteorological observatory Hohenpeissenberg, FRG, *Contrib. Atmos. Phys.*, *49*, 18-33, 1976.
- Bailey, P. L. et al., Comparison of cryogenic limb etalon spectrometer (CLAES) ozone observations with correlative measurements, *J. Geophys. Res.*, *101*, 9737-9756, 1996.
- Barnes, R.A., M. A. Chamberlain, C. L. Parsons, and A. F. Holland, An improved rocket ozonesonde (ROCOZ-A), *3*, Northern mid-latitude

- ozone measurements from 1983 to 1985, *J. Geophys. Res.*, *94*, 2239-2254, 1989.
- Beekmann, M., G. Ancellet, G. Megie, H. G. J. Smit, and D. Kley, Inter-comparison campaign including electrochemical sondes of ECC and Brewer-Mast type and a ground based UV-differential absorption lidar, *J. Atmos. Chem.*, *19*, 259-288, 1994.
- Bruhl, C., et al., Halogen occultation experiment ozone channel validation, *J. Geophys. Res.*, *101*, 10,217-10,240, 1996.
- Callis, L. B., M. Natarajan, J. D. Lambeth, and R. E. Boughner, On the origin of midlatitude ozone changes: Data analysis and simulations for 1979-1993, *J. Geophys. Res.*, *102*, 1215-1228, 1997.
- Chubachi, S., Annual variations of total ozone at Syowa station, Antarctica, *J. Geophys. Res.*, *102*, 1349-1354, 1997.
- Craig, R. A., *The Upper Atmosphere: Meteorology and Physics*, Academic, San Diego, Calif., 1965.
- Cunnold, D.M., L. Froidevaux, J. M. Russell, B. Conner, and A. Roche, Overview of UARS validation based primarily on intercomparisons among UARS and Stratospheric Aerosol and Gas Experiment II measurements, *J. Geophys. Res.*, *101*, 10,335-10,351, 1996.
- De Muer, D., Vertical ozone distribution over Uccle (Belgium) from six years of ozone soundings, *Contrib. Atmos. Phys.*, *49*, 1-17, 1977.
- Dobson, G. M. B., The laminated structure of the ozone in the atmosphere, *Q. J. R. Meteorol. Soc.*, *99*, 599-607, 1973.
- Dutsch, H. U., Two years of regular ozone soundings over Boulder, Colorado, *Tech. Note NCAR/TN-10*, Nat. Cent. for Atmos. Res., Boulder, Colo., 1966.
- Dutsch, H. U., The ozone distribution in the atmosphere, *Can. J. Chem.*, *52*, 1491-1504, 1974.
- Dutsch, H. U., Vertical distribution of ozone on a global scale, *Pure Appl. Geophys.*, *116*, 511-529, 1978.
- Dutsch, H. U., and C. Ling, Six years of regular ozone soundings over Switzerland, *Pure Appl. Geophys.*, *106-108*, 1151-1168, 1973.
- Dutsch, H. U., W. Zullig, and C. Ling, One and one half years of routine observations of vertical ozone distribution near Zurich, Switzerland, *Ann. Geophys.*, *25*, 249-260, 1969.
- Eluszkiewicz, J., D. Crisp, R. Zurek, L. Elson, L. Froidevaux, J. Waters, R.G. Grainger, A. Lambert, R. Harwood, and G. Peckham, Residual circulation in the stratosphere and lower mesosphere as diagnosed from Microwave Limb Sounder data, *J. Atmos. Sci.*, *53*, 217-240, 1996.
- Friedl, R. R., (Ed.), 1996 interim assessment of the atmospheric effects of subsonic aircraft, *NASA Ref. Publ. 1400*, 1997.
- Froidevaux, L., et al., Validation of UARS Microwave Limb Sounder ozone measurements, *J. Geophys. Res.*, *101*, 10,017-10,060, 1996.
- Garcia, R. R., and S. Solomon, A new numerical model of the middle atmosphere, 2. Ozone and related species, *J. Geophys. Res.*, *99*, 12,937-12,951, 1994.
- Gettelman, A., J. R. Holton, and K. H. Rosenlof, Mass fluxes of O₃, CH₄, N₂O, and CF₂Cl₂ in the lower stratosphere calculated from observational data, *J. Geophys. Res.*, *102*, 19,149-19,159, 1997.
- Gotz, F. W. P., A. R. Meetham, and G. M. B. Dobson, The vertical distribution of ozone in the atmosphere, *Proc. R. Soc. London Ser. A* *145*, 416-446, 1934.
- Hering, W. S., Ozone and transport processes, *Tellus*, *18*, 329-336, 1966.
- Hering, W. S., and T. R. Borden, Ozonesonde observations over North America, vol. 2, *Environ. Res. Pap.* *38*, Air Force Cambridge Res. Lab., Bedford, Mass., 1964.
- Hering, W. S., and T. R. Borden, Ozonesonde observations over North America, vol. 4, *Environ. Res. Pap.* *279*, Air Force Cambridge Res. Lab., Bedford, Mass., 1967.
- Hilsenrath, E., et al., Results from the Balloon Ozone Intercomparison Campaign (BOIC), *J. Geophys. Res.*, *91*, 13,137-13,152, 1986.
- Hofmann, D.J., S. J. Oltmans, W.D. Komhyr, J.M. Harris, J.A. Lathrop, A.O. Langford, T. Deshler, B.J. Johnson, A. Torres, and W.A. Matthews, Ozone loss in the lower stratosphere over the United States in 1992-1993: Evidence for heterogeneous chemistry on the Pinatubo aerosol, *Geophys. Res. Lett.*, *21*, 65-68, 1994.
- Holton, J. R., On the global exchange of mass between the stratosphere and troposphere, *J. Atmos. Sci.*, *47*, 392-395, 1990.
- Holton, J. R., P. H. Haynes, M. E. McIntyre, A. R. Douglas, R. B. Rood, and L. Pfister, Stratosphere-troposphere exchange, *Rev. Geophys.*, *33*, 403-439, 1995.
- Jackman, C. H., A. R. Douglas, P. D. Guthrie, and R. S. Stolarski, The sensitivity of total ozone and ozone perturbation scenarios in a two-dimensional model due to dynamical inputs, *J. Geophys. Res.*, *94*, 9873-9887, 1989.
- Jackman, C. H., E. L. Fleming, S. Chandra, D. B. Considine, and J. E. Rosenfield, Past, present, and future modeled ozone trends with comparisons to observed trends, *J. Geophys. Res.*, *101*, 28,753-28,767, 1996.
- Kerr, J. B., D. J. Wardle, and D. W. Tarasick, Record low ozone values over Canada in early 1993, *Geophys. Res. Lett.*, *20*, 1979-1982, 1993.
- Kirchhoff, V.W.J.H., R. A. Barnes, and A. L. Torres, Ozone climatology at Natal from in situ ozonesonde data, *J. Geophys. Res.*, *96*, 10,899-10,909, 1991.
- Komhyr, W. D., R. A. Barnes, G. B. Brothers, J. A. Lathrop, and D. P. Opperman, Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, *J. Geophys. Res.*, *100*, 9231-9244, 1995.
- Logan, J. A., Trends in the vertical distribution of ozone: An analysis of ozonesonde data, *J. Geophys. Res.*, *99*, 25,553-25,585, 1994.
- Logan, J. A., An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models, and development of a gridded climatology for tropospheric ozone, *J. Geophys. Res.*, this issue.
- Logan, J. A., and V.W.J.H. Kirchhoff, Seasonal variations of tropospheric ozone at Natal, Brazil, *J. Geophys. Res.*, *91*, 7875-7881, 1986.
- Logan, J. A., and R. D. McPeters, Ozone Climatology, Chapter 4 in Models and Measurements II, ed. J. H. Park, M. K. W. Ko, C. H. Jackman, and R. A. Plumb, *NASA/TM-1999-0000*, in press, 1999.
- Logan, J. A., et al., Trends in the vertical distribution of ozone: A comparison of two analyses of ozonesonde data, *J. Geophys. Res.*, in press, 1999.
- London, J., The observed distribution of atmospheric ozone and its variations, in *Ozone in the Free Atmosphere*, edited by R. C. Whitten and S. S. Prasad, pp. 11-80, Van Nostrand Reinhold, New York, 1985.
- London, J., and S. C. Liu, Long-term tropospheric and lower stratospheric ozone variations from ozonesonde observations, *J. Atmos. Terr. Phys.*, *54*, 599-625, 1992.
- McCormick, M. P., J. M. Zawodny, R. E. Veiga, J. C. Larsen, and P. H. Wang, An overview of SAGE I and II ozone measurements, *Planet. Space Sci.*, *37*, 1567-1586, 1989.
- McPeters, R. D., and E. Beach (Eds.), *TOMS Version 7 Ozone Gridded Data: 1978-1993* [CD-ROM] Ozone Process. Team, NASA Goddard Space Flight Cent., Greenbelt, Md., 1996.
- McPeters, R. D., and G. J. Labow, An assessment of the accuracy of 14.5 years of Nimbus 7 TOMS Version 7 ozone data by comparison with the Dobson network, *Geophys. Res. Lett.*, *23*, 3695-3698, 1996.
- McPeters, R. D., D. F. Heath, and P. K. Bhartia, Average ozone profiles from the NIMBUS 7 SBUV instrument, *J. Geophys. Res.*, *89*, 5199-5214, 1984.
- McPeters, R. D., T. Miles, L. E. Flynn, C. G. Wellemeyer, and J. M. Zawodny, Comparison of SBUV and SAGE II ozone profiles: Implications for ozone trends, *J. Geophys. Res.*, *99*, 20,513-20,524, 1994.
- McPeters, R. D., S. M. Hollandsworth, L.E. Flynn, J.R. Herman, and C. J. Seftor, Long term ozone trends derived from the 16-year combined Nimbus 7/Meteor 3 TOMS Version 7 record, *Geophys. Res. Lett.*, *23*, 3699-3702, 1996.
- Murphy, D. M., D. W. Fahey, M. H. Proffitt, S. C. Liu, K. R. Chan, C. S. Eubank, S. R. Kawa, and K. K. Kelly, Reactive nitrogen and its correlation with ozone in the lower stratosphere and upper troposphere, *J. Geophys. Res.*, *98*, 8751-8771, 1993.
- Newman, P.A., et al., Measurements of polar vortex air in the midlatitudes, *J. Geophys. Res.*, *101*, 12,879-12,891, 1996.
- Oltmans, S.J., D.J. Hofmann, W.D. Komhyr, and J.A. Lathrop, Ozone vertical profile changes over South Pole, in *Ozone in the Troposphere and Stratosphere*, edited by R. D. Hudson, *NASA Conf. Publ.*, *3266*, 578-581, 1994.
- Oltmans, S. J. et al., Trends of ozone in the troposphere, *Geophys. Res. Lett.*, *25*, 139-142, 1998.
- Park, J. H., M. K. W. Ko, C. H. Jackman, and R. A. Plumb, Model and measurements II, *NASA/TM-1999-0000*, in press, 1999.
- Perliski, L. M., and J. London, Satellite observed long-term averaged aseasonal and spatial ozone variations in the stratosphere, *Planet. Space Sci.*, *37*, 1527-1538, 1989.
- Pittock, A.B., Climatology of the vertical distribution of ozone over Aspendale, *Q. J. R. Meteorol. Soc.*, *103*, 575-584, 1977.
- Plumb, R. A., and M. K. W. Ko, Interrelationship between mixing ratios of long-lived stratospheric constituents, *J. Geophys. Res.*, *97*, 10,145-10,156, 1992.
- Prather, M. J., and E. E. Remsberg, The atmospheric effects of aircraft:

- Report of the 1992 models and measurements workshop, *NASA Ref. Publ.*, 1292, 1993.
- Randall, C. E., et al., Preliminary results from POAM II: Stratospheric ozone at high northern latitudes, *Geophys. Res. Lett.*, 22, 2733-2736, 1995.
- Rasch, P. J., B. A. Boville, and G. P. Brasseur, A coupled three-dimensional general circulation model with coupled chemistry for the middle atmosphere, *J. Geophys. Res.*, 100, 9041-9071, 1995.
- Reid, S. J., and G. Vaughan, Lamination in ozone profiles in the lower stratosphere, *Q. J. R. Meteorol. Soc.*, 117, 825-844, 1991.
- Reid, S. J., G. Vaughan, and E. Kyrö, Occurrence of ozone laminae near the boundary of the stratospheric polar front, *J. Geophys. Res.*, 98, 8883-8890, 1993.
- Roelofs, G. J., and J. Lelieveld, Distribution and budget of O₃ in the troposphere calculated with a chemistry general circulation model, *J. Geophys. Res.*, 100, 20,983-20,998, 1995.
- Rood, R. B., A. R. Douglas, J. A. Kaye, M. A. Geller, C. Yuechen, D. J. Allen, E. M. Larsen, E. R. Nash, and J. E. Nielson, Three-dimensional simulations of wintertime ozone variability in the lower stratosphere, *J. Geophys. Res.*, 96, 5055-5072, 1991.
- Rosenlof, K. H. Seasonal cycle of the residual mean meridional circulation in the stratosphere, *J. Geophys. Res.*, 100, 5173-5191, 1995.
- Rosenlof, K. H., and J. R. Holton, Estimates of the stratospheric residual circulation using the downward control principle, *J. Geophys. Res.*, 98, 10,456-10,479, 1993.
- Shindell, D. T., D. Rind, and P. Lonergan, Increased polar stratospheric ozone losses and delayed eventual recovery owing to increasing greenhouse gas concentrations, *Nature*, 392, 589-592, 1998.
- Shiotani, M., and F. Hasabe, Stratospheric ozone variations in the equatorial region as seen in Stratospheric Aerosol and Gas Experiment data, *J. Geophys. Res.*, 99, 14,575-14,584, 1994.
- Solomon, S. R. W. Portmann, R. R. Garcia, L. W. Thomason, L. R. Poole, and M. P. McCormick, The role of aerosol variations in anthropogenic ozone depletion at northern midlatitudes, *J. Geophys. Res.*, 101, 6713-6728, 1996.
- Solomon, S., et al., Ozone depletion at mid-latitudes: Coupling of volcanic aerosol and temperature variability in to anthropogenic chlorine, *Geophys. Res. Lett.*, 25, 1871-1874, 1998.
- Stolarski, R. S., et al., 1995 Scientific assessment of the atmospheric effects of stratospheric aircraft, *NASA Ref. Publ.*, 1381, 1995.
- Walshaw, C. D., G. M. B. Dobson - The man and his work, *Planet. Space Sci.*, 37, 1485-1507, 1989.
- Wang, Y. H., J. A. Logan, and D. J. Jacob, Global simulation of tropospheric O₃-NO_x-hydrocarbon chemistry, 2, Model evaluation, *J. Geophys. Res.*, 103, 10,727-10,756, 1998.
- World Meteorological Organization (WMO), Atmospheric ozone, 1985: Assessment of our understanding of the processes controlling its present distribution and change, *WMO, Rep. 16*, Geneva, 1986.
- World Meteorological Organization, (WMO), Scientific assessment of stratospheric ozone: 1989, Global Ozone Res. Monit. Proj., *WMO, Rep. 20*, Geneva, 1990.
- World Meteorological Organization, (WMO), Scientific assessment of ozone depletion: 1991, Global Ozone Res. Monit. Proj., *WMO, Rep. 25*, Geneva, 1992.
- World Meteorological Organization, (WMO), Scientific assessment of ozone depletion: 1994, Global Ozone Res. Monit. Proj., *WMO, Rep. 37*, Geneva, 1995.
- World Meteorological Organization, (WMO), Scientific assessment of ozone depletion: 1998, Global Ozone Res. Monit. Proj., *WMO, Rep. 44*, Geneva, 1999.
- World Meteorological Organization, (WMO), Assessment of trends in the vertical distribution of ozone, SPARC Report no. 1, Global Ozone Res. Monit. Proj., *WMO, Rep. 43*, Genève, 1998.
- Yang, H., and K. K. Tung, Cross isentropic stratosphere-troposphere exchange of mass and water vapor, *J. Geophys. Res.*, 101, 9413-9424, 1996.
- Ziemke, J. R., S. Chandra, A. M. Thompson, and D. P. McManara, Zonal asymmetries in southern hemisphere column ozone: Implications of biomass burning, *J. Geophys. Res.*, 101, 14,421-14,427, 1996.

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